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INFLUENCE OF CLAY MINERAL CONTENT ON LANDFILL CAP COVER WITH SPECIAL ATTENTION TO SWELLING

1. Introduction

Landfill cap cover is a complex structure consisting of layers of geotextiles, drainage installation and mineral layers. It is used to protect the population and the environment from the cumulative radiation from a very low level of radioactive waste. Requirements of the Regulation [1] and ITB instruction [2] specify the minimum thickness of the aperture and its physical and mechanical properties. Soil with a high content of fine grained clay fraction is commonly used due to its insulating layer of the mineral sealing. After closure of the landfill it is important to maintain pre-set parameters for a long period of time with special concern to water content.

The moisture loss leads to the changes in the soil parameters of the landfill cap cover. The bonds between the individual grains are hardening which can lead to the formation of cracks. The prolonged aperture settlement makes it rupture which allows the waste to penetrate the environment. In this case, the swelling of the ground which can partly increase at some point, can be beneficial to the landfill in terms of safety as well as it can prolong the time needed for the repair. The mechanism of swelling process which occurs in the case of the aperture is shown with figure 1.

The mineral composition and particle size distribution of soil determines its swelling properties. The mineral composition of each soil clay includes - in varying proportions - the three main minerals: illite, kaolinite, montmorillonite. The study was carried out to determine whether clay soils occurring in Krakow and in the vicinity could be useful for the purposes of landfill cap cover, mainly in terms of expansiveness. A high level of classification research has been carried out with a particular emphasis on the degree of swelling. A broad in-depth analysis of the case proves that it is possible to determine the approximate mineral composition of soils using a simplified method. Classification parameters were chosen according to [3].

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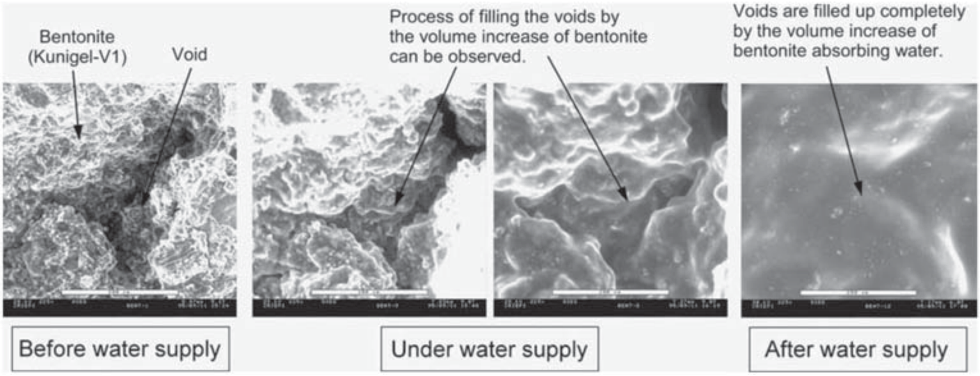

Fig. 1. Stages of swelling process due to water content increase for bentonite soils [4]

The Tertiary compositions occurring in Krakow were used as research material with special attention to reconstituted and undisturbed samples. The samples were analyzed according to Casagrande's plasticity chart while determination of swelling and mineral composition was based on the modified method of free swelling.

2. Clay soil and Clay minerals

In an engineering work, the particle size as well as moisture and geological background determine the mechanical properties of fine-grained soil. For an engineer the clay is a complex soil structure as defined by [5], which states that clay is a mixture of clay fraction and other fraction types that are dependent on geological processes that occurred in the ground. A particle fraction, which determines the engineering properties of soil, is a main fraction of fine grained soils. A 0.002 mm particle size clay soil consists of minerals all of which determine size, shape and properties of water molecules, defining clay soil properties. The clay soil mineral composition is made up of three main groups of clay minerals: kaolinite (Kaolinites), montmorillonite (Smectites), illite (Micas).

Clay minerals are of the phyllosilicates family but they contain an additional, though different layer of Silicates [4]. Silicates combine together into structures of layer silicates within their own group (known as clusters). Clusters consist of two basic units — the first is *silicon tetrahedron* and the second is *aluminium (magnesium) octahedron*. For the purpose of the article, only the two most important minerals and their influence on swelling will be described, kaolinite — which is a non-swelling clay mineral and montmorillonite which is highly expansive.

