THE COMPLEX EQUIVALENCY OF LINER SYSTEMS APPLIED FOR WASTE DISPOSAL

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Abstract

Legal regulation of municipal landfill siting and technical design is incomplete in Hungary so far, however remarkable progress has taken place in the last years as the Environmental Law was put into force. The requirements regarding the base-, and cover lining design of the hazardous and municipal landfills correspond to the current European practice. This fact is proven by numerical equivalency calculations of the liner systems. Uniform judgment about the applicability of the Geosynthetic Clay Liners (GCL-s) is not formed yet. The concern about applicability is the lack of international consensus on assessment methodology and the GCL's limited contaminant retention capacity.

The paper deals with the different types of equivalency according to the different contaminant transport processes, introduces the complex (numerical) equivalency and demonstrates the importance of equivalency calculations using the example of a bottom lining systems with and without the application of GCL's.

Theoretical Considerations (The migration of Contaminants in Porous Media)

The migration of contaminants consists of four different processes: advection (convection), dispersion, adsorption and degradation. The advection is the transport process due to the seepage of the groundwater. The dispersion means the spreading of contaminants caused by local concentration inhomogenity or seepage velocity. On one hand, the diffusion (contaminant transport due to concentration gradient) results in a spreading. On the other hand, there are some hydrodynamic reasons of this phenomena as well. Both the variation of the pathlines of the transport, the changes of pore diameter and the difference of seepage velocities within the pore; and the large scale inhomogenity of rock bodies lead to the dispersion of contaminants. However, the three different processes (diffusion, hydrodynamic dispersion and macrodispersion) listed above result in similar effects and that is the reason why they constitute jointly the dispersive transport. Since the linear adsorption and desorption of contaminant are mostly independent processes, the term of retardation containing both factors is taken into account during the calculations. However, the sorption process is usually non-linear, therefore the non-linear adsorption isotherms (Freundlich or Langmuir) have to be used to characterize the sorption side process. In case of special contaminants, the effect of radioactive decay or biodegradation might be significant. Consequently, their flux has also significant impact on the net process and neglecting those simultaneous phenomena can deteriorate the reliability of the calculations. The effects of all listed phenomena result in changes of contaminant concentration both in time and space, and it can be mathematically written in the following form, called general as a transport-equation:

$$\begin{aligned} R\frac{\partial c}{\partial t} = {} & D_{xx}\frac{\partial^2 c}{\partial x^2} + D_{xy}\frac{\partial^2 c}{\partial x \partial y} + D_{xz}\frac{\partial^2 c}{\partial x \partial z} + D_{yx}\frac{\partial^2 c}{\partial y \partial x} + D_{yy}\frac{\partial^2 c}{\partial y^2} + D_{yz}\frac{\partial^2 c}{\partial y \partial z} + \\ & + D_{zx}\frac{\partial^2 c}{\partial z \partial x} + D_{zy}\frac{\partial^2 c}{\partial z \partial y} + D_{zz}\frac{\partial^2 c}{\partial z^2} - \frac{\partial}{\partial x}\left(\frac{v_x c}{n}\right) - \frac{\partial}{\partial y}\left(\frac{v_y c}{n}\right) - \frac{\partial}{\partial z}\left(\frac{v_z c}{n}\right) - \lambda R c \end{aligned} \tag{Eq.1}$$

where c is the concentration, D_{ij} are the elements of the dispersion matrix, v is the pore velocity of seepage, n is the porosity, λ is the degradation coefficient, R is the rate of retardation, and x, y and z are the axes of the local (Descartes) coordinate system, respectively (1,4).

Types of Barrier Equivalency

We call „A” and „B” barrier systems equivalent if the concentrations at the protected side in time are equal presuming that the hydraulic and the concentration gradients are the same. This means that the „B” being an alternative barrier-system might only be used if the calculated concentrations on the protected side are less than the appropriate values using the „A” standard (recommended) barrier-system (3). Although this description is quite easy and clear, we may face a lot of practical and theoretical problems determining the equivalency. Understanding the problems it is expedient to discuss the types of equivalency.

