Hydraulic Conductivity of Compacted Lime-Softening Sludge Used as Landfill Liners

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Received: 14 May 2019 / Accepted: 12 September 2019 / Published online: 23 November 2019
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Abstract The research goal was to investigate the hydraulic conductivity of compacted lime-softening sludge as a material to be applied to landfill liners. In doing so, the effect of compaction and moulding moisture content on the sludge hydraulic conductivity was assessed. An approximate polynomial $k_{10\text{mean}}$ at hydraulic gradients ≥ 30 for degree of compaction (0.95–1.05) and moulding moisture content (28%–36%) was determined. The results of short-term tap water permeation tests revealed that all hydraulic conductivity values were less than $2.5 \cdot 10^{-8}$ m/s. A lowest hydraulic conductivity of $6.5 \cdot 10^{-9}$ m/s, as well as a corresponding moisture content of 31% were then established. The long-term hydraulic conductivity was measured with tap water, distilled water, NaOH and HCl solutions and municipal waste leachate. The factors of permeating liquids and permeation time significantly affected the initial hydraulic conductivity. The long-term hydraulic conductivity increased for NaOH and HCl solutions and decreased for tap and distilled water. A significant reduction of hydraulic conductivity was observed for leachate permeation. The investigated material met the requirements for the liner systems of inert landfill sites regardless of pH and the limit value for hazardous and non-hazardous waste landfills.

Keywords Lime-softening sludge · Landfill liner · Hydraulic · Conductivity · Leachate · Permeability · Compaction

1 Introduction

1.1 Background

Safe utilization of billions of tons of municipal and industrial wastes is one of today's major problems. Wastes are partially recycled and thermally neutralized, but waste storage in landfill sites is still the common way of its disposal. The landfill sites should guarantee safe waste disposal storage at minimum hazard for the environment. Hence, a landfill site should be equipped with a low permeability liner if the natural soil is not impermeable enough to protect the surrounding environment (the soils, ground water and surface water) from being contaminated by waste leachate generated within the landfill. Properly designed and built landfill liners must achieve consistent performance and be compatible with the expected leachate for the design life of the facility.

Still, tighten requirements concerning natural environmental protection have brought about the need for more attention to be drawn to proper selection of liner system, depending on waste landfills and soils with optimal characteristics and the appropriate criteria of their use in applied liner systems. There are 2 kind of liner systems recommended and predominantly used for hazardous waste landfill: a single composite liner

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and a double composite liner. The single composite liner consists of a drainage layer with a flexible membrane liner underneath, which overlay a compacted mineral layer. The double composite liner system has two single composite liners on top of each other with a leachate detection system between each layer. The liner systems should be applied on mineral subsoil formed by compaction. The single composite liner is applied to non-hazardous and inert landfills as well. The drainage layer for leachate collection and artificial sealing liner are not always required for inert landfills. The drainage layer should be minimum 0.5 m thick and have a permeability not less than or equal to $1.0 \cdot 10^{-4}$ m/s. The flexible membrane liner ought to be minimum 2 mm thick. The European Union regulations for waste landfills require any material used as a mineral liner for hazardous and non-hazardous waste landfills to have a permeability less than or equal to $1.0 \cdot 10^{-9}$ m/s or for inert landfills, less than or equal to $1.0 \cdot 10^{-7}$ m/s. A minimum thickness of a mineral layer, or a total thickness of mineral layers in case of double composite liner, is required not to be thicker than 1 m for non-hazardous waste and inert waste or at least 5 m for hazardous waste (Council Directive 1999). The requirements, which are obligatory in Poland, are compatible with European Union regulations (Dz. U. 2013 poz. 523; Dz. U. 2015 poz. 796).

The criterion of hydraulic conductivity is considered to be of most importance for the construction of landfill site liners. Hence, the cohesive soils, mainly clays and clayey soils are applied in mineral layers. The hydraulic conductivity of material used in landfill liner is required to be tested by at least 2 methods: laboratory and field. Herein, the hydraulic conductivity measured via field tests might be even 2 orders of magnitude higher than that estimated in laboratory tests (Benson et al. 1999).

The hydraulic conductivity depends on the compaction, the dry density and the moisture content so their impact on the permeability cannot be neglected. The common criterion of soil layer is that the compaction in range of moisture content between optimum water content w_{opt} to $w_{opt} + 4\%$ guarantees the minimum value of soil dry density as indicated via the compaction Proctor test: standard method ($\rho_d \geq 0.95 \cdot \rho_{ds}$, i.e. degree of compaction $I_s = 0.95$) and modified method ($\rho_d \geq 0.90 \cdot \rho_{ds}$, i.e. degree of compaction $I_s = 0.95$). The criterion has been found to be not sufficient enough for achieving the required hydraulic conductivity. Thus, other parameters that affect the hydraulic conductivity should also be taken into account. Among these are: particle size distribution, plasticity index, liquid limit,

swelling behaviour, shear strength, compressibility, colloidal activity and initial degree of saturation (Yesiller et al. 2000; Council Directive 1999; Benson et al. 1994; Daniel and Wu 1993; Shelley and Daniel 1993).

It is common that available on-site soils do not have optimal characteristics and they are not suitable to be used for liner systems construction. Hence, the modification methods of local natural soils, which lead to the reduction of permeability, as well as alternative materials, which provide good contaminant sorption properties that meet the requirements for the liner systems, are searched. Then, wide range of wastes and by-products, as well as their mixtures are also studied. What is more, it is beneficial to use them as alternative materials as this contributes to natural resources protection and reduction of the total amount of stored waste in general.

