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Determination of Hydraulic Conductivity Based on (Soil) - Moisture Content of Fine Grained Soils

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1. Introduction

The chapter will be divided into the subchapter material, processes and systems. The first one will focus on the physical, chemical and dynamic material properties and their measuring methods. The second specifies the dynamic of the surface moistening and fluid flow. The comprehensive characterization of materials is prerequisite to understand processes in large (geo)-technical systems and their manipulation. The transition from nano (material) via meso (processes) to macro scale (systems) will be illustrated with an example in the third chapter.

2. Materials

The properties of fine grained soils such as silt or clay considerably influence the migration of water. Especially their small pore sizes, their platy habit, and their high specific surface area generally lead to very low hydraulic conductivities. Therefore, it is indispensable to accurately determine these properties for a reliable assessment of the hydraulic conductivity. Fine-grained soils are soils with a grain size distribution ranging from 0,0002 to 0,2 mm, i.e. soil textures from clay and silt up to fine sand. The hydraulic conductivity, K_f , of these materials is generally smaller than 10^{-4} ms^{-1} . In the following the most important physical, chemical and dynamical properties of fine grained materials that affect the hydraulic conductivity will be explained in detail and their measurement methodologies will be illustrated.

2.1 Material properties

Generally the hydraulic conductivity depends on the soil matrix, the type of the soil fluid (density and viscosity), and the relative amount of soil fluid (saturation) present in the soil matrix. In this chapter we will focus on the important properties relevant to the solid matrix of fine grained soils which include the texture and fabric and its mineral phase content.

2.1.1 Physical material properties

2.1.1.1 Soil density

In soil science one can distinguish between the soil particle density ρ_p which is the mass of the soil particle m_p per volume of the particle V_p and the so called bulk density which is the ratio of the mass to the bulk volume V of a given amount of soil. For the latter two different definitions depending on the scientific discipline or the application are possible, i.e. the wet bulk density ρ_{wb} which is the soil mass m_s , plus the mass of the including water m_w , per unit volume V and its dry bulk density ρ_{db} which is the mass of oven-dry soil per unit volume of moist soil.

$$\rho_p = \frac{m_p}{V_p} \quad (1)$$

$$\rho_{wb} = \frac{m_s + m_w}{V} \quad (2)$$

$$\rho_{db} = \frac{m_s}{V} \quad (3)$$

2.1.1.2 Soil texture (grain size and grain size distribution)

Mineral soils are mostly classified according to grain size and grain-size distribution (grain-size fractions and grading) also generally summarized as the soil texture. A major factor that influences the rate of water flow through soils is the size of the particles. It can generally be stated that the smaller the particles, the smaller the voids and the stronger the flow resistance. This explains the very low hydraulic conductivity of fine-grained materials and their preferred use as sealing elements for example in buffers and backfills of underground storage facilities or landfills. The grain-size distribution gives a good picture of the content of clay minerals since they appear almost entirely in the clay fraction ($< 2 \mu\text{m}$). The compaction properties of the material can be estimated by the character of the entire grain size curve, from which one can conclude, if the material has very low hydraulic conductivities and can be used for water sealing purposes (Pusch, 2002).

Whereas the grain-size distribution of the coarse fractions (gravel and sand) can be determined by sieving, the fine fractions (silt and clay) must be determined by sedimentation of the dispersed soil.

2.1.1.3 Soil structure (porosity and pore size distribution, geometry and shape of the pores, tortuosity)

The term structure of soils generally refers to the pore geometry. The pore volume or porosity is a measure of the void spaces in the soil and is a fraction of the volume of voids over the total volume, between 0-1 or as a percentage between 0-100 %.

$$\Phi = \frac{V_v}{V} \quad (4)$$

where V_v is the volume of void-space and V is the total or bulk volume.

Porosity of surface soil typically decreases as particle size increases. This is due to soil aggregate formation in finer textured surface soils when subject to soil biological processes. Aggregation involves particulate adhesion and higher resistance to compaction. The transport of water, solutes, and gases in soil is not only influenced by the absolute size of the pore volume but also on the nature of how the pores are connected which is summarized

under the term tortuosity. In soil the tortuosity is closely related to soil surface area and the pore-size distribution. Both porosity and tortuosity of fine-grained soils are considerably small due to the plat-like shape of the particles.

2.1.2 Chemical material properties

2.1.2.1 Composition of the mineral phases

Minerals in natural soils originate from degraded rock, most of them belonging to the silicates, sulphates, sulphides, and carbonates. Especially the clay minerals show a platy habit leading to a very high surface area to mass ratio and have a considerable influence on the hydraulic conductivity. The distinction of minerals is mostly based on crystal structure and chemistry. Their crystal structure is responsible for a number of their characteristic chemical properties such as cation exchange capacity or high sorption capacity (Pusch, 2002).

Hydraulic conductivity of swellable clayey and clay-enriched silty soils strongly depends on the density, the type of the adsorbed ions and the salinity of the percolating water (Scheffer, 1992). For example it is the high swelling properties that provide sodium bentonite's unique sealing qualities. As the clay hydrates and swells, the path for water to flow through becomes complex as the clay platelets intersperse. The large fraction of interlamellar, immobile water in smectites yields a much lower hydraulic conductivity than of soils with other minerals at any bulk conductivity. Thus, clays with micas, illites, and kaolinites as major minerals are about 100 to 100000 times more conductive than smectite in montmorillonite form at one and the same void ratio (Pusch, 2002). (See figure1)

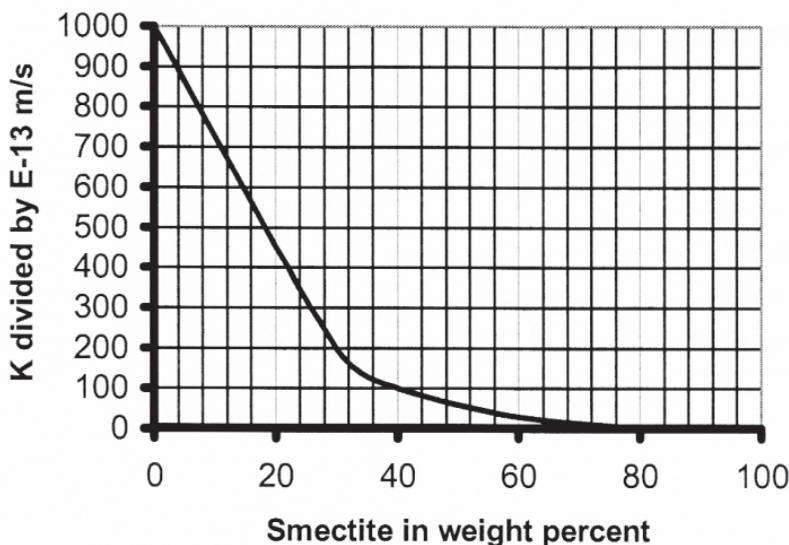


Fig. 1. Approximate relationship between smectite content and hydraulic conductivity (Pusch, 2002).

The type of absorbed cations of the clay is also important with respect to the hydraulic conductivity since bi- and polyvalent cations cause growth in the stack thickness and size, which means that the voids between the stacks of lamellae and thus the hydraulic conductivity are bigger in the Ca than in the Na montmorillonite (Pusch, 2001). Also the electrolyte concentration has a substantial influence on the interparticle distance at low and

moderate densities and thus on the hydraulic conductivity because the stacks of lamellae that form a network with rather much space will coagulate at high electrolyte concentrations.