The Advective Equivalency (Hydraulic Equivalency): Two barriers are advectively equivalent if the concentrations on the protected side - due to equal hydraulic and concentration gradient - are equal taking only the advective transport process into consideration. Since the advective term of the transport equation is given as the seepage velocity multiplied by the concentration in the pore volume, the advective equivalency means the hydraulic equivalency as well. To calculate the hydraulic equivalency the following formula should be used:

$$\sum_{i=1}^{n} M_i \,/ \sum_{i=1}^{n} \frac{M_i}{k_i} = \sum_{j=1}^{m} M_j \,/ \sum_{j=1}^{m} \frac{M_j}{k_j} \tag{Eq. 2}$$

where n and m are the number of layers in the standard and the alternative barrier-system, M_i and M_j are the thicknesses of the layers and k_i and k_j are the hydraulic conductivity values, respectively.

Some calculations on hydraulic equivalency of different barriers are listed in Table 1. It was proved that about 1.5 cm thick GCL (geosynthetic clay liner) is advectively equivalent to 60 cm compacted clay liner, and 7.5 cm thick, multi-layered GCL is needed to assure the same hydraulic equivalency with a geomembrane covered by 3x20 cm compacted clay liner. Practically, it might be said that an average GCL of about 1 cm thickness is advectively equivalent only to 2x20 cm compacted clay liner.

Table 1.: The hydraulic equivalency of different barrier-forming materials

Barrier-material	Average hydraulic conductivity [m/s]	Thick-ness [cm]	Case A [m]	Case B [m]	Case C [m]
Compacted clay liner	1E-9	60	0,6	3,1	93
Mixture of sand and 3% bentonite*	5E-7	(60)	300	1550	46500
Mixture of sand and 5% bentonite*	1E-7	(60)	60	310	9300
Mixture of sand and 10% bentonite*	1E-9	(60)	0,6	3,1	93
Mixture of sand and 15% bentonite*	8E-11	(60)	0,048	0,248	7,44
Geomembrane	8E-13	0,2	0,00048	0,00248	0,0744
Geomembrane with ∅5 mm hole**	4,7E-12	0,2	0,00282	0,01457	0,4371
Geomembrane with ∅1 cm hole**	9,5E-12	0,2	0,0057	0,02945	0,8835
Geomembrane with ∅2 cm hole**	1,9E-11	0,2	0,0114	0,0589	1,767
GCL (Geosynthetic clay liner)	2,5E-11	1	0,015	0,0775	2,325
Hydraulic asphalt liner	3E-11	5	0,018	0,093	2,79

Case A: Equivalent thickness [m] with 60 cm thick, $k=10^{-9}$ m/s compacted clay liner

Case B: Equivalent thickness [m] with 60 cm thick, $k=10^{-9}$ m/s compacted clay liner covered by 2 mm HDPE geomembrane

Case C: Equivalent thickness [m] with 60 cm thick, $k=10^{-9}$ m/s compacted clay liner covered by 2x2 mm HDPE geomembrane with drainage layer between

* Average hydraulic conductivity determined by using the experiments of Chapuis (2)

** Average hydraulic conductivity calculated by using spherical flow field (7)

The Diffusive Equivalency: Two barrier-systems are diffusively equivalent if the concentrations due to diffusion mass transport at identical concentration gradient are equal on the protected side of the barrier. To investigate the diffusive equivalency let us see the analytical solution of the Fick's first law. In a homogeneous medium the change of concentration in time and space is given by:

$$c_i(x,t) = c_{0i}\mathrm{erfc}\frac{x}{2\sqrt{D_i t}} \qquad \text{(Eq. 3)}$$

where c_{0i} is the constant concentration of the i contaminant at the polluted side of the barrier, D_i is the effective diffusion coefficient of the pollutant in the medium, and c_i is the concentration at x distance in time t (5). If „A" and „B" barriers are diffusively equivalent, then the c_i concentrations due to equal c_{i0} concentrations at the same x distance in time t are equal to