A review of the existing literature revealed that most researches have focused on studying the engineering properties of soils amended with additives, which were added in the right proportions affecting the hydraulic conductivity and sorption of tested soils. The researches indicated that addition of shale (Li et al. 2016), lime (Tsai and Vesilind 1998) as well as and sewage sludge ash (Zhang et al. 2018) have a significant effect on soil permeability properties. Both cohesive as well as noncohesive soils, amended with bentonite are commonly used for constructing landfill barriers (Kumar and Yong 2002; Ojuri and Oluwatuyi 2017). Moreover, soils compacted with addition of lime and bentonite (Sivapullaiah et al. 2003; Firoozfar and Khosroshiri 2017) as well as fly ash and bentonite (Tiwari et al. 2000) also allow to achieve permeability low enough to apply at landfill liner. The reports on attempts made to reduce the permeability of soils by using granite polish wastes showed that they are only suitable for landfill liner application when adding bentonite (Patil et al. 2009). While searching for alternative materials for landfill liner application, much attention was given to research of fly ash, as it is one of the largest industrial solid wastes. Laboratory tests revealed that the required hydraulic conductivity could be achieved for fly ashes, but it is difficult (Palmer et al. 2000; Sivapullaiah and Lakshmikantha 2004). However, fly ash mixed with bentonite (Pandey and Jain 2017; Anul and Iqbal 2018) as well as fly ash improved by sewage sludge (Herrmann et al. 2009) can be used as an effective substitute for the traditional clay liner. Furthermore, rubber and bentonite added to fly ash met the hydraulic conductivity limit values (Cokca and Yilmaz 2004).

Addition of gypsum along with lime occurred effective in reducing the hydraulic conductivity of fly ash used in landfill liner systems (Sivapullaiah and Baig 2011). The feasibility of using a mudstone material as well as a coal gangue as landfill material was studied. Tests results indicated that those materials could be applied to landfill soil liners (Sheu et al. 1998; Wu et al. 2017).

1.2 Lime-Softening Sludge Composition

Raw water must be conditioned before industrial or municipal usage because of the high carbonate hardness induced by compounds soluble in water, i.e.: CaCO_3 , $\text{Ca}(\text{HCO}_3)_2$, $\text{Ca}(\text{OH})_2$, MgCO_3 , $\text{Mg}(\text{HCO}_3)_2$, $\text{Mg}(\text{OH})_2$. The aim of the decarbonising process is to reduce water carbonate hardness down to a standard that is acceptable to a given technological process. As a result, soluble compounds are converted into insoluble compounds that can be removed by sedimentation. The lime-softening process is the most common method used at water-treatment plants to lessen water carbonate hardness. This comes about by adding slaked lime $\text{Ca}(\text{OH})_2$. The fundamental chemical reactions are as follows (Stańda 1995):

If there are cations of magnesium and iron in the water and the $\text{Ca}(\text{OH})_2$ is in excess, additional reactions might occur at $\text{pH} > 10.5$, wherein, $\text{Mg}(\text{OH})_2$ and $\text{Fe}(\text{OH})_3$ are induced to precipitate.

It is common for lime sludge obtained from this process to consist of fine-grained CaCO_3 . Precipitation of calcium carbonate is in the form of calcite, or rarely, in that of unstable forms of aragonite and vaterite. In contrast, $\text{Mg}(\text{OH})_2$ precipitates only as a gelatinous sludge. Suspended solids, other iron and magnesium compounds and free carbon dioxide are also removed during the softening process. The precipitated sludge has the ability to absorb oil, soluble compounds and even organic chemicals (Ayoub and Merhebi 2000).

1.3 Lime-Softening Sludge Reuse

The problem of lime-softening sludge utilization was first noticed in running the water-treatment plant in

Oberlin, Ohio State, USA, in 1903 (Che et al. 1988). River, lake and retention ponds and lagoon storage/disposal were at that time the common and generally accepted ways of lime sludge disposal in the USA, as well as in Poland (Sozański 1999).

In Poland, Chojnacki (1966) was the first to address the problem of use and disposal of lime-softening sludge. He also paid particular attention to problems subsequently derived from sludge utilization: limitations of its discharge into rivers and lakes and difficulties with sludge dewatering in lagoons because of high moisture content. The search for new methods of sludge disposal was launched in the 60s and 70s of the XX-ith century upon the tightening of rules concerning natural environment protection. Thus, lime-softening sludge began to be reused as a soil amendment, and was experimented with for engineering construction purposes.

Studies of the geotechnical properties of lime-softening sludge were initiated by Glysson (1972), who discussed the possibilities of lime-softening sludge reuse in geotechnical engineering as a material for building liner systems for waste landfill sites. Furthermore, Glysson (1972) and Raghu and Hsieh (1985) suggested using the lime-softening sludge stored in lagoons for ground levelling and for combination with other wastes for stabilization. Tests carried out by Leeuwen et al. (2011a) showed that the lime-sludge could be successfully reused in wastewater neutralization. Leeuwen et al. (2011b, c) also demonstrated the potential use of dried lime-sludge, modified with stabilizers or mixed with soil, Portland cement or Class C fly ash as fill material for road construction.

Ippolito et al. (2011) and Olivier et al. (2011) reported that lime-softening sludge could be recycled through land application and offer a number of potential benefits for its use as clean below-water table fill, including a reduction in the nutrient loading. In addition, the synthetic precipitation leaching procedure testing conducted by Cheng et al. (2014) showed that lime-softening sludge could be of beneficial use in land applications above the groundwater table as it does not hold potential for release of trace elements into groundwater. However, on the basis on their researches, they emphasized that careful consideration and evaluation of management and disposal options of lime-softening sludge should be conducted, and that any assessment should contain a site-specific evaluation of reuse scenario (Blaisi et al. 2015).

