

Investigation of the functional lifetime of TRISOPLAST® in relation to chemical compositions of pore water solutions in barriers

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ABSTRACT

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Trisoplast® is a mixture of sand, bentonite and a polymer, used amongst others as landfill cover as well as barriers. Its permeability is generally lower than $1-2 \times 10^{-11} \text{ m.s}^{-1}$. Trisoplast keeps its functionality even in contact with different kinds of liquids, is not susceptible to cyclic drying and rewetting and the permeability hardly depends on its dry bulk density. Tests with 10% biaxial strain show only a slight increase in permeability. The functional lifetime of Trisoplast is governed by exchange of adsorbed sodium ions against dissolved calcium ions. This process depends on the composition of the pore water solution in the soil layers directly in contact with Trisoplast and the displacement rate of dissolved calcium ions either by convective or diffusive transport phenomena. The functional lifetime of Trisoplast even under relative extreme conditions is at least 100 years and scores clearly better than the reference sand-bentonite barrier.

Keywords: permeability, biaxial strain, cyclic drying, cation exchange

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Preface

In the Netherlands, legislation prescribes eternal aftercare of closed landfills and has Provincial governments entrusted with the organization and supervision. The level of soil protection, e.g. the quality of barriers, has to be maintained at the level prescribed in the landfill license and should comply with legal minimum requirements. Complete overhaul or reparation is prescribed, once the quality becomes less than minimally required. When the landfill is to be closed, landfill owners are levied to cover the capitalized cost of aftercare. The functional lifetime of barriers significantly determines the height of this amount. The lifetime of Trisoplast® has been investigated and compared to other mineral barrier materials under supervision of representatives of the different provincial governments involved in licensing: J. Ditters (prov. Brabant), E. Foppen (prov. Zuid-Holland), A. Kan (prov. Limburg), J. Kok(VVAV), M. Min (prov. Noord-Holland), M. Power (prov. Drenthe), A. de Wit (NV Afvalzorg Noord-Holland), S. Seuren (prov. Gelderland), R. Franken (RIVM) and W. van der Zon (Geodelft).

Summary

Trisoplast® is a mineral barrier material for landfills consisting of a mixture of sand and bentonite and a polymer. It has been applied on a large scale in the Netherlands often used in constructions combined with a geomembrane. Field investigations and numerical simulations have shown that in such constructions the moisture content of Trisoplast® once installed will not significantly change in future. Elevated temperatures, bi-axial strain up to 10%, liquids with either a high or a low acidity ($1.5 < \text{pH} < 10.5$), crude oil, diesel, phenol, landfill leachate and seawater (provided Trisoplast® could develop in contact with fresh water first) do not significantly affect the permeability. The capacity to attenuate the emission rate of heavy metals is mainly attributed to its extreme low hydraulic permeability. Ranking the performance of different liners in a construction according to the EC-guidelines on the basis of attenuation of heavy metal emission revealed the sequence: Trisoplast® > Clay (40%) > sand-bentonite > clay (10%) > GCL. In this report the approach of the functional lifetime of Trisoplast® is outlined. Lifetime is defined as a period of gradual deterioration of the barrier, which ends with a barrier quality that does not meet the legal or prescribed minimum requirement anymore. Exchange of adsorbed sodium and potassium ions on the bentonite surface against calcium and magnesium ions in the pore water of barriers has been identified as one of the major processes that control the functional lifetime. By this process the composition of the adsorption complex changes and adversely affects the permeability. The interaction between the chemical composition of pore water solution in the barrier and the permeability has been empirically determined through laboratory tests with liquids of different chemical composition: electrolyte concentration of 5 – 250 meq/l, SAR 0.005 – 3.00. The resulting swelling capacity, cation exchange capacity, composition of the pore fluid after the permeability tests and the total surface of the minerals were determined. From these results, a regression model has been derived describing the permeability of Trisoplast® as a function of the SAR-value and the sum of anion and cation concentration in the pore fluid of the barrier. A similar relationship has been derived for the potential swelling capacity. The procedure to estimate the functional lifetime includes the simulation of water and solute transport, exchange of adsorbed against dissolved substances and chemical equilibrium. This yields the evolution of the pore water composition in the barrier. The related permeability is calculated with the developed empirical function. The functional lifetime is found when the average permeability does not meet the legal requirements anymore. The potential water absorption of the bentonite-polymer mixture is slightly higher than the water absorption of pure bentonite: 10.4 vs. 9.1 gr. water per gram bentonite respectively. The relative effect of the chemical composition of the pore water on the swelling capacity of the bentonite and the fully developed bentonite-polymer is more or less equal for both materials.