2.1.2.2 Specific soil surface

The magnitude of the specific surface area of a soil depends largely on the amount of clay and type of clay minerals in the soil. The specific surface area differs largely between types of clay minerals (Table 1). Especially in fine-grained materials one can distinguish between an external and an internal surface, the latter being the interlamellar space of the minerals. The total specific surface area is a factor that can relate grain-scale properties to macro-scale physical and chemical properties of a porous medium. Large specific surface areas lead to much interaction of ions and water molecules with the soil particles. Therefore the total surface determines many physical and chemical properties of the soils (Petersen et al. 1996). In porous media filtration theory, a nonlinear relationship between saturated hydraulic conductivity and surface area has been established for structureless, randomly packed, non-compressible particles (Kozeny, 1927; Carman, 1937; Grace, 1953), the so-called Kozeny-Carman relationship.

Mineral	Specific surface area, m ² /g	
	External	Internal
Kaolinite	10–15	10–15
Illite	100–150	100–150
Smectites	100–150	800–1000

Table 1. Typical specific soil surface area data of clay minerals (Pusch, 2001).

2.1.3 Dynamical material properties

2.1.3.1 Matric potential

Matric potential refers to the tenacity with which water is held by the soil matrix and, in the absence of high concentrations of solutes, is the major factor that determines the availability of water to plants. Differences in the value of matric potential between different parts of the soil also provide the driving force for the unsaturated flow of soil water after any differences in elevation have been allowed for (Mullins, 1991).

The total potential ψ_t of soil water refers to the potential energy of water in the soil with respect to a defined reference state and can be divided into three components:

$$\psi_t = \psi_p + \psi_g + \psi_o \quad (5)$$

with ψ_p as pressure potential, ψ_g as gravitational potential and ψ_o as osmotic potential.

The sum of gravitational and pressure potential is called the hydraulic potential ψ_h . Differences between the hydraulic potential at different places in the soil provide the driving force for the movement of soil water. Matric potential ψ_m is a subcomponent of pressure potential and is defined as the value of ψ_p where there is no difference between the pressure of air or gas in the soil and the gas pressure on the water in the reference state (Mullins, 1991).

The relationship between water content and soil water potential (capillary tension) is described in the water retention curve. The curve is characteristic for different types of soils and is also called soil moisture characteristic. It also depends on the geometry and network of the pores. Fine-grained soils show high residual water contents and high changes in capillary tension are necessary that they release the water.

The retention curve shows a hysteresis which means that depending on the history of the soil with regard to watering and drainage, the shape of the curve is different, a behaviour which is explained by the 'ink-bottle model' (Schuhmann, 2002).

For the analytical description of the water retention curve there are different approaches. Brooks and Corey (1964) combine the mathematical θ/ψ -relationship with the conductivity model of Burdine (1953), but the most common approach is the one from van Genuchten (1980) combined with the conductivity model of Mualem (1976a) which allows a direct determination of the hydraulic conductivity by numerical simulation. Up to now it was state of the art to determine the capillary tension by tensiometers and to determine the moisture with the θ/ψ -curve, yet the influence of the hysteresis does not allow distinct results. Therefore we propose other soil moisture measurement methods to derive soil hydraulic conductivity.

2.1.3.2 Moisture content

As with density there exist several different definitions of water content or moisture. The gravimetric water content can, similarly to the bulk density, be expressed on a dry mass, θ_{db} , or wet mass basis, θ_{wb} , and gives the ratio between the mass of the porewater and the mass of the dry solid substances resp. the solid substance plus mass of water. It can be expressed in percent units

$$\theta_{db} = \frac{m_w}{m_s} * 100 \quad (6)$$

$$\theta_{wb} = \frac{m_w}{m_s + m_w} * 100 \quad (7)$$

The volumetric water content can also be expressed on a volume basis as the ratio between the volume of water and the total volume of the soil sample:

$$\theta_{vb} = \frac{V_w}{V_t} * 100 \quad (8)$$

Since the soil water characteristic from the θ/ψ -curve especially for fine-grained soils is ambiguous, its transformation into a ψ/k_f -curve for the determination of the hydraulic conductivity is ambiguous too. Therefore, instead of the capillary tension, in the following the moisture content will be used as the relevant parameter for a more precise determination of the hydraulic conductivity.

For fine-grained soil samples, the wide distribution of void size means that the various pore water components play different roles. Determination of their relative amounts requires heating to different temperatures. The water in the large voids is lost at about 100 °C, the water in the fine capillaries at 105 °C or slightly more, while the hydration shell of interlayer cations in swellable clay minerals is lost at temperatures appreciably higher than 105 °C.

Although the determination of the water content by thermal analysis is a very accurate method and mostly used as a reference, this method is destructive and non-operational. But fortunately the range of possible soil moisture determination methods has increased

considerably since the beginning of the eighties of the last century. Especially the electromagnetic moisture measurement methods are promising new techniques for a reliable and in-situ determination of the soil moisture and thus of the soil moisture/hydraulic conductivity-relationship in the field. One of these new techniques will be presented in the following chapter.

2.2 Measuring methods

2.2.1 Physical methods

2.2.1.1 Bulk density

The methods available for the measurement of soil bulk density fall into two groups. In the first are the long established direct methods, which involve measurement of the sample mass and volume (core sampling, rubber ballon, sand replacement, clod). In the second group the attenuation or scattering of nuclear radiation by soil is used in conjunction with a calibration relationship to give an indirect measurement of bulk density (Mullins, 1991).

2.2.1.2 Soil texture (particle size distribution, grading curve)

The particle size distribution analysis is one of the most principle determinations in soil science and its knowledge already allows relatively good estimations of soil hydraulic properties. Grain size distribution is very important for the bulk density.

Principally with particle size distribution analysis there exist two problems: (1) from the wide range of particle sizes it follows that the analysis cannot be carried out by one single method alone and (2) since the particles show different stabilities it is nearly impossible to exactly distinguish between primary particles and aggregates, the latter being valid especially for fine grained materials such as clay and silt. Therefore a chemical and/or physical pre-treatment of the soil sample is indispensable to minimize aggregation of particles. The separation of the different particle sizes is carried out by sieving with exactly defined mesh sizes. The fine grained fraction, i.e. particle ranging from $<63 \mu\text{m}$ to $2 \mu\text{m}$, are normally separated by so called sedimentation analysis, using the different sinking properties of the grains in liquids (Stokes' law). This law is applied in several methods like aerometer according to Casagrande, Andreasen Pipete, Koehn Pipete, Atterberg or Kopecky, that yield the so called grading curve. This method reaches its limitations when the clay breaks up into fine, medium, and coarse clay. For determination of the clay fractions sedimentation is speed up by centrifugation applying several times of earth gravitational force.

All these methods are time consuming and determine size distribution of spherical equivalent particles as Stokes' law is based on the assumption of spherical particles. Faster methods of particle size distribution apply laser light scattering, X-ray absorption, acoustic spectrometry or dynamic light scattering. A systematic comparison of advantages and disadvantages in relation to particle size and particle shape was given by Latief (2010). The shape of platy clay minerals has a strong influence on their sedimentation behaviour and thus influences the determined equivalent particle size. Therefore, some methods allow introduction of a shape factor (e.g. Konert & Vanderberge, 1997) for calculation of particle size distribution.

2.2.1.3 Soil structure (pore distribution)

Beside the calculation of the porosity Φ from the density values, it can also be measured directly by an air pycnometer which is based on the Boyle-Mariotte law ($p_1V_1=p_2V_2$).

The relationship between the decrease of pressure (from p_1 to p_2) and the pore volume in the sample must be taken from calibration curves. This yields the air-filled pore volume V_A . To extract the total volume V , an additional water content determination to determine the volume of water V_W in the sample is necessary.

$$\Phi = \frac{(V_A + V_W)}{V} \quad (9)$$

2.2.1.4 Specific surface area

The determination of the specific surface area helps to identify expandable minerals and to estimate the degree of cementation of expandable clays. The measurement is carried out by determining the external and internal surface areas. The external surface area is measured by the adsorption of non-polar gases like nitrogen which cannot enter the interlamellar space. The internal surface is determined by measuring the total surface using water and subtracting from it the separately determined external surface (Pusch, 2002).