A commonly recommended way of lime-softening sludge disposal is in agricultural reuse as an amendment (Glysson 1972; Raghu and Hsieh 1985; Che et al. 1988; Sozański 1999; Ratnayaka et al. 2009). Moreover, lime-softening sludge could be used in cement production as a limestone additive or instead of limestone (Harris 2002; Leeuwen et al. 2011a) and potentially for SO_x removal in coal power plants and coal power station flue gas (Leeuwen et al. 2011b; Ratnayaka et al. 2009). Ratnayaka et al. (2009) also recommended the use of lime-softening sludge in industry for products such as cosmetics.

The production of granulates from lime-softening sludge was begun in Holland in 1989. The granulate is used as a construction materials additive, as an additive in chicken feed, for neutralization pickling solution in electroplating, for sulphur removal from gas in coal power plant installations, for groundwater alkalisation, for mortar production and in high-ways construction as an asphalt mass additive. According to Sozański (1999), lime-softening sludge granulate might also be reused in steel production, in fertilizer production, in sewage sludge stabilization, in sewage dephosphatation and in calcium carbonate recalcination. About 100,000 tones of lime-softening sludge is produced in Poland every year, but only an insignificant part is recycled and neutralized. Lime-softening sludge is not a well-recognized material with regard to its geotechnical properties. There is a shortage of available data in a technical literature of international scope, which concerns detailed investigation of the sludge properties affecting its reuse in landfill liners.

The main purpose of this work was to investigate the hydraulic conductivity, affected by the compaction and the moulding moisture content, in the short-term permeation tests and evaluate the simultaneous effect of time factor and the permeating liquid on the hydraulic conductivity, in the long-term permeation tests, for the lime-softening sludge as a material applied to landfill liners.

2 Materials and Methods

2.1 Lime-Softening Sludge

For the purposes of this study, the lime-softening sludge produced in treating the water of the Rzeszów Power Plant was tested. The original source of water was the Wisłok River. A solution of lime, in the form of lime milk with concentration about 2%, was applied to utilize it for cooling purposes. The coagulation process was carried out simultaneously. Here, a ferric sulphate FeSO₄·7H₂O was used as the coagulant. The derived sludge was directed to a filter press and removed into a container underneath after dewatering. About 100–230 tons of sludge is produced in the Rzeszów Power Plant every year, and new possibilities of sludge disposal are being sought at present. Figure 1 shows the stockpiled lime-softening sludge in the Rzeszów Power Plant wet waste storage.

The main compound of the tested lime-softening sludge was calcium carbonate CaCO₃ (up to 87.25%), which appeared as calcite. The sludge also contained small amounts of iron, magnesium and aluminium

Fig. 1 The stockpiled lime-softening sludge

Table 1 The geotechnical properties of lime-softening sludge (Dąbska 2007)

No.	Property	Value range	Remarks
1	Bulk density ρ (Mg/m ³)	1.61–1.79	After filtering on filter press
2	Moisture content w (%)	46.1–55.1	
3	Specific density of solids ρ_s (Mg/m ³)	2.66–2.81	Proctor standard method
4	Plastic limit w_p (%)	34.1–40.9	
5	Liquid limit w_L (%)	58.4–65.3	
6	Shrinkage limit w_s (%)	34.0–37.3	
7	Plasticity index I_p , %	17.5–31.2	
8	Maximum dry density ρ_{ds} (Mg/m ³)	1.28–1.40	
9	Optimum water content w_{opt} (%)	28.9–37.7	

compounds and vestigial amounts of such elements as manganese, potassium and sodium. The sludge was characterized by pH values between 8.35 and 8.66. The lime-softening sludge also showed a particle-size distribution that is similar to natural clayey silts, silt-clays and silts, with clay content less than 23.7%, silt content between 63.0% to 93.7% and sand content less than 22.0% (Dąbska 2007). The geotechnical properties of lime-softening sludge are found in Table 1.

2.2 Permeating Liquids

The permeating liquids used for the permeability tests were: tap water (WW), distilled water (WD), NaOH solution at $\text{pH} \geq 11.0$ (NaOH), HCl solution at $\text{pH} \leq$

3.0 (HCl) and municipal landfill leachate from an operating landfill site in Otwock (O1 and O2). Table 2 and Table 3 summarize the chemical properties of the permeating liquids.

2.3 Methodology of Hydraulic Conductivity Tests

The hydraulic conductivity tests were performed by way of the falling head technique, following the ISO/TS 17892–11 (2004) standard procedure. The tests were carried out on cylindrical specimens 0.06 m height, with diameter 0.1 m, prepared in rigid-wall cylinders with porous filters placed bottom and top. The specimens were moulded directly in the cylinders while these were held within the hydraulic press.