$$\frac{x_A}{\sqrt{D_A}} = \frac{x_B}{\sqrt{D_B}} \qquad \text{(Eq. 4)}$$

In Table 2. the results of diffusive equivalency calculations based on data from the literature (3,7) are summarized. Investigating the results of the calculations it seems to be obvious that the efficiency of the HDPE geomembrane and the GCL barriers against diffusive transport processes is much lower than that of the compacted clay liners. Depending on the product the HDPE geomembrane of 2 mm thickness or a GCL of 10 mm thickness is equivalent to 5-10 cm compacted clay liner regarding only the diffusive transport.

Table 2.: Diffusive equivalency of geomebrane and compacted clay liners

Pollutant	Effective diffusion coefficient [x10^{-10} m^2/s]	Effective diffusion coefficient [x10^{-12} m^2/s]	Equivalent thickness of compacted clay with HDPE geomembrane of 2 mm thickness [cm]	Equivalent thickness of HDPE geo-membrane with comp. clay liner of 60 cm thickness [mm]
	Comp. clay liner	Geo-membrane		
Methanol	4.8	0.8	4.9	0.00167
Acetone	3.4	0.6	4.8	0.00187
Ethyl-methyl ketone	3	0.55	4.7	0.00202
Acetic acid- ethylester	2.8	0.15	8.6	0.00017
Formaldehyde-solution	5.9	0.8	5.4	0.00110
Chloroform	3.1	0.25	7.0	0.00039
Carbon tetrachloride	2.9	0.25	6.8	0.00045
Trichloro ethylene	2.9	0.25	6.8	0.00045
1,2-Dichloroethane	3	0.25	6.9	0.00042
Tetrachloro ethylene	2.5	0.25	6.3	0.00060
Chlorobenzene	2.7	0.25	6.6	0.00051
Benzene	3	0.2	7.7	0.00027
Ethylbenzene	2.3	0.2	6.8	0.00045
Xylene	2.4	0.2	6.9	0.00042
Toloene	2.7	0.2	7.3	0.00033
Pentane	2.7	0.2	7.3	0.00033
Hexane	2.4	0.2	6.9	0.00042
Heptane	2.2	0.2	6.6	0.00050

Advective-Dispersive Equivalency: The above mentioned calculations of equivalency took only one transport phenomenon into consideration, namely, the advection or the diffusion. For investigation of more complex transport procedure Shackelford (8) proposed a solution. Shackelford derived the 1D transport equation's analytical solution in the following form:

$$\frac{c}{c_0} = \frac{1}{2}\left[\mathrm{erfc}\left(\frac{1-T_R}{2\sqrt{T_R/P_L}}\right) + \exp(P_L)\cdot\mathrm{erfc}\left(\frac{1+T_R}{2\sqrt{T_R/P_L}}\right)\right] \qquad \text{(Eq. 5)}$$

where c is the concentration of the solution on the polluted side of the barrier, c_0 is the initial concentration, $\mathrm{erf}(z)=\frac{1}{\sqrt{\pi}}\int_0^z \exp(-\xi^2)d\xi$, $\mathrm{erfc}(z)=1-\mathrm{erf}(z)$ is the standard error function and its complementer function, $T_R=\frac{v_s\cdot t}{R_d\cdot x}=\frac{v_R\cdot t}{x}$, $P_L=\frac{v_s\cdot x}{D^*}=\frac{v_s\cdot L}{D^*}$, where x is the distance from the surface of the barrier. Using this method the average transit time of a pollutant in a given barrier media was calculated considering a uniform flow field during a predefined pathline. (If a given concentration of the pollutant on the protected side of the barrier at a given time is allowed, we can calculate the average pathlength in the uniform flow field, which is equal to the required minimal L thickness of the barrier.).