Water absorption capacity according to Enslin/Neff describes the property of soil at 60 °C dried up to weight constancy to absorb water by capillary action and to retain it. It mainly depends on content and type of the clay minerals. The Enslin-Neff values are known to be determined by the amount of exchangeable Na^+ and to a lesser extent by the smectite content (Neff, 1959). Hence the Enslin-Neff method provides an indirect estimate of the Na^+ content. Evaporation during the test has to be restrained as it is known to have a serious affect on the water-uptake capacity values measured by the Enslin-Neff device (Kaufhold & Dohrmann, 2008). Nevertheless, the test provides an index value which gives some indication of the size of the specific surface and activity of the fine grain.

2.2.2 Chemical and mineralogical methods

2.2.2.1 Phase analysis

Identification and quantification of mineral phases is performed by X-ray diffraction analysis and analysis of X-ray diffraction patterns from powdered samples by Rietveld method or pattern summation methods supported by complementary analyses (e.g. CEC, XRF, STA) (Omotoso et al., 2006, Emmerich, 2011).

- **X-Ray Diffraction Analysis**

X-ray diffractometry (XRD) is the standard analysis in mineralogy providing rapid information on clays and non-clay minerals present in a powder sample. An X-ray beam of defined wavelength (e.g. $\text{Cu K}\alpha$) is diffracted at the lattice of each mineral. According to Bragg's law the diffracted beam is commonly recorded at angles between 2-80° 2θ . The intensity ratio of peaks for each mineral depends on many factors (like chemical composition, preferred orientation and others, see e.g. Moore & Reynolds, 1997). Identification of the phases is made according to the JCPDS register (International Centre for Diffraction Data; JCPDS 1983) incorporated in commercial software. Identification of clay minerals often requires additional XRD analysis of so-called textured samples. The sample is slurried by appropriate chemical and/or physical treatment and the $>2 \mu\text{m}$ fraction separated in a centrifuge and discarded. The remaining suspension ($< 2 \mu\text{m}$) is placed as drops on a slide or sucked on top of a ceramic disc whereby the clay mineral orientates during the sedimentation and drying process more or less on a parallel basis (texture) (Moore & Reynolds, 1997). Subsequent solvation with ethylene glycol will identify the swellable clay mineral phases. Calcinating at 550°C destroys the existing kaolinite through

dehydroxilation allowing chlorite to be identified (<http://pubs.usgs.gov/of/2001/of01-041/htmldocs/flow/index.htm>).

- **Cation Exchange Capacity (Methylene Blue and Cu-Trien methods)**

Determination of cation exchange capacity (CEC) is performed by homoionic saturation of exchangeable positions by an Index cation (Dohrmann & Kaufhold, 2010) and indicates the amount of smectites in fine grained materials. Common index cations are methylene blue (MB), Ba^{2+} , NH_4^+ or copper triethylenetetramine (Cu-Trien). Despite MB is widely used (VDG P 69 1988) the method suffers certain restrictions that are discussed in detail by Kahr and Madsen (1995). If the CEC is determined by MB of a sodium exchanged material at neutral pH it would result in similar values as determined with the ammonium (NH_4^+) acetate method. MB can even be used for determination of SA if montmorillonite surface area per charge corresponds with the area of the MB of 130 \AA^2 , i.e. the interlayer charge of the montmorillonites must amount to 0.28-0.33 charges per half unit cell.

Occupation of exchangeable positions in natural state of clay containing is of particular significance for water adsorption, swelling and resulting microstructure and thus permeability of clays especially bentonites. Exchanged cations can be determined by AAS or ICP-OES from supernatant of CEC determination if precaution is taken to prevent dissolution of soluble minerals (Dohrmann & Kaufhold, 2010). According to Müller-Vonmoos & Kahr (1982) the cations bound to the exterior and interior surfaces of montmorillonite can be exchanged for an alcoholic ammonium rhodanide solution in the presence of soluble salts, too. The ion distribution of exchangeable bound ions (Ca^{2+} , Mg^{2+} , Na^+ , K^+) is of particular significance for water absorption and swell capacity, the microstructure and thus the water permeability of bentonite.

- **Swell Capacity Method**

The ASTM D 5890 method is used to determine the swell capacity of bentonite. Two grams of dried (105°C) pulverised sample is placed in 90 ml of de-ionised water in standard 100 ml glass cylinders. Of the sample, 0.1 g at a time is sprinkled into the water within 30 seconds until the whole amount has been used up. The cylinder is filled to 100 ml with de-ionised water and temperature is measured. After 24 h, the swell volume is determined in ml ignoring any flocculated material.

- **Fluid Loss Test**

The fluid loss test is a very reliable test regarding the permeability behaviour of bentonites. It enables the evaluation of fluid loss properties of a clay mineral film deposited on a filter paper from a 6% solids slurry of clay mineral at 100 psi (690 kPa) pressure as a measure of its usefulness for the permeability of hydraulic conductivity reduction. This method was adapted from American Petroleum Institute drilling fluid specifications for bentonite.

2.2.3 Dynamical methods

2.2.3.1 Matric potential

Namely the tensiometer and the pressure-membrane (or pressure-plate) apparatus, that either measure, or generate, the matric potential as defined above are used. These instruments measure the difference in pressure across a semipermeable 'membrane' in contact with the soil on one side and the soil solution (i.e. the equilibrium dialysate) on the other. The 'membrane' is permeable to solution but not to solids (Passioura 1980).

Principally the tensiometers are suitable for the determination of the hydraulic conductivity in the field and a measurement precision of $\pm 1.0 \text{ vol.-%}$ is principally

sufficient, yet there can occur big uncertainties when transforming the capillary tension into moisture or hydraulic conductivity due to described hysteresis effect. Also maintenance and calibration of the tensiometers especially in fine-grained soils are quite complex since they tend to run dry very quickly due to the high water suction of fine-grained soils. Also the measurements are rather punctual. However they can be used as a reference method for our purpose.

2.2.3.2 Moisture content

Since moisture content is a decisive criterion of many porous materials, there exists a broad variety of different methods. Generally one can distinguish between direct methods, where the moisture is determined directly by physical or chemical methods, or indirectly by determining a property that is mainly a function of the water content.

The most common direct method is the thermogravimetric method, where a sample of the soil is dried at 105°C to a constant weight. Other direct methods use e.g. calcium carbide, sulphuric acid, or phosphorus pentoxide, which react with the water present in the sample. Because all these methods require sampling and considerable laboratory equipment, their use for soil moisture measurement in the field is not very practicable. Here the indirect methods are more common and more differentiated using mostly physical parameters such as electrical, radiometric, acoustic or thermal soil properties. A good survey is given in Schmugge et al. (1980).

In this chapter we will focus especially on the electromagnetic methods that use the dielectric properties of the soil. The principle is based on a functional relationship between the dielectric permittivity of the soil and its volumetric water content. Different methods take advantage of the high relative permittivity of water ($\epsilon_r = 80$) compared to that of dry soil ($\epsilon_r = 3-5$). One of the most well known meanwhile is the Time Domain Reflectometry (TDR) (Topp et al. 1980), where the transit time t of an electromagnetic pulse on a wave guide of length l , which is buried in the soil, is measured. The relative permittivity ϵ_r is determined according to:

$$\epsilon_r = \left(\frac{c_0 t}{2l}\right)^2 \quad (10)$$

with c_0 as the velocity of light in free space. Relating the measured ϵ_r of different soil samples to the volumetric water content determined by the thermogravimetric method, allows to establish a so called calibration function. One of the most well-known calibration functions is the Topp-polynomial (Topp et al. 1980) which yields the volumetric water content θ_v according to:

$$\theta_v = -5.3 * 10^{-2} + 2.92 * 10^{-2} \epsilon_r - 5.5 * 10^{-4} \epsilon_r^2 + 4.3 * 10^{-6} \epsilon_r^3 \quad (11)$$

Conventional TDR-sensors are normally fork-like metallic wave guides of several tens of centimetres that penetrate the soil, giving a rather punctual measurement. Yet the determination of hydraulic conductivity on a field-scale basis based on soil moisture measurement requires more large-scale sensors why a flat-band-like TDR-cable sensor called TAUPE has been developed (Brandelik & Huebner, 1999). Due to a plastic coating of the copper wave guides this sensor is capable of sensing up to 30 m of the surrounding soil. Both an integral soil moisture value and a moisture profiling along the length of the buried cable according to a new TDR inversion technique can be accomplished (Schlaeger, 2005).

