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## DETERMINATION OF POLLUTANT TRANSPORT PARAMETERS BY LABORATORY TESTING

## DETERMINATION PAR ESSAIS EN LABORATOIRE DES PARAMETRES DE DEPLACEMENT DES CONTAMINANTS

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### SYNOPSIS

The paper gives an overview of the research program on pollutant transport within environmental geotechnics that is currently being executed at the Institute for Soil Mechanics and Foundation Engineering of the Ruhr-University Bochum. It concerns a new laboratory testing method for determining the parameters of diffusive and sorptive pollutant transport. In this research project, supported by the German "Federal Minister for Research and Technology", the different effects of various pollutants on defined mineral liners are pursued and shown by emission-graphs. Based on computer aided interpretation of the test results the characteristic parameters of diffusion and adsorption can be determined. In this paper test data for potassium transport are presented and discussed. The results of the tests can be used to evaluate mineral liner materials and systems of sufficient chemical resistance to prevent pollutant emission and hazardous material degradation.

### INTRODUCTION

Main sphere of environmental geotechnology is concentrated on planning and constructing of landfills according to regulations which assign central importance to the prevention of pollutant emission into groundwater and air. The actual regulations for basal lining systems demand a multi-barrier-system of various components each of them operating as a redundant to the others. In case of failure of all other barriers a mineral liner is preserved to retent hazardous substances as last technical barrier. Within the last decade increasing safety regulations demand for mineral liners permeability coefficients less than  $5 \times 10^{-10}$  m/sec. [1]. In this case diffusive mass transport is more important than convective transport.

However, while standard tests are available for the determination of the hydraulic conductivity coefficient, only few testing methods have been developed which enable geotechnical engineers to measure diffusion parameters.

An important role plays the mechanism of adsorption, a term which involves different mechanisms like ion exchange capacity, precipitation or crystallization, that hinder pollutant emissions. This part is more difficult to determine as it depends on mineralogy and chemistry of the soil and varies in a broad range. Nevertheless it is necessary to consider sorption effects as they contribute towards the prevention of pollutant emissions. Especially smectite type clays can technically be adapted against certain chemical attacks which appear propable in monodumps.

As only little knowledge of the pollutant transport phenomena was noticed experts from various scientific disciplines worked together with the Bochum Institute for Soil Mechanics and Foundation Engineering and discussed new testing methods.

Theoretical principles to describe the mass transport phenomena have been presented by hydrogeologists and soilphysicists. A very useful assessment is laid down by the fundamental transport equation derived by Van Genuchten [2]. The equation indicates the concentration alteration of substances as a function of time:

$$D \cdot \frac{\delta^2 c}{\delta z^2} = V \cdot \frac{\delta c}{\delta z} + \frac{\delta c}{\delta t} + \frac{1}{a} \frac{\delta n}{\delta t}$$

$c$  = concentration of adsorbate in the fluid stream, moles per unit volume of solution

$n$  = amount of adsorbate on the adsorbent, moles per unit volume of packed bed

$V$  = velocity of fluid through interstices of the bed

$z$  = distance variable along the bed

$D$  = diffusion coefficient of the adsorbate in solution in the bed

$a$  = fractional void volume in the bed

In the laboratory test methods were evolved. In a first step a new apparatus was developed, which can be used to determine the migration resistance within water-saturated soils during a stationary phase [3].

Based on this starting point an advanced test equipment was developed in cooperation with Dywidag Environmentals, Munich, to investigate diffusion and sorption parameters, the so-called DKS-permeameter (Diffusion, Konvektion, Sorption) [4], shown in figure 1.

## DKS-PERMEAMETER

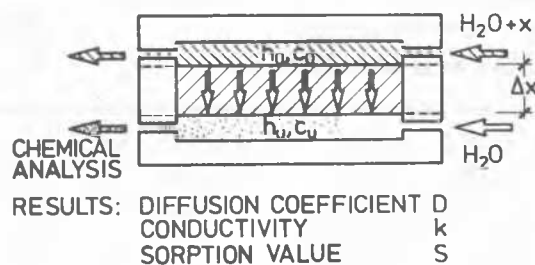


Figure 1: DKS-permeameter

A soil sample is placed in the centre of the permeameter, between the haze plate and the cap in a space of 80 mm x 80 mm x 20 mm. A flow of a solution with a constant contaminant concentration ( $H_2O + x$ ) parallel to the surface of the soil sample is induced, while clean water ( $H_2O$ ) is flowed past the opposite side of the sample. Using this arrangement, one obtains a constant concentration gradient through the sample and diffusion results. All parts of the permeameter coming in contact with the experimental solutions are made of PE, PVDF or Viton. These resistant materials do not have interactions with the considered solutions. Figure 2 shows the flow of solutions within the experimental kite.