Table 2 The chemical properties of permeating liquids

No.	Permeating liquid	Permeating time day	Chemical property						
			pH	Electric conduction $\mu\text{S}/\text{cm}$	Ca^{2+} $\text{mgCa}^{2+}/\text{dm}^3$	Mg^{2+} $\text{mgMg}^{2+}/\text{dm}^3$	Na^+ $\text{mgNa}^+/\text{dm}^3$	Fe^{3+} $\text{mgFe}^{3+}/\text{dm}^3$	Cl^- $\text{mgCl}^-/\text{dm}^3$
1	Distilled water	1–26	8.38	108	0	0	9.9	0	23.3
2		26–91	7.80	48.3	0	0	10.6	0	21.3
3	Tap water	1–35	7.49	1,137	63.2	20.1	130	0.048	241.4
4		35–91	7.70	1,513	59.0	17.4	125	0.057	259.0
5	0.2% NaOH	1–26	11.7	11,600	0	0	1,240	0	20.3
6		26–72	–	–	–	–	–	–	–
7	0.43% HCl	72–91	11.8	10,011	0	0	1,265	0	20.5
8		1–17	2.46	2,714	1.0	0.2	6.5	–	355
9		17–31	2.63	2,772	1.0	0.1	6.1	–	355
10		31–66	–	–	–	–	–	–	–
11		66–87	2.38	2,733	0.7	0.2	6.4	–	234

Table 3 The chemical properties of leachate

No.	Parameter	Unit	Value
1	pH	—	8.35
2	Electric conduction	$\mu\text{S}/\text{cm}$	12,615
3	ChZT	mgO_2/dm^3	3,744
4	Ca^{2+}	$\text{mgCa}^{2+}/\text{dm}^3$	42.9
5	Mg^{2+}	$\text{mgMg}^{2+}/\text{dm}^3$	20
6	Na^+	$\text{mgNa}^+/\text{dm}^3$	240
7	K^+	mgK^+/dm^3	145
8	Fe^{3+}	$\text{mgFe}^{2+}/\text{dm}^3$	8.24
9	Mn^{2+}	$\text{mgMn}^{2+}/\text{dm}^3$	5.4
10	Cl^-	$\text{mgCl}^-/\text{dm}^3$	2,201
11	SO_4^{2-}	$\text{mgSO}_4^{2-}/\text{dm}^3$	— ^a
12	PO_4^{3-}	$\text{mgPO}_4^{3-}/\text{dm}^3$	150
13	NH_4^+	$\text{mgNH}_4^+/\text{dm}^3$	1,226
14	NO_3^-	$\text{mgNO}_3^-/\text{dm}^3$	—
15	N og	mgN/dm^3	1,305
16	N org	mgN/dm^3	121
17	Pb^{2+}	$\text{mgPb}^{2+}/\text{dm}^3$	0.504
18	Zn^{2+}	$\text{mgZn}^{2+}/\text{dm}^3$	0.424
19	Cu^{2+}	$\text{mgCu}^{2+}/\text{dm}^3$	624
20	$\text{Cr}^{2+}, \text{Cr}^{7+}$	$\text{mgCr}^{(7+)+(2+)}/\text{dm}^3$	0.392
21	Ni^{2+}	$\text{mgNi}^{2+}/\text{dm}^3$	0.932
22	Cd^{2+}	$\text{mgCd}^{2+}/\text{dm}^3$	0.088

^a Measurement was not possible

The hydraulic conductivity tests of compacted lime-softening sludge were carried out in two stages of permeation: short-term and long-term.

The short-term tests were performed in order to recognize the permeating properties of lime-softening sludge and establish the hydraulic conductivity dependence on the compaction and the moulding moisture content in relation to the compaction curve obtained from the Proctor test. Tap water was used as a permeating liquid. In comparing the physical parameters of lime-softening sludge to that of cohesive soils, it was assumed that the lowest hydraulic conductivity occurred at proper compaction at moisture content $w \geq w_{\text{opt}}$. Thus, sludge as compacted using the standard Proctor energy with $\rho_{\text{ds}} = 1.4 \text{ Mg}/\text{m}^3$ ($I_s = 1.00$) and $w_{\text{opt}} = 28.9\%$ was adopted as the main compaction curve. The hydraulic conductivity of lime-softening sludge was determined at degrees of compaction $I_s = 0.95, 0.97, 1.00, 1.03$ and 1.05 with assumed moulding moisture contents $w = 28, 30, 32, 34$ and 36% according to the plan presented in Table 4. Theoretically, forming

Table 4 The plan of short-term permeation test – number of series

No.	Degree of compaction I_s (—)	Moulding moisture content w (%)				
		28	30	32	34	36
1	1.05	3	—	3	—	—
2	1.03	—	4	—	3	—
3	1.00	3	—	8	—	3
4	0.97	—	3	—	3	—
5	0.95	3	—	3	—	3

specimens with parameters above zero air voids ($S_{\text{sat}} = 1.0$) was not possible. In practice, the moulding moisture contents responding to these measurement points were lower by about $\Delta w \approx 0.22\%$ on average than the assumed moisture content. The specimens were formed, but this involved lower compaction and lower moisture content. This came about as a result of squeezing pore water with sludge during specimen formation, and this in turn resulted from changes in the sludge physical properties. Taking into account inaccuracies in measurements and parameter changes at compaction, the assumed measurement points were moved along the zero air voids line. These points were included in the theoretical analysis and the values of the assumed parameters were taken into approximation as the $\Delta w \approx 0.22\%$ was situated towards the error moisture content measurement limit. A series of 42 short-term tests were carried out (Table 5).