The observed reference permeability of Trisoplast® is about half of the permeability of a mixture with sand and 13% bentonite, so the addition of polymer reduces the permeability by 50%. The (relative) effects of the chemical composition of pore

water on the performance of Trisoplast® and mixtures of sand with 13% bentonite are more or less equal at relatively high salt concentrations. Trisoplast® is less sensitive in concentrations of up to 400 meq/l, a value that is rarely exceeded under normal conditions. Mixtures of sand and 7.5% bentonite (standard BES) are much more sensitive than Trisoplast®. The functional life time of Trisoplast® and sand-bentonite (7.5%) barriers have been calculated for a profile containing bottom ash (residual material from the waste incineration processes), directly below the barrier and a second profile with a sandy layer separating the barrier from the bottom ash. Two different moisture conditions were assumed: (1) field situation (moisture content of layers under average field conditions)(pF2) and a relatively dry situation (pF3). Calculations show that the performance of Trisoplast® in all cases remains better than legally required. Contrary the functional life time of the sand-bentonite barrier was limited and varied from 10 years (without separating sandy layer and field capacity) to till more than 80 years (with a sandy separation layer and dry conditions, pF3). Under relatively severe conditions, the functional lifetime of Trisoplast® is at least 100 years. When, however, a rather limited increase of the permeability is acceptable, ageing of the mineral barriers can be effectively attenuated by treating the top of the supporting layer with a Na-rich material. Calculations show that the exchange rate is reduced significantly but the positive effect has vanished completely after 100 years. Laboratory experiments have shown that once the permeability of the mineral barriers have increased their original low permeabilities cannot be regained by applying solutions with relatively high sodium concentration So recovery is not likely. For designing barrier constructions and selecting construction materials, it is recommended to determine the chemical composition of soil layers that become into close contact with barriers. Materials that significantly have a detrimental effect on the permeability of the barrier should not be applied. Pre-investigations to determine the permeability of barriers should be executed with test fluids of the same chemical composition as the pore fluid in those soil layers. The test fluid should directly be applied to samples and without pre saturation using tap water. When such a test proves that the permeability remains lower than required, the barrier will keep its functionality during extremely long periods.

1 Introduction

In 1995 the Dutch Government issued the 'Prohibition on Landfilling Wastes Decree' to reduce the amount of waste sent to landfill. This decree states that only waste that cannot be reused or recycled may be landfilled and prohibits the landfilling of 32 categories of household and industrial solid waste. The waste that may be landfilled consists mainly of contaminated soil that cannot be cleaned, non-reusable building and demolition wastes and waste from incinerators, ash from incinerated sludge and contaminated building materials. Only 4% of the waste produced in 2010 may be landfilled. In 2000 waste processors landfilled about six million tonnes of waste. Isolation, maintenance and monitoring are the main concerns for landfill operators. Current landfills are sealed from the environment by a composite barrier to prevent pollutants leaching into the subsoil or the groundwater. Closed landfills are systematically checked and maintained and the surface layer is replaced when necessary (VVAV).

Under climatically conditions as prevail in the Netherlands, landfills are potential chemical time bombs when no precautions are made (Boels and Flemming, 1994). Liners and covers are projected to 'eternally' prevent soil and groundwater pollution. External factors of physical, chemical and biological nature should not adversely affect the barrier performance. Barriers should not be susceptible to: (1) irregular settlements resulting in bi-axial strain (positive as well as negative), (2) leachate, (3) waste or polluted soil (4) elevated, low or changing temperature, (5) drying and rewetting and (6) root penetrations . Although adverse effects cannot be ruled out completely, the functional lifetime of barriers should be as long as possible in order to minimise costs of aftercare, including eventual overhaul of the barrier construction.

Barrier requirements

In the Netherlands, the construction of barriers and covers includes a geo-membrane and a mineral barrier. The two layers are in direct contact with each other. This results in a higher level of protection than is reached by the sum of the separate layers because the series connection has an added value. If one layer fails the other layer will take over. To judge the influence of external factors on the durability of the single mineral barrier the protection by the geo-membrane should not be taken into account. The performance of the mineral barrier is most important at the spots where the geo-membrane fails as a result of installation failures or ageing processes. Derived from EC-legislation and directives, minimum requirements have been prescribed. For mineral barriers the maximum leakage rate should be less than 0.0001 m/d (1.157×10^{-9} m/s) when a water column of 1 m is applied on the top of the barrier and free outflow conditions prevail underneath the barrier and less than 1.59×10^{-10} m/s when a geo-membrane is combined with a mineral barrier (Hoeks et al. 1991; Boels, 1993). This requirement provides freedom to the designer: a moderate permeability has to be compensated by a certain thickness of the barrier, while thin barriers require extremely low permeability. In practise a number of requirements

have been adopted to assure a sufficiently long term performance. Effects of chemicals (leachate), elevated temperature of 40 centigrade continuously, biaxial strain of up to 10%, micro organisms and drying and rewetting cycles, should be minimal and at least not jeopardise the minimum requirements. The water content of mineral materials can significantly affect their behaviour when subjected to strain. The materials should maintain their low permeability even when exposed to strain under non-saturated conditions as are to be expected in practice. When strained (up to 10% biaxially) fissuring and crack formation should be absent or material characteristics should at least ensure restoration upon rewetting (Boels and Schreiber, 1999).