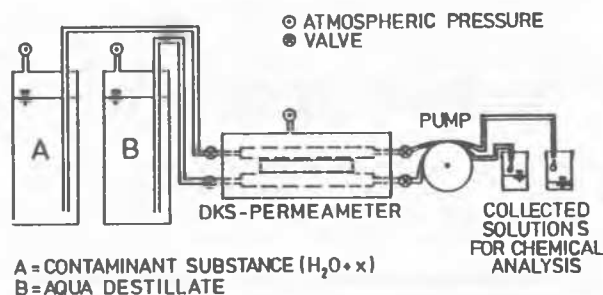


Figure 2: Layout of the DKS-permeameter system.

A liquid reservoir is filled with a contaminant solution while a second reservoir contains the same volume of distilled water. The velocity of the flow can be controlled by a pump. The concentration of chemicals in liquids on both sides of the soil sample are determined frequently by chemical analysis so that the mass of pollutants moving through the sample is known. Also the effect of an overburden pressure on the soil sample can be considered [5]. The latter matches the boundary conditions of a landfill, an important fact, which cannot be considered in other testing methods.

In addition it is not only possible to accomplish with the DKS-permeameter pure diffusion experiments but also to realize a conventional permeability test. This can be achieved by a hydraulic gradient which can be chosen

between 1 and 500, as pressure can be given onto the solutions in the reservoirs.

However, the tests discussed in this paper are exclusively limited on the diffusion effect, as the convective mass transport is of minor importance in the clay material under consideration.

Figure 3 shows the equipment situated in a temperature controlled room as diffusion crucially depends on temperature.

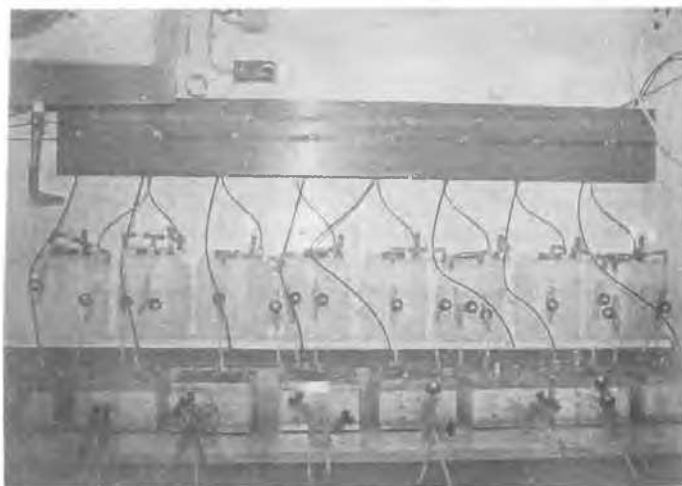


Figure 3: Test bench with eight DKS-permeameters.

## TEST MATERIAL AND SAMPLE PREPERATION

For sample preparation dry soil can be compacted by vibration to a dry bulk density of  $1,76 \text{ g/cm}^3$  (porosity = 0,32) while natural clays are to be fixed by the water content of the liquid limit.

All discussed experiments were carried out with a mixture of sodium-bentonite, quartz-flour and gravel (see table 1). This mixture was exactly classified by chemical and mineralogical composition as well as by grain-size distribution.

Table 1 shows the mineralogical composition and the soil-mechanical parameters of the mentioned soil sample.

Table 1: a: Soil-mechanical parameters of the test soil.

grain bulk density	2,80 g/cm <sup>3</sup>
plastic limit	26,4 %
liquid limit	191,0 %
plasticity index	164,8 %
coeff. of hydr. cond.	$2-3 \times 10^{-11} \text{ m/s}$
clay content < 0,002 mm	14,4 %
porosity	32 %

b: Mineralogical composition of the test soil.

sodium-bentonite Montigel F	14,4 %
quartz-flour SP 8	21,6 %
quartz-gravel	64 %

The experiment demands full water saturation of the soil sample. The dry soil sample gets moistened with clean water within 48 hours so that the bentonite can dwell and fill all holes between the quartz particles. A simulated landfill overburden pressure does counteract to the expanding pressure by a load unit. The end of the dwelling can be controlled by measuring.