3. Processes

3.1 Dynamic of the surface moistening

The moistening of surfaces obeys certain natural laws which were established during the last 200 years. The development was started in the beginning of the 19th century by several scientists from the fields of physics and chemistry. In this chapter we will exemplify some important laws with respect to surface moistening.

3.1.1 Laws

3.1.1.1 Young-laplace equation

In 1805 Thomas Young and Pierre-Simon Laplace both described independently from one another a fundamental equation with respect to interface science. The Young-Laplace equation describes the correlation between surface tension, pressure and surface curvature of a system consisting of two phases. Such a system could be e.g. a liquid drop on a solid surface or a liquid in another immiscible liquid. Surface tension of a liquid results from attractive interaction of the liquid molecules. A molecule located within a liquid is surrounded by other molecules, so the resultant force is zero. This does not apply to a molecule at the surface, since a part of interaction is missing at this place. The molecule is bordered by air molecules on the upper side and these intermolecular forces are of weak nature. This leads to an inward looking force. The energy required to overcome this force is the surface tension, sometimes also called surface energy.

3.1.1.2 Lucas-Washburn & modified Lucas-Washburn

The predefined aim of the studies of Lucas (1918) and Washburn (1921) was to develop a theoretically established law, which determined the capillary head existing in an arbitrary capillary system, as a function of time. The first approach was to immerse a wettable cylindrical tube vertically into a solution. The surface tension of the liquid becomes noticeable as the length of the cross section ($2 \cdot r \cdot \pi$) multiplied by the surface tension (σ), perpendicular to the direction of the tube. This force elevates the liquid to a height where it is equilibrated by the gravity.

$$2r\pi\sigma = r^2\pi m_s h_0 \quad (12)$$

where h_0 is the maximum pressure head and m_s the specific mass. Thus the maximum height entirely depends on the surface tension, on the radius of the tube and on the specific mass of the solution measured. The penetration speed of the liquid due to the pulling force diminishes with the height because the mass of the liquid increases. Moreover the rise of the liquid is slower the tougher the liquid is. After the viscosity of the solution has been taken into account (Poiseuille) and assuming that wetting isn't complete, the Lucas-Washburn equation for the capillary rise is

$$h^2 = \frac{r\sigma \cos \theta t}{2\eta} \quad (13)$$

Here θ is the contact angle, t is the time and η is the viscosity. The contact angle is between 0° and 90° , so $\cos(\theta)$ lies between 0 and 1.

3.1.2 Methods

3.1.2.1 Contact angle measurement

The basis of the contact angle measurement goes back to Thomas Young (1805), who related the contact angle to the surface tension:

$$\cos \theta = \frac{\sigma_s - \sigma_{ls}}{\sigma_l} \quad (14)$$

θ is the contact angle, σ_s the surface free energy, σ_{ls} the solid-liquid surface energy and σ_l is the surface tension of the liquid. We can distinguish between three cases relating to the contact angle. If $\theta < 90^\circ$, the liquid wets the solid surface, if $\theta > 90^\circ$, the sample is hydrophobic and the liquid doesn't wet it or wets it only partially and if $\theta = 0^\circ$, the solid surface is totally wettable, the liquid spreads over the surface.

In practice the liquid drop is put on the solid surface, which has to be as straight as possible, plane and also clean. A light source, which is positioned in the rear lets the drop appear dark. θ can be measured directly using a goniometer or with the help of an optical calculating system which employs the equation of Young-Laplace. The goniometer measuring leads to a relatively large error ($\pm 2\%$) and is not applicable for small angles and irregular contact lines (Dimitrov et al., 1991). In case of small drops the hydrostatic effects can be neglected and the contact angle can be calculated from the height of the drop (Butt et al., 2006).

3.1.2.2 Dynamic contact angle measurement

The processes happening at the solid-liquid interface during wetting and dewetting are best described by the dynamic contact angle. The interface at the contact between liquid drop and solid surface doesn't appear suddenly, but it needs a certain time until a dynamic equilibrium is reached. In practice the measuring of the dynamic contact angle works in the way that a liquid drop is spread on the solid surface and then extended by means of a needle. The solid-liquid interface migrates outwards and the contact angle can be measured by defining certain degrees steps. Studebaker & Snow (1955) developed an equation for the determination of the dynamic contact angle.

$$\cos \theta_2 = \frac{\sigma_1 \eta_2 t_1}{\sigma_2 \eta_1 t_2} \quad (15)$$

The authors determined dynamic contact angles of powder samples by measuring the time required for a liquid to imbibe the powder bed. This time was then compared to a reference sample with $\cos \theta = 1$ (contact angle = 0°). This method assumes that the differences in the penetration rate are due only to differences in contact angle, after taking surface tension and viscosity into account (Yang & Zografis, 1986).

3.1.2.3 Capillary rise method

Jones & Ray (1937) investigated the determination of the surface tension of water and several salt solutions. They developed a differential method to determine this property of liquids. The experimental set-up of the capillary rise method consists of a tight cylindrical tube and a broad tube being connected with each other. The vertical level difference between the meniscus in the tight tube and the extended one has to be measured. The density of the liquid, which also needed, may be determined directly by the use of a hydrometer.

$$\sigma = \frac{r h g (D - \beta)}{2} \cos \theta \quad (16)$$

r is the radius of the tight tube measured at the height of the meniscus, h the capillary rise, g the acceleration of free fall, D the true density of the liquid, β the density of the gas phase (air plus water vapor) at the temperature and the barometric pressure when the experiment is made and θ is the contact angle. θ should be zero in glass and silica tubes if the tubes are clean (Jones & Ray, 1937). First the elevation between the lowest levels of the menisci must be read off to get the approximate value of the capillary rise. This value has to be corrected for the liquid by means of the Rayleigh formula. Jones & Frizzell (1940) for their part examined the influence of the concentration of the solution on the capillary rise. Therefore they used diluted salt solutions of different concentrations. The most noticeable feature of the results was that the penetration height of water was higher than those of the diluted solutions. This was interpreted as an evidence for a higher surface tension of water compared to the salt solutions. Measurements based on Washburn's equation do not only depend on the particle size but also on the pore size distribution. Addition of fine particles to the measured bed increases the penetration rate of liquid and improves precision of the measurement (Dang-Vu & Hupka, 2005).

3.1.2.4 Wilhelmy-Plate

The Wilhelmy-Plate method is utilized to determine the surface tension of a liquid. It can also be used in order to study the contact angle during capillary rise. In doing so a plate is contacted with the surface of the examined liquid where a meniscus forms at the contact point of the two phases. Due to this meniscus a force between the phases appears which originates from the wetting. By pulling the plate upwards a force (surface tension) manifests itself.