Kaolinite — consists of both silica and octahedral sheets which can change their positions. Due to double bonding between those layers – created by Van Der Vaals forces and hydrogen bonds — kaolinite has non-swelling behavior. Cation-exchange capacity is about 3–15 meq/100g, but besides that posses negative charge, and due to small specific surface, carries much greater surface charge density. Well-crystallized particles of kaolinite have well-formed six-sided plate shape, poorly crystallized kaolinite obtaines a hexagonal plate shape. Because of slight differences in the oxygen-to-oxygen distances in the tetrahedral and octahedral layers, there is some distortion of the ideal tetrahedral network. As a result, kaolinite, which is the most abundant member of the subgroup and a common soil mineral, is triclinic instead of monoclinic [4].

Montmorillonite — consists of an octahedral sheet sandwiched between two silica Sheets. All the tips of the tetrahedra point toward the center of the unit cell. The oxygens forming the tips of the tetrahedral are common to the octahedral sheet as well. The anions in the octahedral sheet that fall directly above and below the hexagonal holes formed by the bases of the silica tetrahedral are hydroxyls. Due to dioctahedral structure charge deficiency, which is usually about 0.66 per unit cell, may result in all aluminium ions in favor of magnesium ions. Under electron microscopy it is possibile to see that monmorillonite particle are more like thin flakes [4].

The summary of selected methods for determining the mineral composition and their brief description with an indication to the test procedures is presented with table 1.

3. Testing material and research methodology:

The ground samples were obtained from an area within a scientific polygon of Cracow University of Technology (Politechnika Krakowska) located in Krakow. It is a tertiary composition lying under a layer of Quaternary silts and sands with a consistency defined as *very stiff*. Soil samples of a category A and B according to [6] were taken from a depth of 3.5–5.0 m using a manual method. In the research studies, the researchers used the cohesive remoulded material that met the criteria of applicability tests and also a category A samples if the standards [7] and [8] or technical specifications [9] were applicable. The test procedures were used to determine the physical parameters of soil, all of which can be specified in standard [7], and technical specifications [9]. The physical parameters obtained are as follows: bulk density is about 2.10g/cm^3, dry density is 1.70g/cm^3 and the specific density was determined at 2.67g/cm^3 while the natural water content was about 25%. The liquid limit has been obtained with the use of Casagrande percussion apparatus, according to [7] and [10]. Plastic limits have been determined in accordance with [7]. As a result, the Atterberg limits were obtained at 33% for a plastic limit, and 83% for a liquid limit, all of which puts plasticity index at 50–60%. According to [7], particle size distribution classified Krakow clay as over 25% of clay fractions, 60–65% of silts and 10% of sands.

To understand the mechanical behavior, one of the stages was to use two classification tests for expansiveness prediction. The first classification test was based on an Atterberg plasticity classification chart of cohesive soils. The second test was on the basis of a classification chart for expansive fine grained soils using Free Swell Ratio. The above test allows one to determine a dominant clay mineral [10]. It could be useful to follow [10, 11] and others to claim that using an Atterberg Chart requires using Casagrande apparatus which is not mentioned in standards [5, 9]. The use of an Atterberg Chart excludes the use of Cone Penetration Test, owing to different results of plastic and liquid limits. The final results of classification

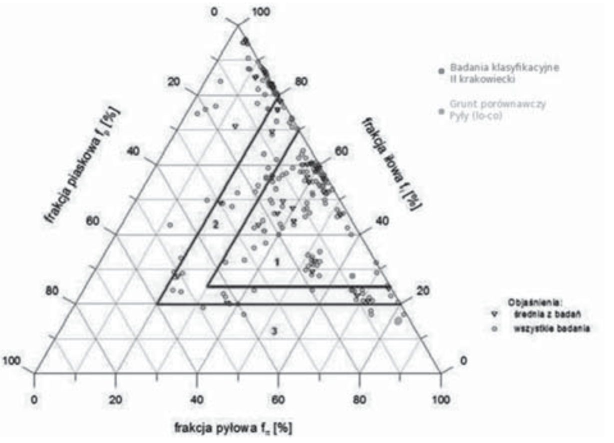

Fig. 2. Fraction content due to [15] with classification for cap cover utility by [3]

TABLE 1

Classification of fine grained soils for landfill cap covers [14]

Criterion	Unit	Limit according to [3]	Krakow Clay
Filtration coefficient	m/s	max. 10^{-9}	fullfilled
Clayey fraction content (grain size < 0,002 mm)	%	min. 20%	min. 25%
Sum of clay and silt fraction	%	min. 30%	> 50%
Liquid limit	%		~65–75%
Plasticity Index	%	$10 < I_p < 30$ and $30 < I_p < 60$	above 30%, usually about 55–60%
Clay minerals content	%	min. 20 %	more than 20%
Organic parts content	%	< 5%	< 5%