After a saturation of 14 days a minimal hydraulic conductivity coefficient between  $1,9$  and  $3,2 \times 10^{-11}$  m/s does result. During this phase of saturation a hydraulic gradient of 200 is induced in advance of determining the hydraulic conductivity coefficient. When the consolidation is finished the definitive experiment is started with the flow of solutions through the permeameter as described above. During the experiments which last between 56 and 100 days the temperature is kept constant at 20° C.

**DETERMINATION OF DIFFUSION AND SORPTION PARAMETERS**

As figure 2 shows the transported solutions are collected behind the pump and get analyzed on the amount of transported pollutants. The results of the chemical analysis get illustrated by computer and characterised by emission graphs.

In figure 4 three decisive phases resulting from the interactions between diffusion and sorption are shown.

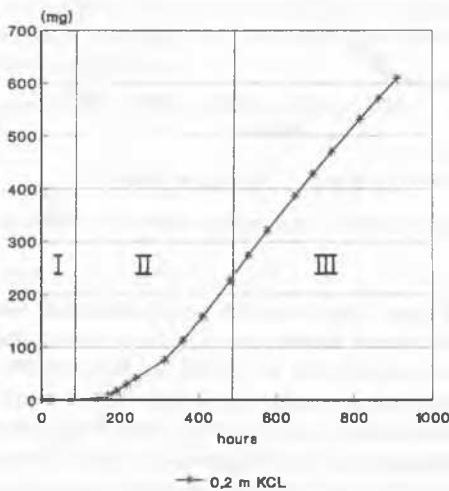


Figure 4: Phases of the pollutant transport experiment.

Phase I: No emission of potassium out of the soil sample can be seen, as all cations are retarded within the sample.

Phase II: Instationary phase; still the sorption capacity of the sample is not exhausted, but an increasing amount of pollutants emits out of the sample.

Phase III: Stationary phase; the sorption capacity of the sample is exhausted and pollutants migrate through the sample without any retardation. The amount of cations corresponds to the influence of diffusion and has reached a maximum.

From the quantity within the stationary phase the following diffusion parameters can be calculated:

$D$  = effective diffusion coefficient of the adsorbate in solution in the soil sample

$W$  = tortuosity factor

The emission-time curves can be backanalysized very precisely with the pollutant transport program Depotrans, developed by Dywidag Environmental, Munich [4].

As mentioned above fine grained clay mineral particles are able to adsorb a certain amount of pollutants during the so-called instationary phase of the pollutant transport. The curved line in figure 5 which results as a function of this sorption effect can be compared with the hypothetical curve, which is postulated to be non-sorptive. With the help of the Depotrans-simulation-program it is possible to evaluate this hypothetic graph by using the calculated diffusion-parameters (which give us the angle of ascent) and declaring the sorption parameters of the soil to the zero-position. Depotrans does now only consider the saturation of the pore-water. Offhands the resulting graph is situated parallel but higher than the experimental graph, and at an optional point of time within the stationary phase, it is possible to calculate the amount of adsorbate directly. Figure 5 shows a characteristic graph of a potassium-emission; the hypothetic non-sorptive graph is drawn in addition.

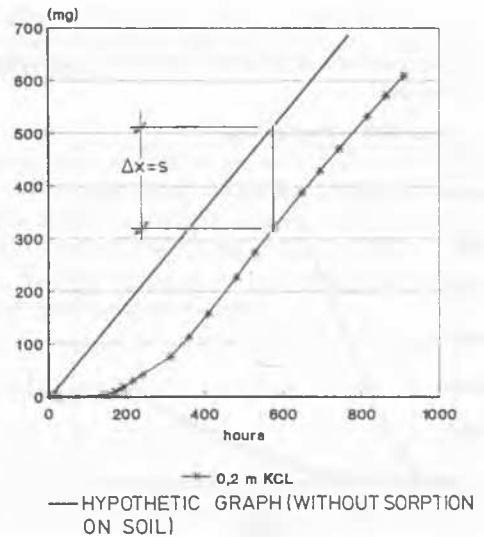


Figure 5: Calculation of the amount of adsorbed pollutants.

## TEST RESULTS

### Diffusion and sorption parameters

Table 2 shows some diffusion and sorption parameters of potassium determined at the bentonite-quartz mixture.