3.1.2.5 Sessile drop

The interface science makes use of different methods with regard to measure both, properties of liquids (e.g. surface tension) and properties of solids (e.g. static/dynamic contact angle or surface energy). The sessile drop method is an example for a measurement on a solid. For this purpose a drop of liquid (in most of the cases a reference solution is used) is spread on a solid surface and the static contact angle of the liquid is measured optically. Bachmann et al. (2000) developed a sessile drop method by modifying Young's equation on two points, since it is strictly applicable only to completely uniform and plain surfaces:

- i. A correction factor was introduced, which is defined as the ratio between the actual and the apparent area. This leads to the equation of Wenzel

$$\cos(\theta_{obs}) = r \cos(\theta) \quad (17)$$

Eq. (17) was developed due to the fact that the observed contact angle is smaller than the ideal (intrinsic) angle as long as this is below 90° and larger if the intrinsic angle is above 90° . So, the precision of the contact angle measurement therefore depends on the magnitude of it.

- ii. The Cassie-Equation is considered as an empirical approach describing the apparent contact angle on a chemically heterogeneous surface

$$\cos(\theta_{obs}) = f_1 \cos(\theta_1) + f_2 \cos(\theta_2) \quad (18)$$

3.2 Fluid flow

3.2.1 Hydrology

According to the physical law of conservation of mass the equation of water balance is valid. The input precipitation (N) is equal to the sum of the current total evaporation (Haude, 1958) ET_a (evaporation of soil, transpiration of plants and evaporation of interception), the sum of runoff (surface runoff Q_O , lateral runoff Q_L and leaching Q_V) and change of water content in soil layers within the observation period ($\Delta\theta = W_A - W_E$).

$$N = ET_a + (Q_O + Q_L + Q_V) + (W_A - W_E) \quad (19)$$

The description of water balance in vertical soil profiles is the fundamental requirement for the description of water balance in areas. Numerous investigations on the regionalization of point methods of measurement nowadays provide well-founded transmission options. The following sections consider the water balance of a vertical profile.

3.2.2 Analytical basics

The major focus of the investigation lies on the flow of a solution (in this case water) through the soil matrix. The water flow not only underlies gravity, but is also influenced by the soil properties (Hillel, 1980). Driving forces like gravity (hydrostatic forces), adsorption, cohesion, osmotic forces due to dissolved salts etc. cause water movement through their resultant (Czurda, 1994). Concerning the water movement we distinguish between advective flow and diffusion. Diffusion is irrelevant in the case of materials owing high hydraulic conductivity. The mathematical expression of the potential can be used to describe the flow of water through soil. It should be noted that the theory of the potentials considers neither the geometry of the pore space nor the mechanisms of water binding. These are included in the matric potential. The mechanical energy is taken into account but not the thermal energy. The stationary flow within the unsaturated zone is described by Darcy/Buckingham (equation of motion). Darcy's law features flow by means of a unit volume in dependency of the hydraulic conductivity of soil and in dependency of a potential gradient. The flow velocity depends on the hydraulic conductivity (k_f -value) of the soil and on the total potential (Ψ). k_f depends on the water content θ .

$$v_x = -k_{fx}(\theta) \cdot \frac{\partial \Psi}{\partial x} \quad v_y = -k_{fy}(\theta) \cdot \frac{\partial \Psi}{\partial y} \quad v_z = -k_{fz}(\theta) \cdot \frac{\partial \Psi}{\partial z} \quad (20)$$

k_f does not vary linearly depending on water content, but it follows a relationship which is characteristic to each soil. The hydraulic conductivity decreases with the square of the capillary radius. The air in the soil is considered to be stationary. In case of transient conditions Darcy's law (equation of motion) is combined with the equation of continuity (validity of conservation of mass).

$$\frac{\partial v_x}{\partial x} + \frac{\partial v_y}{\partial y} + \frac{\partial v_z}{\partial z} = -\frac{\partial \theta}{\partial t} - S \quad (21)$$

S represents a term containing a sink or a source. Combining (20) with (21) considers the change in water content during water flow. This equation is known as the partial differential equation of unsaturated flow (unit s^{-1}) within the soil matrix.

$$\frac{\partial \Psi}{\partial t} \cdot C + S = \frac{\partial}{\partial x} \left[k_{fx}(\theta) \cdot \frac{\partial \Psi}{\partial x} \right] + \frac{\partial}{\partial y} \left[k_{fy}(\theta) \cdot \frac{\partial \Psi}{\partial y} \right] + \frac{\partial}{\partial z} \left[k_{fz}(\theta) \cdot \frac{\partial \Psi}{\partial z} \right] \quad (22)$$

$C (=d\theta/d\psi)$ is defined as specific moisture capacity of the soil. Richard's equation contains the relationship between water content (θ) and soil water tension (Ψ , set equal to the potential) and also the relationship between hydraulic conductivity and water content. These relationships are extremely non-linear. Therefore, the solution of equation (24) (calculation of water and solute transport) requires numerical methods (Jentsch, 1992; Philip et al., 1974). It is foreseeable that we need to know two parameters in order to describe the water movement within the soil: total potential (Ψ , represented by the soil water tension under described boundary conditions) and water content.

3.2.3 Soil mechanics and soil hydraulics

Soils store and transport water within their pore system. This property is determined by the hydraulic conductivity and the texture of the pores. The hydraulic conductivity depends on the water content of the soil. Considering an unsaturated "ideal" soil with a uniform microstructure without macropores the following relationships are relevant:

- ratio volumetric water content to soil water tension also called the soil moisture characteristic or pF-curve
- ratio soil water retention to hydraulic conductivity
- ratio volumetric water content to hydraulic conductivity

The soil water tension equals to the sum of bonding forces performed by the soil matrix with regard to the water. The pF-curve shows a characteristic course for each different soil (Scheffer, 1992). It depends on the particle size distribution, the formation and cross-linking of the pores and also of their size. The proportion of organic matter and the chemical composition of the wetting phase also affect the pF-curve. The water holding capacity of sand is low (if $pF > 4.2$) the residual saturation θ_r equals a water content less than 3 vol. %), so sand releases water situated within the pore volume at low soil water tension differences (low specific water capacity C , that means a slight gradient of the tangent with respect to the pF-water content-curve at $0 < pF < 2.5$). In contrast, clay shows a high residual saturation (if $pF = 4.2$, water content equals ~ 30 vol. %) and the release of water needs high changes of the soil water tension (high specific water capacity C , that means high gradient).

Depending on the history of the soil with respect to watering and dewatering the shapes of the pF-curve for a single soil are different, an effect called hysteresis. It is assumed that the capillaries which connect the pores show a smaller cross section than the pores themselves. If the soil water tension is high, the convex meniscus holds on to the upper border of the pore (by means of retention forces). This soil water tension is higher than necessary in order to move the concave meniscus (formed by wetting resistance) into the capillary in case of watering. The main influencing factors of the pF-curve are grain size, soil structure and hysteresis.

There are manifold approaches for the analytical description of the pF-curve. The approach of Brooks & Corey (1964) combines the mathematical ratio of θ to Ψ with the conductivity model of Burdine (1953). The definition of effective water content, also called relative saturation index or soil-water-retention, is

$$S_e = \frac{\theta - \theta_r}{\theta_s - \theta_r} \quad (23)$$

with θ as current water content, θ_r as residual water content (simplified = 0, easier to measure at pF = 4,2) and θ_s as water content at saturation (measured at pF = 0), conform to porosity.