Thus, the calculation consists of the following steps:

- Determination of the constants of the calculation (I: hydraulic gradient, k: hydraulic conductivity, n: porosity, c: allowable concentration at the protected side of the barrier at time t, c_0: concentration at the polluted side of the barrier, D*: effective dispersion coefficient of the pollutant in the investigated barrier, R_d: retardation coefficient),
- Determination of the seepage velocity in the pore volume: $v_s=\frac{k\cdot I}{n}$,
- Estimation of an L (required barrier-thickness) which assures that the c concentration on the protected site is lower at time t then the recommended limiting value,
- Determination the $P_L=\frac{v_s\cdot L}{D^*}$ value,
- Determination of c/c_0 value and from a published nomogram (8,9), the T_R value using the previously calculated c/c_0 and P_L values is to be determined,
- The average transport time of the pollutant can be calculated using $t_1=\frac{T_R\cdot R_d\cdot L}{v_s}$. and if $t_1>t$, then the barrier of L thickness is efficient, if not the calculation must be repeated using higher L value.

The Complex (Numerical) Equivalency: The presented equivalency calculations, however, have several limitations. The most crucial problems are as follows:

- Not all, but only some of the transport processes are considered;
- The barrier-forming material must be homogeneous (only one layer);
- Linear and monolayer adsorption is supposed (Henry-isotherm, Langmuir-isotherm) or no adsorption allowed at all;
- Constant concentration at the polluted side of the barrier required;
- Hydraulic gradient, hydraulic conductivity, porosity, effective diffusion or dispersion coefficient for the pollutant in the investigated barrier medium must be constant both in space and time.

For practical calculations these simplifications are too strict, so a new calculation method using the numerical solution of the transport-equation had to be introduced. Using the implicit finite difference method with the Peclet and Courant stability criteria all the above mentioned problems could be eliminated. The barrier is handled as a column of elements, where each element is characterized with its own thickness, hydraulic and transport properties. The mass equilibria of the pollutant due to any transport process is taken into consideration which is represented in the 1D transport-equation (Eq.1.). Using the FD method the effluent concentration vs. time function is calculated for any layer in the barrier-system irrespectively to the varying influent concentration. As a first step, the average seepage velocity in the barrier system is to be calculated. Then, the transport properties of the medium should be determined. As a precondition, the initial concentration distribution in the barrier system, as boundary condition the constant or varying concentration at the polluted side of the barrier is used. The calculation is performed layer by layer, starting with the top layer of the barrier. The concentration vs. time relationship at the bottom of the layer was calculated using the initial and boundary conditions. This concentration distribution in time is the input for the second layer, etc.(3).

For the complete numerical calculation of the equivalency the mentioned procedure must be run two times. At the first time, the calculations should be run using the data of the standard barrier system, and the second time applying the investigated alternative barrier system. The equivalency is proven if the concentration at the protected side of the alternative barrier system is lower than in case of the standard barrier system at the same time interval (10).

Calculation Methodology of the Complex Equivalency: Since incompatibility problems may arise between different leachates and barrier-forming media (particularly in case of compacted clays, GCL, geomembranes, geotextiles, etc.) it is highly recommended to perform the numerical equivalency calculations simultaneously for different contaminants. There are six groups of contaminants with rather different behaviour for the different barrier elements, which are the following:

- Cations of alkali metals and alkali-earth metals (Na, K, Mg, Ca)
- Anions of halogenides (Cl, Br, I)
- Toxic metals and heavy metals (Sr, Cd, Cu, Ni, Zn, Pb, Fe)
- Chlorinated hydrocarbons (mono and dichloro benzene, carbon tetrachloride, etc.)
- Alcohols and their derivatives (alocohols, aldehydes, ketones)
- Aromatic organic compounds (benzene, xylene, toluene, etc.)