Table 2: Diffusion and sorption parameters

solution	D	W	adsorbed amount
0,2m KCl	$1,34 \times 10^{-6}$	0,068	1,4
0,05m KCl	$1,23 \times 10^{-6}$	0,063	0,8
0,2m KNO <sub>3</sub>	$1,26 \times 10^{-6}$	0,064	1,4
0,05m KNO <sub>3</sub>	$1,23 \times 10^{-6}$	0,063	1,2

D = effective diffusion coefficient [cm<sup>2</sup>/sec]

W = factor of tortuosity [-]

adsorbed amount [g/l]

All parameters were calculated as an average from twin tests.

The determination of D is based on the diffusion coefficient in free solution  $D_0$ . This parameter is determined rather precise by different authors. Table 3 shows this parameter for the mentioned ions.

Table 3: Diffusion coefficient in free solution  $D_0$  (after [6]).

K <sup>+</sup>	$19,6 \times 10^{-6}$ cm <sup>2</sup> /sec
Cl <sup>-</sup>	$20,3 \times 10^{-6}$ cm <sup>2</sup> /sec
NO <sub>3</sub> <sup>-</sup>	$19,0 \times 10^{-6}$ cm <sup>2</sup> /sec

### Emission graphs

In the following diagrams some characteristic results for a model-pollutant (potassium) are presented.

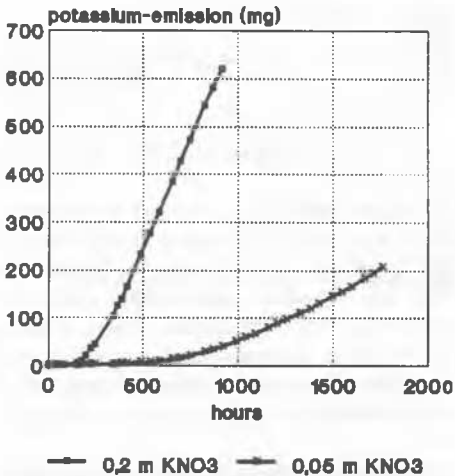


Figure 6: Test results with different potassium concentrations.

Figure 6 shows the effect of different concentrations of solutions of potassiumchloride. The higher concentration (0,2 m) does reach the stationary phase of the experiment after 20 days while the experiment with the lower concentration (0,05 m) does last for almost 90 days until all sorption effects are finished and the stationary phase does begin.

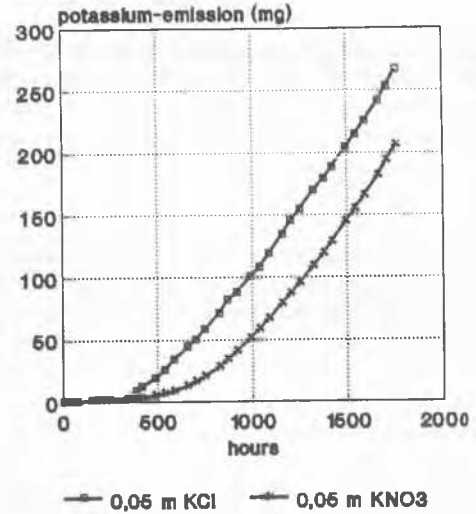


Figure 7: Influence of different anions on the emission of potassium (low concentration).

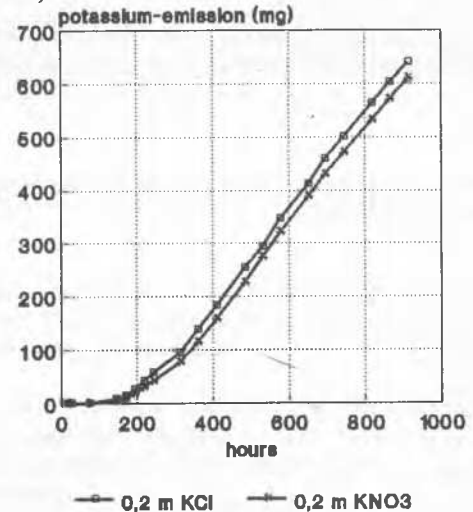


Figure 8: Influence of different anions on the emission of potassium (high concentration).

The comparison of figure 7 and 8 shows that lower concentrations tend to a lower adsorption on smectit surfaces than high concentrations. Also the influence of various anions within the solution can be depicted by low concentrations. Chloride-ions can run faster through the soil sample than nitrate-ions. Both anions effect a negative charge within the soil sample by trespassing. This treats positive potassium-cations to accelerate as they try to equal the negative charge. In the opposite the anions now get retarded by the positive cations. The more  $D_0$  of cations and anions diverge, the better this effect can be seen (see Table 3).