Besides the approach of Campbell (1974) the common approach originates from Van Genuchten (1980) and Mualem (1976a), VGM in the following. The VGM approach prevailed in the literature and will be considered subsequently. Therefore we combine the (Ψ)-relationship with the conductivity model from Mualem. Unlike the model of Brooks & Corey-Burdine (Berger, 1998) this approach doesn't take any sharp air inlet into account. Thus this approach is solvable from the analytical point of view if we consider certain boundary conditions

$$S_e = \frac{\theta - \theta_r}{\theta_s - \theta_r} = \left[\frac{1}{1 + (\alpha h)^n} \right]^m \quad (24)$$

α, m, n parameters of shape, dependent on grain distribution of the soil:

$\alpha = 1/h_b$; h_b = soil water tension (ψ) at air inlet point,

$m = \lambda/(\lambda+1)$; λ = index of pore size,

$n = \lambda+1$; hence $m = 1-1/n$

h soil water tension (ψ) at the water content θ

So the description of the hydraulic conductivity/soil moisture-relationship becomes possible from the analytical point of view based on the prediction model of Mualem:

$$\frac{k_f(\theta)}{k_s} = S_e^\gamma \cdot \left[1 - \left(1 - S_e^{\frac{1}{m}} \right)^m \right]^2 \quad (25)$$

$k_f(\theta)$ hydraulic conductivity at the current water content

k_s hydraulic conductivity at water saturation. k_s is often set constant, but also depends on the texture of the soil

γ takes the influence of tortuosity into account, usually $\gamma = 0.5$.

By means of inverse identification of parameters (Schultze et al., 1996) the approximation of the values from numerical simulations to those from field trials is possible. Thus the sought parameters are provided directly. The measuring of water tension by means of tensiometer represented until now the state-of-the-art. The water content of the soil was determined via pF-curve (with knowledge of the water tension) and furthermore the k_f -value. Until now no exact results could be achieved due to the influence of hysteresis.

3.2.4 Soil moisture

In order to solve the Richards equation (22), knowledge of properties of the soil is required. Until now these properties were taken from the fundamental pF-curve. There the pF-value represents the common logarithm taken from Buckingham's "capillary potential" expressed as cm of the water column. The shape and uniqueness of the pF-curve depends on the texture. The texture includes pore size distribution (structure) as well as storage and arrangement of the particles (texture). For analytical modeling further parameters are

necessary. These parameters describe the water content of the soil at different boundary conditions. Alongside the natural saturation water content (θ_s) where $pF = 0$, the absolute saturation (θ_s^*) is also an important parameter. θ_s^* correlates to the porosity of the soil. Absolute saturation cannot be achieved by rewatering because -according to structure and texture- certain parts of the pores remain air-filled. According to extensive investigations the following equation is valid

$$\theta_s = 0.8 \div 0.95 \theta_s^* \quad (26)$$

Thus soil is liable to be seen as three-phase system. A further parameter is the remaining water or residual saturation defined as θ_r , where the aqueous phase is not constant anymore. The associated pF -value is 4.2. The distribution of the three components water, air and soil is described by various "mixing models". The aqueous phase is distinguished between "free water" and "bound water" that is adsorbed on or within the particles. The electrostatic forces surrounding the solid (here: soil particle) act outwards. These forces result from molecules that are not compensated electrical all-round. The wetting property of a solid with regard to water depends on the strength of these forces. If the cohesion forces of the water molecules are less than the surface forces, the water molecules absorb on the surface. Bound water is able to absorb further water molecules by means of associate forces, however, this binding is not stable (Huebner, 1999).

Bound water prefers ionic bonds. The surfaces of fine-grained materials such as clays are saturated by ions. The sorption forces between ions and the surface of the clays are greater than the non-polar sorption forces between surface and water molecule. As a result the wetting property of clays increases and a hydrate envelope around the metal cations is established. Also crystal water, i.e. water bound within the lattice of the soil particles, is present and water can condensate within the capillaries. If two water films get into touch, the water molecules flow together and form carrying menisci within the soil pores and more water molecules are attracted by the surface tension. If the soil air is saturated with vapor, the water condenses above the concave meniscus. At this place the vapor pressure is smaller than above the convex or the flat meniscus. The molecular forces get saturated by steam or by liquid water.

The main part of the water in the soil is not influenced by molecular forces. It has zero potential and underlies gravity. The water contents θ , θ_s and θ_r have been determined in on lab-scale depending on soil water tension. Because of the hysteresis it was distinguished between the watering and dewatering of the sample. The reasons for the hysteresis of the pF -curve are that the advancing contact angle between soil matrix and soil water is greater than the retreating contact angle, the effects regarding the geometry of pores, water bound on clay mineral surfaces, and enclosed air. Statements leading to a reliable approximation of the hydraulic conductivity are therefore impossible. The extension of uncertainty by 70 times results from empiric measures (Ψ is applied logarithmic, k_f is applied exponential). This uncertainty provides the basis for the assessment of the water content (θ) as a relevant measurement parameter.

4. Systems

The comprehensive characterization of materials is prerequisite to understand processes in large (geo)-technical systems and their manipulation. This can be achieved best by the knowledge of material properties, measuring methods to determine water content and

processes that describe the interaction of matter and water. Examples for technical systems in that sense are e.g. sealing systems for landfills and subsurface storage of waste, monitoring of soil water content over large areas using power lines, or monitoring system for groundwater recharge in the unsaturated zone.

4.1 Monitoring system for surface sealings

4.1.1 Configuration of sealing system and monitoring layer

For the monitoring of the volumetric water content θ_v , the TAUPE TDR-system described in chapter 1.2.3.2 was used. Specifications for a monitoring system for surface sealing systems on landfills (figure 1) defined from legislating body (BAM, Federal Institute for Materials Research and Testing) are

- detection of increase over more than an order of magnitude in permeability of a mineral sealing or capillary barrier,
- detection of local relative variations in volumetric water content of more than 5 %,
- positioning information of 100 m², that means a circle with radius around 5.5 m around true position.

The sealing system installed at the landfill is build up as a capillary barrier as shown in figure 2.

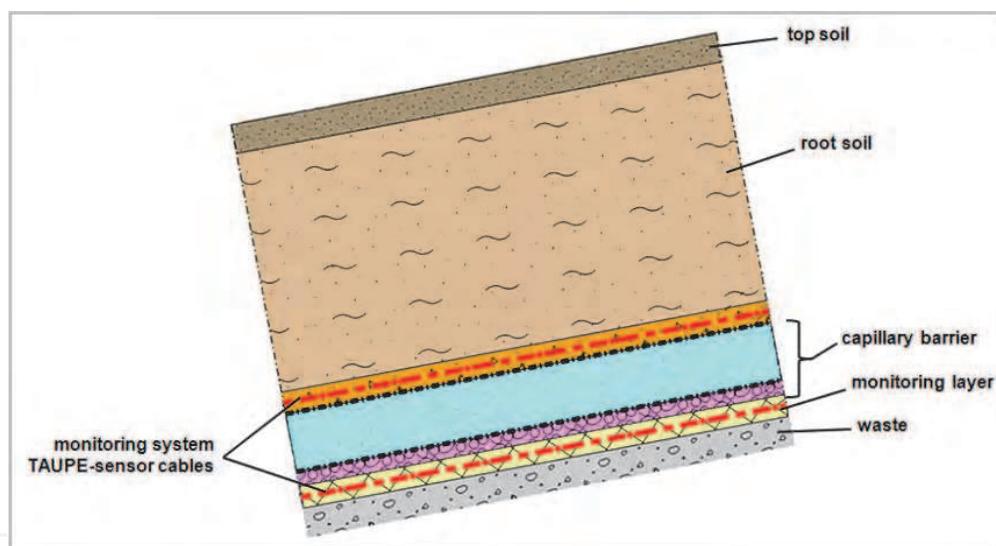


Fig. 2. Simplified schematic of mineral capillary barrier consisting of a fine-grained material capillary layer above a coarse-grained material capillary block in a surface sealing system of a landfill.