For each group it is recommended to make the calculation using the data of the most hazardous component. The environmental hazard might be estimated using the c/c_0 value (allowed concentration at the protected side/maximal concentration on the polluted side). The equivalency calculation of any contaminant groups can be omitted if the occurrence of that leachate components is irrelevant. If the leachate composition in the planning phase is not known, a chemical analysis of leachates at similar pollution sources is advised. The general (representative) transport properties of the different contaminant groups for some barrier elements are listed in Table 3. Using the numerical calculations, the equivalency of standard and alternative barrier systems or their elements can be determined at much higher safety level, because the prescription made only by the hydraulic equivalency does not assures the equivalency if other transport processes are also taking place (3, 10).

Table 3.: Representative hydraulic and transport-parameters for calculations of the numerical equivalency

Property	Representative value			
	HDPE geo-membrane	Compacted clay liner	Substratum[1]	Drainage layer[2]
hydraulic conductivity [m/s]	10^{-13}	10^{-9}	10^{-8}	10^{-4}
Effective diffusion coefficient [m^2/s] 1.group 2.group 3.group 4.group 5.group 6.group	 $2\cdot10^{-16}$ $3\cdot10^{-16}$ 10^{-16} $2\cdot10^{-13}$ $6\cdot10^{-13}$ $2\cdot10^{-13}$	 10^{-10} $5\cdot10^{-10}$ $3\cdot10^{-11}$ $3\cdot10^{-10}$ $2\cdot10^{-10}$ $3\cdot10^{-10}$	 10^{-10} $5\cdot10^{-10}$ $3\cdot10^{-11}$ $3\cdot10^{-10}$ $2\cdot10^{-10}$ $3\cdot10^{-10}$	in saturated conditions $5\cdot10^{-9}$ $7\cdot10^{-9}$ $5\cdot10^{-10}$ $6\cdot10^{-10}$ 10^{-9} $6\cdot10^{-10}$
Dispersivity [m] (layer thickness[m])	0,0001 (0,002)	0,01(0,2) 0,025(0,6)	0,08 (3)	thickness dependent[3]
„A” parameter of the Langmuir isotherm [mg/kg]	T=0,001 meq/100g[4]	T=10 meq/100g[4]	T=5 meq/100g[4]	T=2 meq/100g[4]
„K” parameter of the Langmuir isotherm [m^3/g]	0,1	0,03	0,1	0,01
λ decay factor [1/s]	0	0	0	0
n porosity [-]	0.000001	.5	0.45	0.33
n_0 eff. porosity [-]	0.000001	0.02	0.04	0.33

[1] only at barriers for hazardous waste deposits

[2] only at drainage layers between isolation layers

[3] dispersivity might be determined depending on layer thickness using data from the literature

[4] The parameter in case of contaminant groups 1.-2.-3. might be calculated from CEC (T[meq/100g]) values. A[mg/kg]=M*10*T[meq/100 g], where A is the parameter of the Langmuir isotherm, M is the relative atomic weight. In case of group 4., 5. and 6. the values are to be determined during laboratory measurements

A practical application of complex equivalency calculations

The barrier efficiency of two types of GCL (further Type A and Type B) vs. compacted clay were investigated using different laboratory tests (9,10) (permeability tests at different hydraulic gradient, different normal load, investigation of behaviour after freeze-thaw and dry-wet cycles, investigation of overlapping GCL parts). The testing results for the two GCL's were rather different. As cause of the alteration the clay mineralogical composition of the filling material was found. The GCL type B filled with high montmorillonite content (81%) bentonite powder was much effective during the tests than the type A GCL either filled with lower smectite content powder or granulate (59%) (Fig. 1).