When chloride is present, potassium does reach the stationary phase earlier than a solution of potassiumnitrate. On the other hand in case of high

concentrations this effect gets overlaid by the amount of soluted ions. In both cases the smectite-surfaces tend to a more intensive exchange of cations than by the corresponding low concentration.

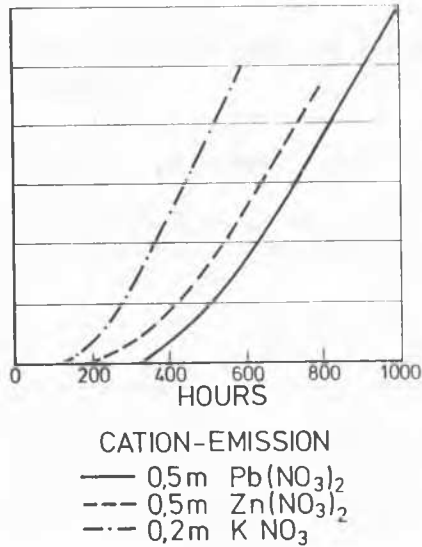


Figure 9: Comparison of the sorption of different cations (lead, zinc, potassium).

The qualitative comparison of different cations shows the increase of sorption from potassium over zinc to lead (figure 9). At first potassium does break through even as it is present in a lower concentration. This signifies a relative low affinity of potassium to the sodium-bentonite. Zinc gets adsorbed in a significant higher extent which is illustrated by the later break-through and the extended instationary phase. Lead, finally, does emit out of the soil-sample at the latest point of time, but does reach the stationary phase faster than zinc. The most important reasons determining both, the time of break-through of the ions, and the moment of reaching the stationary phase are the geochemical attributes, as valency, size and weight of the ions. The observed effect of extensive retardation of lead cations by sodium-bentonite gives an optimistic view on preventing the contamination of the environment by lead pollution.

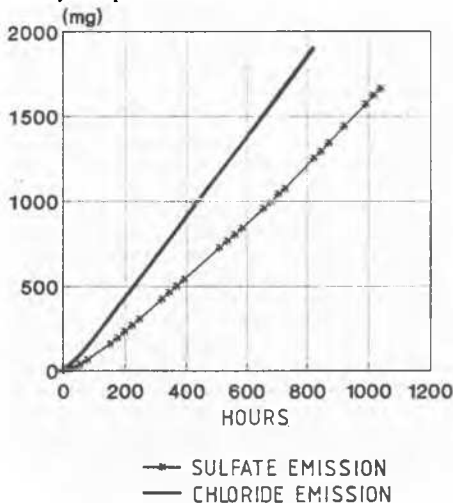


Figure 10: Comparison of emission of sulfate and chloride anions.

In the last figure we can depict sorption effects of sulfate anions within the soil sample. Usually anions are credited as conservative tracers without remarkable tendencies for ion-exchange. Indeed, the emission graph of chloride shows a very short retardation, only induced by saturation of pore-water and the time spent on passing through the soil sample. The noticed delay of the sulfate emission cannot be explained with these postulations, however. There must be additional sorption effects either on the broken-ends of the smectite-layers or precipitation and mineralisation effects within the soil sample.

## SUMMARY

A new laboratory testing method, the DKS-permeameter, was developed by Dywidag Environmentals, Munich, together with the Institute for Soil Mechanics of the Ruhr-University Bochum in order to determine mass-transport parameters under the consideration of landfill boundary conditions. A series of experiments was carried out with a classified soil sample by varying model pollutants (potassium) with different concentrations and solutions.

In addition to the determination of diffusion and sorption parameters interesting effects concerning the sorption characteristics of different metal ions were detected. Also a certain content of anion sorption within bentonite samples was discovered.

Still there are numerous effects to be examined and additional experiments are to be carried out. Though the first results give confidence in solving fundamental problems of measuring the pollutant transport phenomena.

## PROSPECTS

In the course of the research project pollutant mixtures of organic and inorganic substances will be examined, finally dump leachates will be taken in consideration, too.

With the help of the calculated sorption and diffusion data it will be possible to tailor landfill mineral basal liners fitting to expected hazardous dump contents. In addition geotechnical engineers will be able to estimate the aptitude of outcropping clays for usage of mineral liners or to refine them if necessary. An aspect which can not be rated high enough as construction and material costs increase permanently.

## ACKNOWLEDGMENT

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