Sensors of the monitoring system were installed in monitoring layers above and below the capillary barrier to determine the quantity of permeated water through the barrier. Water content of a soil layer situated below the capillary barrier without appreciable hysteresis is directly related to its permeability, i.e. the unsaturated hydraulic conductivity. Landfill regulations determine saturated hydraulic conductivity as quality parameter of mineral sealing systems. A critical limit of $< 5 \times 10^{-9}$ m/sec can be deduced from experimental relation between volumetric water content and unsaturated hydraulic conductivity, which can reach values of a few orders of magnitude below that limit. Materials used in a monitoring layer must meet the following requirements

- its hydraulic properties must be defined unambiguously,
- inflow from layers above must lead to a significant change in volumetric water content,
- uniquely defined monitoring parameters for saturated/unsaturated hydraulic conductivity correspond to definite volumetric water contents. Percolation through monitoring layer must be equivalent to percolation in total sealing system.

Adequate for a monitoring layer are sandy to silty materials with a range of saturated hydraulic conductivity of 10^{-5} to 10^{-6} m/sec. A decrease in unsaturated hydraulic conductivity should be at least four to five orders of magnitude and cover the range of 10^{-10} m/sec to 10^{-5} m/sec. The monitoring layer then can discharge a break-through of the sealing system without building up backwater. Sensors are installed in monitoring layers at depth of around 2 m and 2.3 m, respectively.

4.1.2 Calibration of a material for the monitoring layer

An adequate material was tested in the laboratory using measurement equipment for the determination of complex dielectric permittivity. It consists of a vectorial network analyzer and a coaxial type probe cell (see inset in figure 2) in the same frequency range of 100 MHz to 1100 MHz as used with TDR method. The material has been exposed to different amounts of water, with a part of it being dried at 105 °C in oven for evaluation of gravimetric water content related to dry mass. A second part was inserted in the coaxial probe cylinder for determination of permittivity. Figure 3 shows the relation between volumetric water content and square root of measured permittivity. Results from TDR measurement can directly be converted via the indicated regression function.

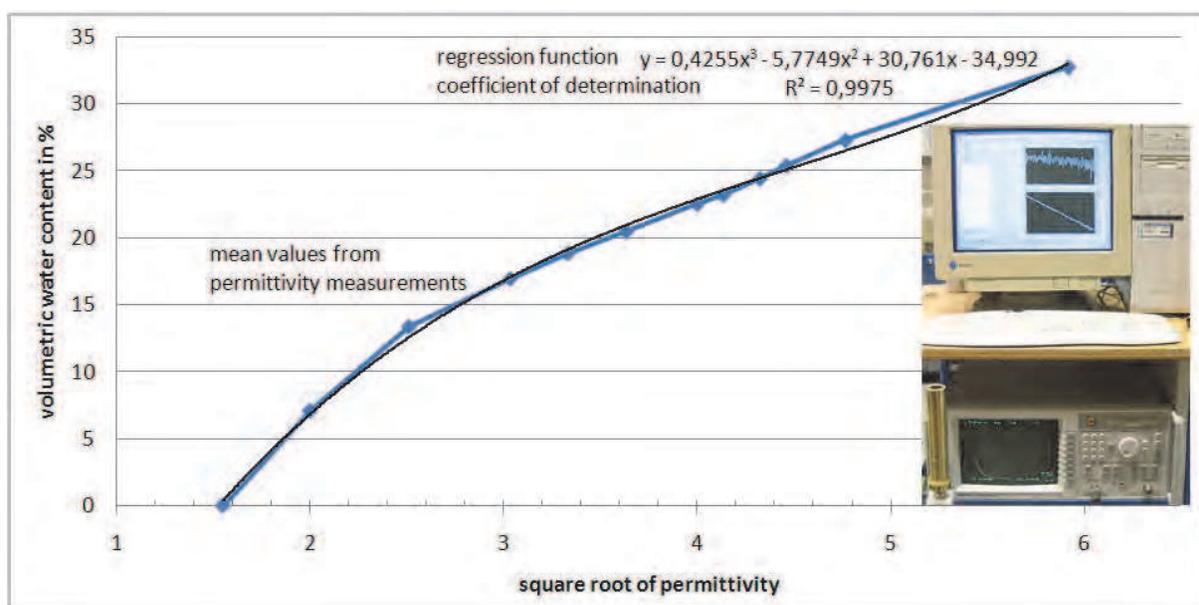


Fig. 3. Material calibration function for the monitoring layer. Inset shows the permittivity measurement system with coaxial probe cylinder.

4.1.3 Monitoring system

A test site for a monitoring system for surface sealing has been established from 2004 to 2005 at the landfill situated in Oberweier/Germany (Figure 4). In two monitoring layers (see figure 1) 230 sensors have been installed and connected via 34 multiplexer to a TDR system.

To keep the length of the connecting coaxial cables between TDR system and sensors below 150 m, two central units with separate TDR devices cover 120 and 110 sensors, respectively. Sensor length is 10 m and distances between adjacent sensors are between 8 and 10 m, depending on hill slope. Data collection takes place two times a day.

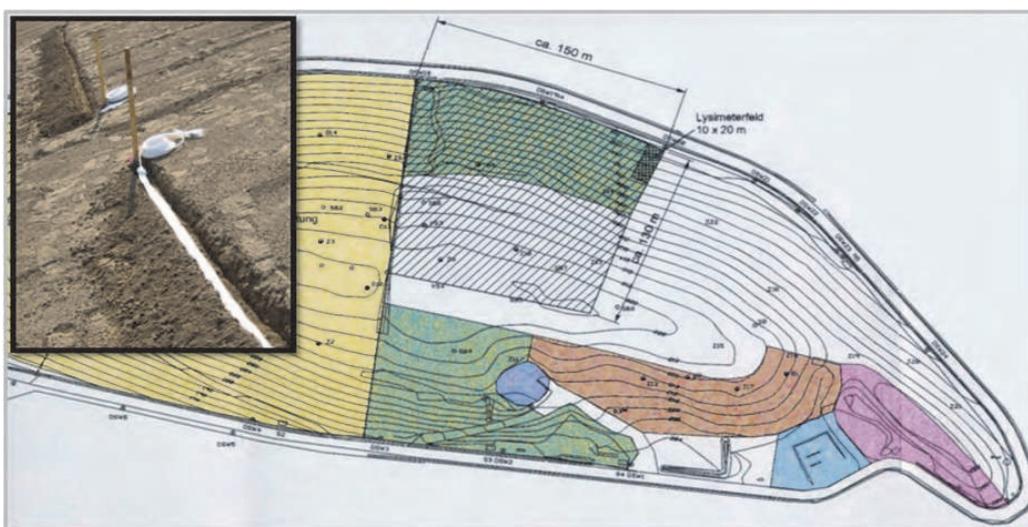


Fig. 4. Plan of landfill in Oberweier. Sensors and measurement equipment has been installed in the shaded area of approximately 150 m x 130 m. Inset shows TAUPE TDR-cable sensors during installation in lower monitoring layer.

4.2 Data evaluation

TDR signal data comprise only a part of the total signal length and is constraint to the transition between coaxial cable and start of sensor and shortly beyond end of sensor (see figure 5). First rise of the reflection signal occurs at start of sensor and second rise at end of sensor (Topp et al., 1980). Exact starting and ending points are defined by calculating the inflection points of the slopes to fit tangents to the curve and finding crossing points with horizontal lines. From time difference propagation time is calculated.