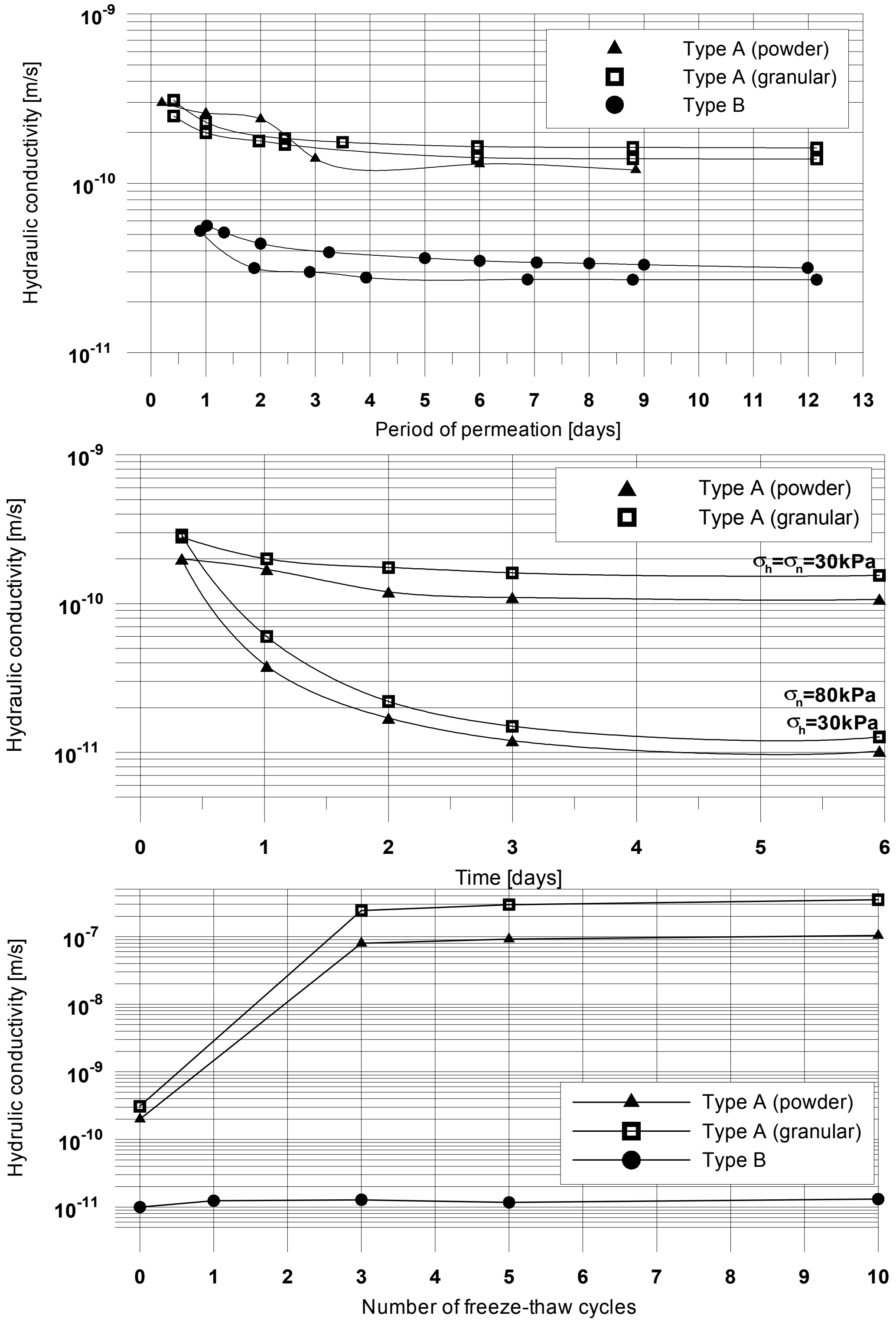
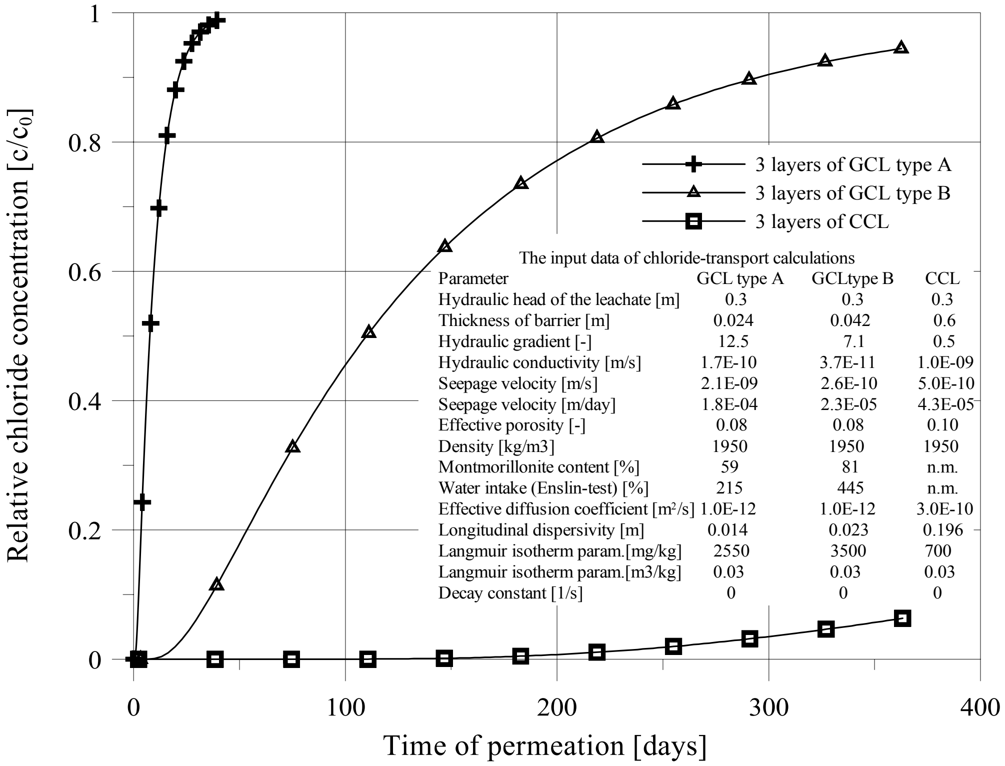