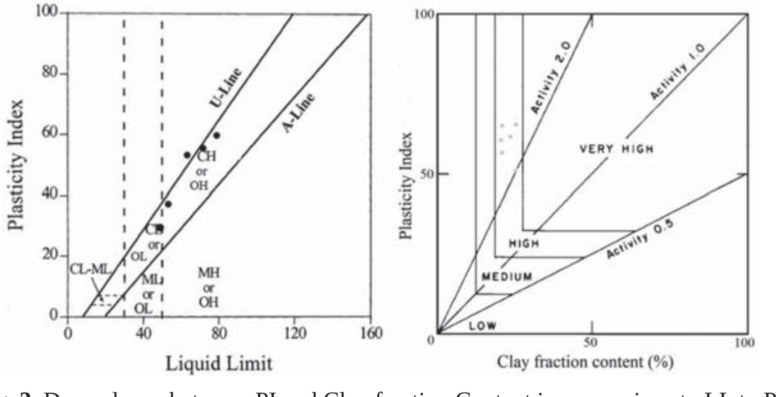

Fig. 3. Dependency between PI and Clay fraction Content in comparison to LL to PI

TABLE 2

The most popular methods of determining clay minerals content

No.	Method	Description	Additional information
1	X-ray diffraction	— main requirement is to use pure material only due to difficulties in the recognition of pure minerals (different and complex spaces between them), — it is possible to determine clay minerals by known basal spacing between silicates layers, — the need to avoid background "noises",	— Possible to identify clay minerals with high precision, — process is not easy and/or quantitative, — mostly for recognition of structural groups and sub-groups, — Ability to record long spacing minerals — from 5–10-15 up to 25A,
2	Differential Thermal Analysis	*Based on*: Le Chatelier invention *Method assumes*: measurement of temperature during the dehydration of samples *Temperature*: ~1000 degrees *Problem*: soil test results obtained in different labs vary	— easy to prepare, — peak appearance with relation to temperature shows reaction corresponding to specific mineral, — e.g. Kaolinite at 550–600°C.
3	Electron microscopy	*Based on*: Assumption based on electron microscopy theory *Method assumes*: high precision and Electron diffraction is similar to X-ray diffraction, Possibility of viewing the sample during scanning	Possibility of determination: — microstructure characteristics of clay minerals in nanometer scale, — the microchemistry of clay particle, — clay poly-type structures,
4	Determination of specific surface	*Based on*: BET theory *Defined as*: either a surface to volume or surface to mass ratio *Method assumes*: measurement of gas absorption and its relation to possible surface of clay minerals	Specific surface is inversely proportional to particle size
5	Cation Exchange Capacity	Distinguish cation exchange from ligand exchange, and exchange of diffuse layer absorbed cations *Method assumes*: determine the extractable cations (K+, Ca2+, Mg2+, and Na+) and estimate H+ and Al+3 from soil and buffer pH measurements	— direct measurement is costly, — for calcined clay it's much higher, — results might be inadequate for soils consisting of organic matter, — depends on method; the determined values of CEC are dependent on the method used.
6	Determination of Atterberg Limits	*Based on*: Classification of cohesive soils due to Atterberg limits, basing on Atterberg plasticity chart *Method assumes*: by using Atterberg's classification chart it is possible to determine expansiveness of clay *Requires:* Cassagrande percussion apparatus	— in CEN ISO TS-17892 Atterberg limits are determined only with cone penetrometer test, — in BS1377-2 there's a commentary on using this method: it is difficult to maintain the apparatus in accordance with standards and the test results are subject to the judgment of the operator
7	Thermo-gravimetric Analysis	*Based on*: theory of drying velocity for particular clay minerals *Method assumes*: measure of gaining/losing weight while heating	— this method is complementary with Differential Thermal Analysis, — results are compared to curves from DTA.

tests with comparison to existing limits according to [2] are presented in the grid below: Table 1: Classification of fine grained soils for landfill cap covers.

Particle size distribution as well as other physical and mechanical properties allow the use of clay material from the vicinity of Krakow in mineral barriers of landfill cap cover.

4. Determination of dominant Clay mineral

The test methods used to determine the mineral composition in Table 1 are widely used in laboratory practice. It should be noted that each method requires the use of high quality research equipment which is expensive to buy and operate not to mention the fact that it is impossible for researchers to determine minerals composition in a fast but cheap method. In some cases, it would be advisable to use low-cost methods that present mineral composition of the soil samples in a qualitative way. Several research teams around the world have chosen the Free Swell Method to determine mineral composition [16], and its usefulness has been defined as good.

Free Swell Method — a method similar to the one used in the research studies for Krakow clay — was first described in 1956 by Holtz and Gibbs. The method was originally based on a comparison between the sediment parameters obtained from a blend of dry soil and 10 cm^3 of distilled water and the ones obtained from a blend of dry soil with paraffin.