The purpose of long-term testing was to evaluate the simultaneous effect of time factor and the permeating liquid on the long-term hydraulic conductivity of the compacted lime-softening sludge. The specimen with parameters corresponding centrally to that of the experimental plan was chosen for the test, i.e. with the degree of compaction $I_s = 1.00$ and the moisture content $w = 32\%$ (Table 4). Table 6 lists the characteristics of the tested specimens. The long-term tests of hydraulic conductivity were carried out on 6 specimens and lasted up to 3.5 months. Tap water, distilled water, NaOH and HCl solutions, as well as municipal landfill leachate (2 specimens) were used as permeating liquids. Readings were taken every few days in the initial period, and every 2 weeks towards the end of the test period. In case of leachate permeation, for ease of test, the measurements were taken after 3 days, 1 week, 2 weeks and about 1, 2, 3 and 3.5 months to 108 days of permeation, beginning from the first measurement date. The

Table 5 The short-term hydraulic conductivity results

No.	Degree of compaction I_s (–)	Dry unit density ρ_d (Mg/m ³)	Bulk density ρ (Mg/m ³)	Planned moisture content w_z (%)	Moulding moisture content w (%)	Final moisture content w_f (%)	Average hydraulic conductivity $k_{10\text{mean}}$ at $i \geq 30$ (10 ^{–9} m/s)
1	1.05	1.47	1.88	28.0	27.71	31.83	2.351
2			1.88		27.85	33.43	4.373
3			1.90		28.88	31.23	8.395
4			1.94		31.60	33.60	0.437
5 ^a			1.94		31.94	34.85	5.563
6 ^a			1.94		32.12	33.99	4.576
7	1.03	1.44	1.86	30.0	29.15	31.95	5.629
8			1.86		29.17	31.35	5.636
9			1.86		29.33	34.12	0.938
10			1.88		30.37	33.87	1.120
11 ^a			1.92		33.17	35.34	9.074
12 ^a			1.92		33.18	34.68	7.502
13 ^a	1.00	1.40	1.92	28.0	33.19	35.93	1.946
14			1.79		27.71	34.06	5.026
15			1.79		27.85	32.85	8.365
16			1.81		28.88	33.78	14.589
17			1.84		31.50	35.35	10.002
18			1.84		31.51	32.78	14.582
19	1.00	1.40	1.84	32.0	31.61	35.73	2.291
20 ^a			1.85		31.94	36.23	7.935
21			1.85		32.12	35.98	4.667
22			1.85		32.20	34.65	5.376
23			1.85		32.20	33.51	3.461
24			1.85		32.20	34.06	4.539
25 ^a	0.97	1.36	1.90	36.0	35.79	34.91	20.782
26 ^a			1.91		36.38	36.07	9.343
27 ^a			1.91		36.62	36.16	5.789
28			1.76		29.36	34.91	8.356
29			1.76		29.36	34.90	4.881
30			1.77		29.64	35.81	2.913
31 ^a	0.95	1.3	1.81	34.0	33.17	36.76	11.316
32			1.81		33.18	37.25	8.547
33			1.81		33.25	36.99	3.243
34			1.70		27.71	36.31	16.697
35			1.70		27.85	37.46	18.365
36			1.71		28.88	36.96	25.077
37	0.95	1.3	1.75	32.0	31.61	38.48	5.115
38			1.76		31.94	38.06	17.251
39			1.76		32.12	37.01	9.700
40			1.81		36.38	38.83	13.371
41 ^a			1.82		36.62	38.20	20.343
42			1.81		35.65	35.82	2.397

^a The pore water with sludge squeezed from the apparatus during specimens forming under the press

Table 6 The geotechnical properties of the tested specimens – the long-term permeation tests

No.	Permeating liquid	Degree of compaction I_s (–)	Dry unit density ρ_d (Mg/m ³)	Bulk density ρ (Mg/m ³)	Planned moulding water content w_z (%)	Moisture content w (%)	Final water content w_f (%)
1	WD	1.00	1.40	1.84	32.0	31.50	37.96
2	WW			1.84		31.50	35.54
3	NaOH			1.84		31.50	36.21
4	HCl			1.85		31.95	35.79
5	O1			1.85		32.40	34.58
6	O2			1.84		31.25	35.38

specimens were held under a permanent hydraulic gradient within a range of 35 to 40 between the measurements. The apparatus were deaerated before each measurement session, and air appeared under the apparatus cover only in the initial period of HCl solution permeation. In the case of HCl, due to the possible occurrence of chemical reaction, the apparatus was deaerated twice a week. Still, air bubbles appeared in the glass standpipe twice during the first 2 weeks of permeation. After about 1.5 months, when no air bubbles were observed during the apparatus deaeration, the apparatus was subsequently deaerated only directly before every measurement.

All tests were run on saturated specimens. This was obtained by exposing them to a 1.20 m liquid pressure head for 3–4 days. In the case of the leachate, the O2 specimen was left under a 1.20 m leachate pressure head for 18 days. The apparatus were deaerated before each test-run.

The short-term and long-term tests were performed at rising hydraulic gradients and lessening hydraulic gradients between 30 and 40 - as per requirements of laboratory tests on hydraulic conductivity for material applied to landfill sites liner systems. In the experiment, 2–3 measurements at hydraulic gradients ≥ 30 rising, as well as at lessening were done. The readings were taken every 15 min. In the case of the landfill leachate, the changes of leachate level were registered every 60 min after 1 week of continuous permeation as indicated by the fading leachate passing through the specimen. The mean hydraulic conductivity values k_{mean} were determined for each test.

The temperatures of all permeating liquids were controlled and their impact on hydraulic conductivity was taken into the consideration. The obtained hydraulic conductivity values were reduced to values at temperature + 10 °C i.e. $k_{10\text{mean}}$ (Pisarczyk 2015). Additionally, the initial hydraulic gradient i_0 was determined. The final moisture contents were established after the test runs were completed.

3 Results and Discussion

3.1 Short- Term Hydraulic Conductivity Tests

The short-term conductivity results are summarized in Table 5.