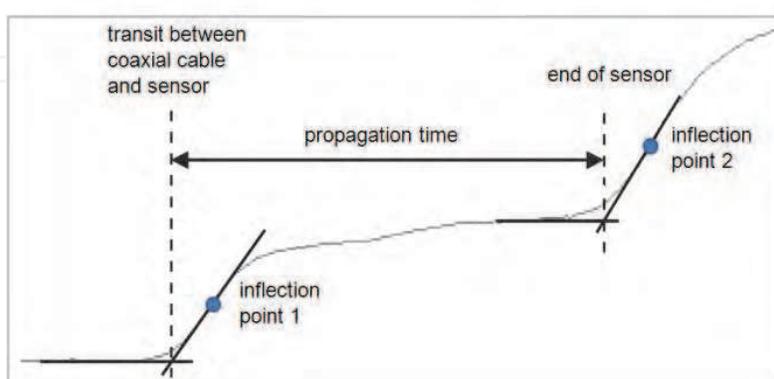


Fig. 5. Typical form of TDR reflection signal and determination of propagation time

Data from all sensors are automatically evaluated using an appropriate software system. Resulting propagation time defines an integral value for volumetric water content along a complete sensor according to the material calibration function in figure 2. Adding results for

each sensor over time delivers variations in water content all over the landfill. To give an easier access to the hydraulic behavior at locations of different sensors the landfill is divided in vertical transects between top of the landfill and its base. This is shown for 2010 in figure 6 on eight sensors for both monitoring layers.

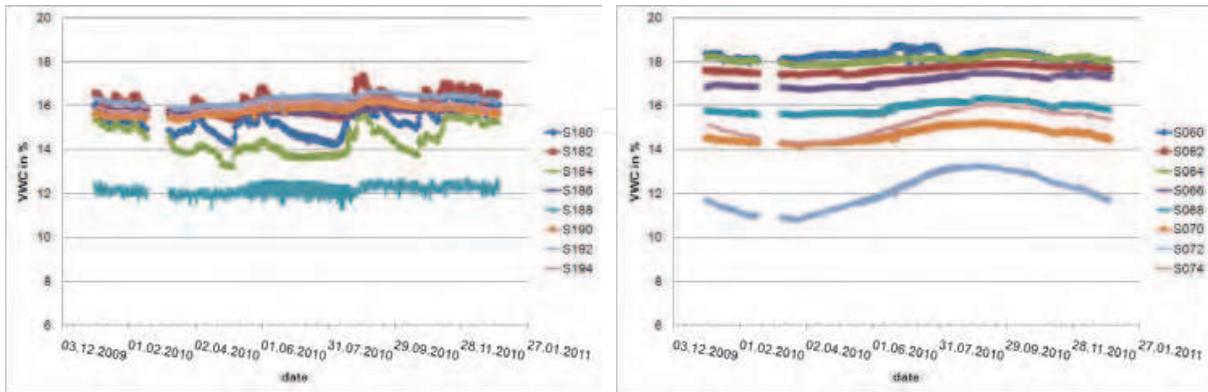


Fig. 6. Volumetric water content as a function of time in monitoring layer above (left: sensors 180, 182, 184, 186, 188, 190, 192, 194) and below (right: sensors 60, 62, 64, 66, 68, 70, 72, 74) capillary barrier. Data gap in March 2010 was due to a failure in power supply.

In the monitoring layer above capillary barrier discharge of precipitation at the surface can be found directly, especially at the borders of the landfill due to a poor connection to the area outside of the test site. Preferential flow parallel to the slope in layers with local secondary capillary barriers, which have been built unintentionally during installation process, leads to a more steady value of volumetric water content. Normally short time reactions describe the hydraulic system above the sealing.



Fig. 7. Intrinsic situation of volumetric water content in monitoring layer above (left) and below sealing system (right). Vertical sections according to figure 6.

In contrast, situation below the sealing system shows little short time variations, what demonstrates the functionality of the capillary barrier. Except at the borders little influences of discharge from the surface occur. Depending on chemical reactions in waste and seasonal temperature changes the volumetric water content can vary locally in the order of up to 2 % due to temperature depending permittivity of water.

Graphs in figure 7 show the situation above and below the capillary barrier at a certain time as colored graphs. Rectangular blocks show the volumetric water content of each sensor and

the red color indicates possible problems due to locally high water content above arbitrarily chosen 18 %. Situation below the sealing shows more dark blue and red spots than above, which is a result of the installation since the first section below the sealing has been constructed during heavy rain in October 2004 and the second section with the capillary barrier was built in spring 2005 during the dry season. Water exchange with atmosphere via evapotranspiration is low due to depth of monitoring layers.

4.2.1 Hydraulic conductivity

Volumetric water content of the layer below capillary sealing received from the monitoring system is the input parameter for the determination of hydraulic conductivity according to figure 8. In 2010 the sensors detected volumetric water contents between 12 and 18 %. Critical limit of 5×10^{-9} m/sec gives a monitoring value for volumetric water contents of 22 % and has not emerged during the observation period between 2005 and 2010.

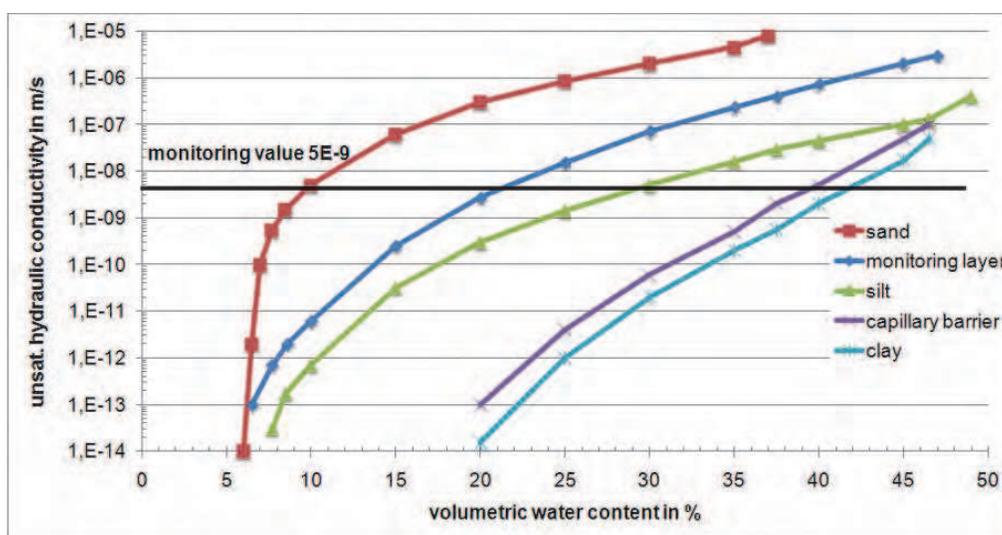


Fig. 8. Hydraulic conductivity of different materials as function of VWC; material of monitoring layers fulfills requirements of BAM.

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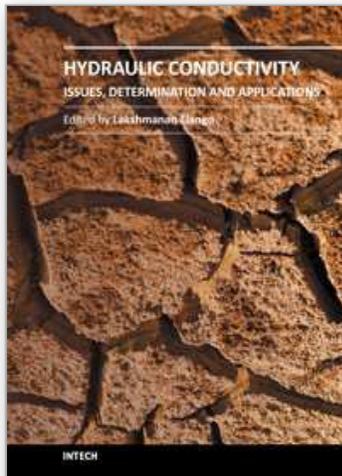
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There are several books on broad aspects of hydrogeology, groundwater hydrology and geohydrology, which do not discuss in detail on the intrigues of hydraulic conductivity elaborately. However, this book on Hydraulic Conductivity presents comprehensive reviews of new measurements and numerical techniques for estimating hydraulic conductivity. This is achieved by the chapters written by various experts in this field of research into a number of clustered themes covering different aspects of hydraulic conductivity. The sections in the book are: Hydraulic conductivity and its importance, Hydraulic conductivity and plant systems, Determination by mathematical and laboratory methods, Determination by field techniques and Modelling and hydraulic conductivity. Each of these sections of the book includes chapters highlighting the salient aspects and most of these chapters explain the facts with the help of some case studies. Thus this book has a good mix of chapters dealing with various and vital aspects of hydraulic conductivity from various authors of different countries.

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