The aftercare legislation in the Netherlands stipulates that a barrier or any other object that is part of the constructions has to be replaced when the minimum requirements are not met any more. So a significant functional life time of barriers proves to be cost effective.

Trisoplast® is a relatively new material, which consists of a mixture of sand, bentonite and a polymer and has an extremely low permeability and is rather unsusceptible to different dissolved chemicals. It is applied on a large scale in the Netherlands and its characteristics have been extensively tested. This report presents an overview of the characteristics of Trisoplast®, results of laboratory research as well as numerical modelling to estimate the functional lifetime of Trisoplast®.

2 Characteristics of TRISOPLAST®

2.1 Effect of design on moisture conditions.

In the Netherlands, the sealing system normally contains a combination of a mineral barrier and a geomembrane. The design of landfill caps include a coarse grained mineral layer covering the waste body. This layer serves as gas drainage and supporting layer for the mineral barrier. On top of the mineral barrier a geomembrane is applied. A layer for draining of excess rainfall (200 – 400 mm / y) is installed above the geomembrane and normally consists of a coarse grained sand (0,3 m). The drainage layer is covered with a soil layer of 0.5 – 0.8 m (fig. 1 A.).

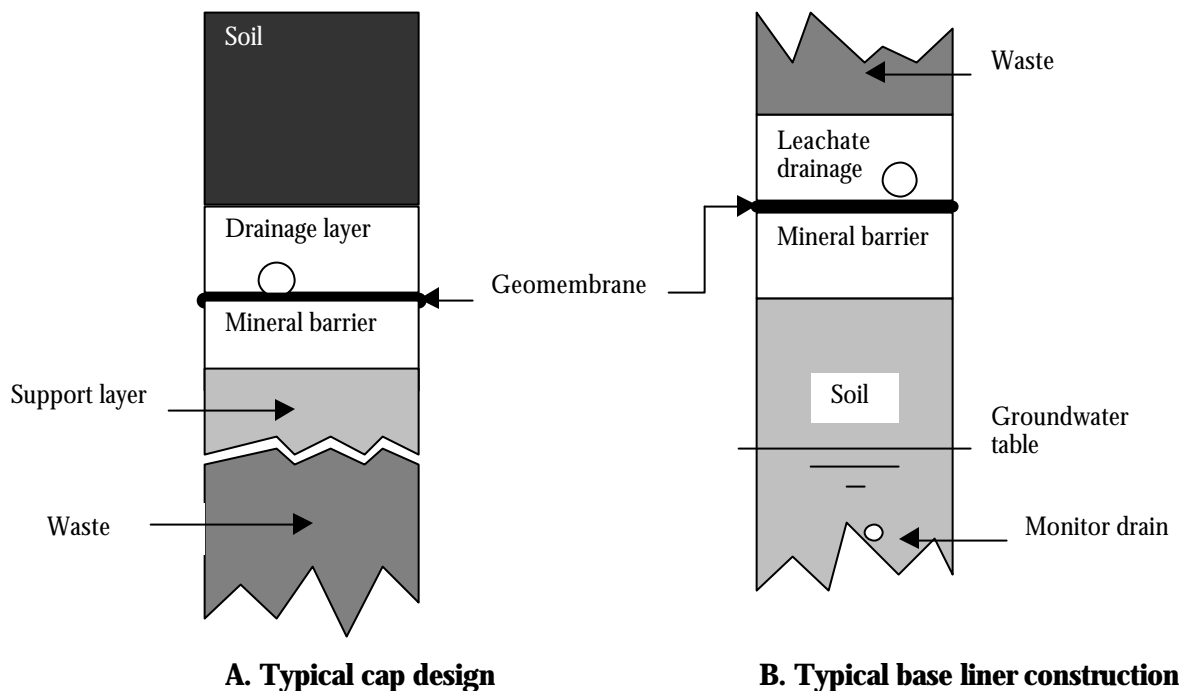


Fig. 1 Typical landfill design in the Netherlands

The base liner has a similar design (fig. 1 B.). Here (changing) temperature and water potential gradients can cause displacement of liquid water and water vapour. Field observations showed near saturation conditions and no significant variation of water content within TRISOPLAST®. Model calculations show that vapour transport has no major effect on water displacement (and drying) of TRISOPLAST® below a geomembrane in cap constructions when the temperature in the waste body is not elevated. The annual water content changes are small (fig. 2) and result after a period of 100 years in a reduction of the moisture content of less than 3%. When, however, the temperature in the waste body is elevated, the water content of TRISOPLAST® in covers increases significantly. Depending on the initial moisture content,

saturation is almost achieved within 100 year when an average temperature gradient of about 5 °K/m exists (Boels and Beuving, 2000).