Fig. 1: The hydraulic behaviour of two GCL's during laboratory tests

However this difference in hydraulic behavior is transmitted to the barrier efficiency because of the advective inequivalency. Fig. 2.. demonstrates this effect. Both for the GCLs and both for a compacted clay liner break-through curves were calculated, where the difference of barrier efficiency is obvious. Allthough there is a much higher efficiency of GCL Type B than GCL type A caused by the advective inequivalency, but the advantage of CCL application is also clear regarding the much higher efficincy during the diffusive transport. With this calculation we would like to focus on the facts, that:

- Each type of GCL must be investigated with several laboratory tests to know its behaviour during the diffrent transport processes.
- There might be a big difference between GCLs of well known brandings in transport behaviour.
- The advantage of GCL vs. CCL is clear durin advective isolation, but there is a contraversary effect in case off diffusive transport occurence.



The input data of chloride-transport calculations

Parameter	GCL type A	GCLtype B	CCL
Hydraulic head of the leachate [m]	0.3	0.3	0.3
Thickness of barrier [m]	0.024	0.042	0.6
Hydraulic gradient [-]	12.5	7.1	0.5
Hydraulic conductivity [m/s]	1.7E-10	3.7E-11	1.0E-09
Seepage velocity [m/s]	2.1E-09	2.6E-10	5.0E-10
Seepage velocity [m/day]	1.8E-04	2.3E-05	4.3E-05
Effective porosity [-]	0.08	0.08	0.10
Density [kg/m3]	1950	1950	1950
Montmorillonite content [%]	59	81	n.m.
Water intake (Enslin-test) [%]	215	445	n.m.
Effective diffusion coefficient [m^2/s]	1.0E-12	1.0E-12	3.0E-10
Longitudinal dispersivity [m]	0.014	0.023	0.196
Langmuir isotherm param.[mg/kg]	2550	3500	700
Langmuir isotherm param.[m3/kg]	0.03	0.03	0.03
Decay constant [1/s]	0	0	0

Fig.2. The calculated break-through curves (GCL, CCL)

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