For some tests, especially for high Kaolinite content soils, negative values of Free Swell Ratio were common result. Later, by observation [10] that equilibrium sediment volume of Kaolinite-rich soils in non-polar liquids like carbon tetra chloride gives results greater than the equilibrium sediment volume in comparison to suspension made with distilled water and dried soil specimen. This test procedure was modified to current *Modified Free Swell Ratio* with aim to categorize dominant mineral packages. Test results obtained from the modified Free Swell Ratio for Krakow's Clay and some other comparative soils are presented in Table 4.

TABLE 3
Classification of soils based on free swell ratio [10]

Free Swell Ratio	Soil Expansivity	Figure 4	Clay Type	Dominant Clay Mineral Type
< 1.0	Negligible	I	Non-swelling	Kaolinitic
1.0–1.5	Low	II	Mixture of swelling and non-swelling	Kaolinitic and Montmorillonitic
1.5–2.0	Moderate	IIIA	swelling	Montmorillonitic
2.0–4.0	High	IIIB	swelling	Montmorillonitic
> 4.0	Very High	IIIC	swelling	Montmorillonitic

5. Test results

Test results obtained with the method of Free Swell Test are presented in table 4 and with figure 4 also shows two groups of one specimen, the first one with 3rd degree of expansivity (shown as triangles) and the second one with 2nd degree for 50–50% kaolinitic-montmorillonitic soils (presented as dots). During the analysis of Free Swell Ratio we compared the obtained results with existing mineral analysis performed with a few methods mentioned in Table 1. Most of the results classified Krakow Clay as a partly swelling clay-type. It might suggest that Krakow Clay can change its volume over the time after the closure of the landfill. For several specimens, (there were five but only three are mentioned in this work), we obtained higher values of $V_{d,cc}$ than of $V_{k,cc}$ that places the samples at a IIIA group. In comparison to the whole base of all conducted analyses, the obtained results are not of a poor quality because the samples might have had different expansivity properties due to various depth extraction.

TABLE 4
Modified Free Swell Ratio test results for dominant clay mineral determination for Krakow Clay

Specimen no.	Liquid Limit	Plasticity Index	Free Swell Ratio	Degree of expansivity (Table 3)	Dominant Clay Mineral(s) Predicted from MFS
10C/7	88	59	1.22	II	Kaolinitic and Montmorillonitic
10C/10	90	62	1.9	IIIA	Montmorillonitic
10C/12	89	59	1.59	IIIA	Montmorillonitic
S_Z_56	85	58	1.45	II	Kaolinitic and Montmorillonitic
S_Z_78	85	58	1.55	II/IIIA	Montmorillonitic
S_Z_03	80	61	1.29	II	Kaolinitic and Montmorillonitic
S_Z_103	78	50	1.15	II	Kaolinitic and Montmorillonitic

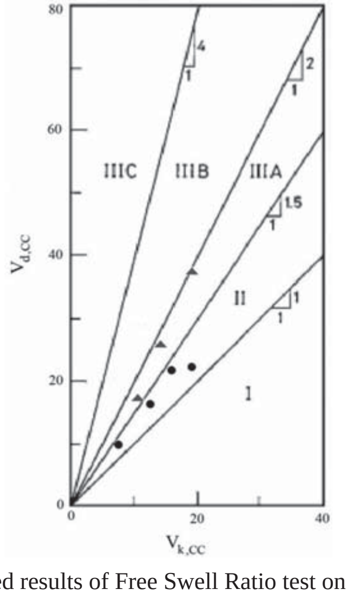

Fig. 4. Obtained results of Free Swell Ratio test on Krakow Clay

6. Summary

There has been a conclusion drawn regarding the usefulness of simple analysis methods to identify the mineral composition and the use of clay shrouds from the regions of Krakow for the landfill cap covers. Classification tests for Low Level Radioactive Waste Landfill usefulness [3] define Cracow Clay as applicable to landfill cap cover. What is more, particle size distribution as well as physical properties especially with Atterberg limits and clay fraction content (Fig. 3) prove the above statement. Apart from that, all the methods of soil minerals presented in table 1 give much more precise results but they are very demanding during test procedures, though.

Those test procedures also require sophisticated instrumentation e.g. X-ray apparatus, high temperature oven for DTA, and what is most important — a wide database of knowledge about technology of a specific test.

The Free Swell Ratio, predicts the degree of soil expansivity much more realistically and gives additional information about the clay mineralogy of soils (Table 3). The method is very simple, quite cheap and gives fair and approximate but qualitative results which have been proved in the presented work.

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