The parabolic dependence of lime-softening sludge hydraulic conductivity on its compaction and moulding moisture content was established on the basis of the short-term tests. An experimental rote-quasi-uniformal - PS/DS.-P: $\lambda(\bar{\lambda})$ plan was used (Polański 1984) in order to show the impact of these parameters on the lime-softening sludge permeation properties. The approximate polynomial $k_{10\text{av}} = f(w, I_s)$ at $i \geq 30$ for 2 input values: the moisture content w and the degree of compaction I_s , was determined in the range of w [28%; 36%] and I_s [0.95; 1.05]. The hydraulic conductivity arithmetical mean values for specimens prepared at the same moulding water content and degree of compaction ($k_{10\text{mean}}$) were taken for every point of plan as input values ($k_{10\text{av}}$). The values of hydraulic conductivities as applied to the approximation are presented in Table 7.

The approximate polynomial, which is representative of the assumed ranges of input values, was:

$$k_{10\text{av}}(i \geq 30) = 7.478 \cdot 10^{-8} - 1.175 \cdot 10^{-8} w + 5.742 \cdot 10^{-7} I_s + 1.693 \cdot 10^{-10} w^2 + 1.310 \cdot 10^{-9} w \cdot I_s - 3.323 \cdot 10^{-7} I_s^2 \quad (1)$$

The influence of moulding moisture content and degree of compaction on the hydraulic conductivity estimated on the basis of the polynomial is presented in Fig. 2.

The mean values of hydraulic conductivity for edge points of zone recommended for compaction as regards the moulding moisture content was then plotted. The graphic interpretation of hydraulic conductivity

Table 7 The values of hydraulic conductivities k_{10av} applied to the approximation

No.	Input values		Hydraulic conductivity k_{10av} at $i \geq 30$ (10^{-9} m/s)
	Moulding moisture content w (%)	Degree of compaction I_s (-)	
1	30	0.97	5.383
2	30	1.03	3.331 ^a
3	34	1.03	6.174
4	34	0.97	7.702
5	28	1.00	9.326
6	36	1.00	1.197
7	32	1.05	3.525
8	32	0.95	1.069
9	32	1.00	8.958
10	32	1.00	5.992
11	32	1.00	4.000 ^b

^a mean of 4 measurements^b mean of 2 measurements

depending on the moisture content for points responding to the compaction curve is presented in Fig. 3. From the analysis, it followed that as regards permeating properties; lime-softening sludge was similar to that of cohesive soils. The minimum hydraulic conductivity occurred in the wet side of the compaction curve. The hydraulic conductivity decreased beyond the optimum moisture content as

the increment of moisture content affected the increased ability to break down “particle aggregates” as well as the reorientation of particles and the interparticle pores reduction. The hydraulic conductivity reached its minimum at moulding moisture content slightly higher than the optimum when the pore volume and distances between particles were the smallest. Further rise in moulding moisture content caused the hydraulic conductivity increase as the free water pushed soil particles away, increasing soil porosity. Hence, there was a certain range of moulding moisture content at which it was possible to obtain the minimum hydraulic conductivity (Benson and Daniel 1990; Lambe 1954; Mitchell et al. 1965).

Flow through the sludge within a range of moisture content between ($w_{opt}-1\%$) to ($w_{opt}+7\%$) and degree of compaction $I_s = 0.95-1.05$ came about only after crossing the initial hydraulic gradient i_0 as in cohesive soils (Roza 1950). The initial hydraulic gradient of lime-softening sludge was determined to be $i_0 \geq 13$. The estimated mean value of sludge hydraulic conductivity k_{10mean} at hydraulic gradients ≥ 30 varied between $4.37 \cdot 10^{-10}$ m/s to $2.5 \cdot 10^{-8}$ m/s, and is characteristic of clayey sandy silts and deemed as being very low (Pisarczyk 2017).

The moulding moisture content and the compaction affected the hydraulic conductivity of the tested sludge. The minimum value of hydraulic conductivity was obtained at the wet side of the compaction curve at a

Fig. 2 The short-term hydraulic conductivity versus the moulding moisture content and the degree of compaction estimated on the basis of polynomial

Fig. 3 The hydraulic conductivity versus (a) the moulding moisture content and (b) dry density

moisture content of about $w = w_{\text{opt}} + 2\%$. The minimum hydraulic conductivity value, estimated on the basis on the approximation for $k_{10\text{av}}$ at hydraulic gradients ≥ 30 , for points with parameters responding to that on the compaction curve, was about $6.5 \cdot 10^{-9}$ m/s and occurred at $w \approx w_{\text{opt}} + 1\% \approx 31\%$. So as to guarantee the lowest hydraulic conductivity, a moisture content $w = w_{\text{opt}} + 2\%$ should be assumed for the investigated lime-softening sludge.

The estimated values of hydraulic conductivity for the lime-softening sludge were similar to the values presented in the literature, i.e. between 10^{-10} m/s to 10^{-7} m/s (Glysson 1972; Raghu and Hsieh 1985; Maher et al. 1993). Glysson (1972) observed that the sludge hydraulic conductivity increased as the porosity index increased, i.e. the compaction influence on the permeation properties of sludge. Glysson (1972) also concluded that lime-softening sludge is a material with low to very low hydraulic conductivity.

3.2 Long-Term Hydraulic Conductivity Tests

The time and permeating liquids factors impact on the sludge hydraulic conductivity $k_{10\text{maen}}$ at hydraulic gradients ≥ 30 is presented in Fig. 4. The permeating liquids and obtained leachate were observed during a 3.5-month permeation time. Changes in the liquid medium structure were noticed after 1.5–2 months permeation, in the glass stand-pipe directing the distilled water, tap water, as well as NaOH and HCl solutions. Herein, flocculent solidification occurred. This made the hydraulic gradient decrease practically unfeasible at the last conductivity measurements (after about 3 months

of permeation). After about 1–1.5-month test duration, a white bloom was seen on the vessel surface in which the obtained leachate was collected for the NaOH test. After about 2 months, the outflow pipe was coated with a white, crystalline substance that was soluble in water. Dark solids were noticed on the walls of municipal leachate containers in the period of constant hydraulic gradient ≥ 30 between the hydraulic conductivity readings. Finally, a clear segregation of leachate was noticed in the glass stand-pipe i.e. darker solid containing liquid under the lighter.

The initial leachate obtained from the tap water, distilled water, NaOH and HCl test solutions was almost similar to the permeating liquids in colour. The colours changed in time from light yellow to dark yellow, whereas the obtained leachate was light yellow in colour in the first days for the municipal leachate permeation. That colour became more and more intense in time, reaching dark brown in the end. After a month, the obtained leachate was only a little lighter in colour than the permeating leachate. The quantity of collected leachate also decreased in time, and after about 2 months, leachate outflow was practically imperceptible.

There was no unpleasant smell in case of distilled water, NaOH and HCl solutions. An unpleasant odour was noticed in the tap water test, while a very unpleasant odour, similar to the smell of leachate from municipal waste landfill, was encountered in the leachate permeation tests. There was also black coating removable by scraping on the tap water test specimen's surface. This changed in colour to brown-green at a depth of about 0.005 m even up to half the height of the specimen (Fig. 5). The top surface of the specimen permeated

Fig. 4 The time and permeating liquids impact on the sludge hydraulic conductivity $k_{10\text{maen}}$ at hydraulic gradients ≥ 30

Fig. 5 The lime-softening sludge specimen affected by the tap water in long-term permeation test

with distilled water was without major contamination and changes (Fig. 6). The top surfaces of the specimens affected by the NaOH and HCl solutions seemed to be slightly less hard compared to the surfaces of specimens permeated with tap and distilled waters (Fig. 7 and 8). The surface of the specimen affected by HCl solution was darker compared to the state before permeation and was loose to a depth of about 0.005 m. Underneath there was a hard material, the colour of which was similar to that of bronze in the central part but whitish elsewhere (Fig. 8). The most significant changes were observed in specimens permeated with municipal waste landfill leachate. Their surfaces were a residue black in colour (Fig. 9). However, there was no noticeable change in the structure or colour of the sludge after removing the 0.02-m layer of material.

Fig. 6 The lime-softening sludge specimen affected by the distilled water in long-term permeation test

Fig. 7 The lime-softening sludge specimen affected by the 0.2% solution of sodium alkali (NaOH) in long-term permeation test

The results of hydraulic conductivity initial measurements showed that the values of conductivity $k_{10\text{mean}}$ were similar and varied between $1.5 \cdot 10^{-8}$ m/s to $1.0 \cdot 10^{-8}$ m/s for tap water, distilled water and NaOH and HCl solutions. There was no significant increase of hydraulic conductivity in the first week of tap water, distilled water and HCl solution permeation, however, hydraulic conductivity suddenly decreased up to $6.0 \cdot 10^{-9}$ m/s and afterwards increased in the first week when the lime-softening sludge was permeated with the NaOH solution. A gradual decrease of hydraulic conductivity was also observed in the distilled water (lightly alkaline medium, $\text{pH} = 7.80\text{--}8.38$) and tap water (neutral medium, $\text{pH} = 7.49\text{--}7.70$) long-term tests. Here, hydraulic conductivity stabilized at level $k_{10\text{mean}} \approx 3.0 \cdot 10^{-9}\text{--}4.0 \cdot 10^{-9}$ m/s after about 70 days of permeation

Fig. 8 The lime-softening sludge specimen affected by the 0.43% solution of hydrochloric acid (HCl) in long-term permeation test

Fig. 9 The lime-softening sludge specimen affected by the leachate (O2) in long-term permeation test

in both cases. In contrast, the hydraulic conductivity gradually increased and stabilized at the level $k_{10\text{mean}} \approx 2.0 \cdot 10^{-8}$ m/s in the case of the permeation of the extreme reaction liquids: 0.2% NaOH solution (strongly alkaline medium, pH = 11.7–11.8) and 0.43% HCl solution (strongly acid medium, pH = 2.38–2.63), regardless of solution. Herein, the lime-softening sludge hydraulic conductivity measured at first leachate passage through the specimen was lower than the hydraulic conductivity determined for other permeating liquids, and stabilized at level $k_{10\text{mean}} \approx 5.0 \cdot 10^{-9}$ – $8.0 \cdot 10^{-9}$ m/s at hydraulic gradients ≥ 30 . The results confirm the decrease in the hydraulic conductivity of clay to municipal leachate relative to the hydraulic conductivity to distilled water (Rauen and Benson 2008).

There was a significant hydraulic conductivity time dependent behaviour when the lime-softening sludge was permeated with leachate to $k_{10\text{mean}} \approx 4.7 \cdot 10^{-11}$ – $6.8 \cdot 10^{-11}$ m/s in 108 days of testing. The most intensive decrease occurred in the first month of permeation. As the water and leachate exposure time increased, the lime-softening sludge hydraulic conductivity showed a reduction that corresponded to changes that were similar to that of cohesive soils, for which also the hydraulic conductivity decreased in time (Thomas and Brown 1992; Aldaeef and Rayhani 2014). Of note, Majer (2005) concluded that the hydraulic conductivity of compacted low permeable landfill liners decreases in time, but with stabilization occurring after about 2 years from its construction.

The hydraulic conductivity initial value of lime-softening sludge permeated with landfill leachate was similar to the values obtained for leachate passage

through clays, for which the conductivity changed in time and varied between $1.0 \cdot 10^{-10}$ m/s and $6.0 \cdot 10^{-9}$ m/s (Sai and Anderson 1991). Daniel et al. (1988) also observed a time influence on the hydraulic conductivity for leachate passage through cohesive soils. Their work revealed that in a 3-month period, for three kinds of landfill leachate and three different cohesive soils, the initial hydraulic conductivity varied between $5.0 \cdot 10^{-10}$ m/s and $1.0 \cdot 10^{-8}$ m/s and decreased in time, in the extreme case, up to $3.0 \cdot 10^{-10}$ m/s. Moreover, the ratio of final hydraulic conductivity k_f and the initial one k_i was on average $k_f/k_i = 0.6 \div 0.9$. A hydraulic conductivity ratio was established as being $0.02 \div 0.06$ for natural soils affected by leachate in time (Francisca and Glatstein 2010). Here, the ratio k_f/k_i was established to be 0.01 for the tests carried out for leachate. The obtained results point towards a higher decrease of conductivity in time when compared with literature data. It is thought that differences in chemical characteristics of the leachate used in the tests, as well as the diverse characteristics of the tested sludge could be a reason for such ratio discrepancies. Indeed, the presence of biomass in used leachate could cause a decrease in the long-term hydraulic conductivity by pore clogging (Francisca and Glatstein 2010). This was observed in the carried out tests. The studies of Roque and Didier (2006) indicate that in the permeation of leachate with pH = 7.0 through the specimen, the lowest hydraulic conductivity of cohesive soils (in this case, about 10^{-9} m/s), was obtained at $w = w_{\text{opt}} + 2\%$. These results underline the similar characteristics of hydraulic conductivity dependence on the moisture content for water and leachate permeation.

4 Conclusion

The hydraulic conductivity tests performed on lime-softening sludge revealed that the compaction and the moulding moisture content significantly affected lime-softening sludge hydraulic conductivity. The results of short-term tap water permeation tests also showed that all hydraulic conductivity values at hydraulic gradients ≥ 30 were less than $2.5 \cdot 10^{-8}$ m/s, when water content was between 27.7% to 36.7% and the degree of compaction was between 0.95 to 1.05.

In planning the set of experiments, the lowest hydraulic conductivity was predicted to be $6.5 \cdot 10^{-9}$ m/s at the corresponding moisture content 31%. However,

permeation time and the permeating liquid affected the hydraulic conductivity of lime-softening sludge. The long-term tests revealed that the factors permeating liquids and permeation time had significant effect upon the initial hydraulic conductivity. The long-term hydraulic conductivity showed negligible dependence when permeated with strong acid and strong alkaline solutions as it increased for NaOH and HCl solutions from $2.0 \cdot 10^{-8}$ m/s to $12.0 \cdot 10^{-9}$ m/s. A reduction of hydraulic conductivity in time was observed for liquids characterized by neutral or light alkaline reaction as it decreased for tap water, from $1.0 \cdot 10^{-8}$ m/s and for distilled water, from $1.5 \cdot 10^{-8}$ m/s, to $3.0 \cdot 10^{-9}$ – $4.0 \cdot 10^{-9}$ m/s.

The hydraulic conductivity estimated for permeating municipal leachate was significantly different from the results obtained for other permeating liquids. Here, leachate passage through the specimen brought about a significant reduction of lime-softening sludge hydraulic conductivity – from $5.6 \cdot 10^{-9}$ – $8.1 \cdot 10^{-9}$ m/s to $4.7 \cdot 10^{-11}$ – $6.8 \cdot 10^{-11}$ m/s – i.e. the required value of hydraulic conductivity for materials used as landfill liners ($k_{10} \leq 1.0 \cdot 10^{-9}$ m/s). Thus, a significant decline was expected just after one week's permeation. With regard to municipal leachate passage through landfill liners made of lime-softening sludge, a decrease of hydraulic conductivity of even 2 orders of magnitude in 3 months permeation time is held to be possible. Pore clogging and bioactivity is thought to be the main reason for which the hydraulic conductivity of lime-softening sludge decreased for municipal leachate permeation. Herein, flow through the layer could not even occur at hydraulic gradients $i \leq i_0 = 13$. Hence, the menace of possible washing carbonates and other compounds away would be eliminated as a consequence of hydraulic conductivity decrease in time and permeating leachate quantity imitation.

Thus, the laboratory tests of geotechnical properties confirmed that in some determined conditions, the lime-softening sludge could replaced the natural soil layers in waste-landfill liners as the investigated material meets the requirements for the liner systems of inert landfill sites ($k \leq 1.0 \cdot 10^{-7}$ m/s) regardless of permeating solution pH and the limit value for hazardous and non-hazardous waste landfills ($< 1.0 \cdot 10^{-9}$ m/s).

The physic-chemical, mechanical and permeation properties of lime-softening sludge should be more widely recognized in order to safely re-use it. Lime-softening sludge should be additionally tested in the field for working out the appropriate test methodology

and for verifying the results of laboratory geotechnical measurements. The changes of leachate and sludge chemical properties after permeating liquids passing through are going to be emphasized in further research. The stress will be put on the changes in structure of the sludge affected by permeating liquids as well.

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