Under similar condition, however, basal barriers will reduce their moisture content when they are installed above a coarse grained soil with a groundwater table below (Holzlöhner, 1988; Collins et al. 1988).

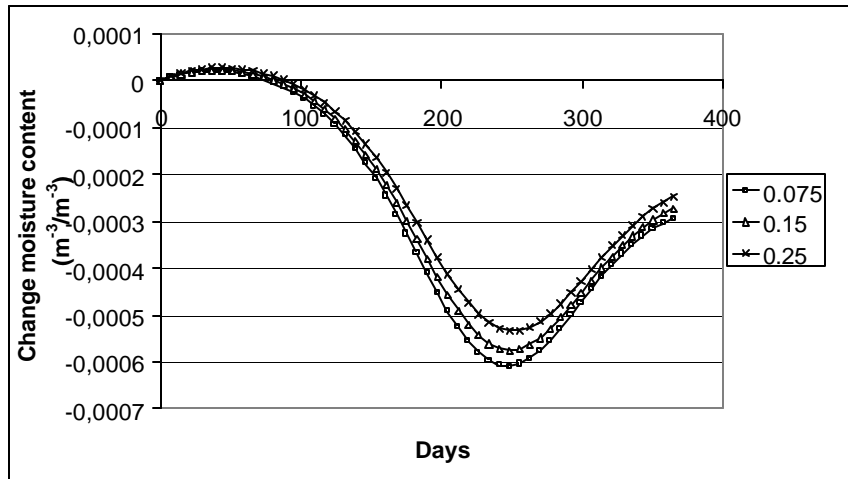


Fig.2 Calculated change of moisture content ($m^3.m^{-3}$) of landfill caps caused by dynamic temperature gradient induced vapour transport at different depths (0.075 – 0.25 meters) below the geomembrane of landfill caps

In a capping situation displacement of liquid water causes an equilibrium condition within two years after installation in the mineral barrier and its supporting layer. Because of the coarse nature of waste material, exchange of liquid water with the waste body is not likely, unless a significant amount of leachate accumulates in the waste body.

From this investigation one can conclude that effects of strain on the permeability of barriers should be tested with a water content that will exist when an equilibrium situation has been achieved in the liquid water phase. In practise this corresponds to a water content present during installation.

In deviation of the standard Dutch regulations the design of landfill caps occasionally only contains a single mineral layer without a geomembrane. This because older regulations apply for some older landfills that were closed some time ago. In other countries as well as for other applications a design without a geomembrane is still commonly used.

Here the moisture conditions of the layer can be influenced by drying effects from above and by influence of roots. Even in combination with a geomembrane these effects also can influence the mineral barrier in areas where the geomembrane is damaged or might fail in future. As the correct performance of the mineral barrier is needed precisely there effects should not be neglected even in combination with a geomembrane.

2.2 Drying cycles and crust formation

Mineral barriers can dry out during dry seasons and/or by the influence of plant roots. Drying cycles can lead to crack formation and the forming of aggregates in the clay or bentonite which results in a higher fraction of big pores.

Wet/dry cycles in a mineral barrier can also lead to a higher concentration of calcium ions in the barrier than the concentration present in the soil layer on top of the barrier. Calcium ions can concentrate in the barrier. Repeated drying and wetting of cracks can even lead to crust formation at the crack surface by salts dissolved from the above layers.

The bentonite-polymer complex of Trisoplast shows strong swelling and shrinkage behaviour, which does not change after a number of drying-wetting, cycles (Weitz, et al. 1994). When, however mixed with sand at the prescribed ratio, the sand fraction constitutes a skeleton with a certain pore system. This skeleton does not show swelling and shrinkage behaviour, under a load of 20 kPa (~ 1m) and when the bentonite content is less than 15% according to a study on sand/ bentonite mixtures (Groeneveld et al. 1991). Mineral barriers that do not have a skeleton of sand are more sensitive to crack formations.

Further research in Germany indicated that mineral barriers with clays and geosynthetic clay barriers might fail due to desiccation and plant root penetration. While Trisoplast performed much better in laboratory experiments than traditional mineral barriers with respect to crack formation, these laboratory results also needed additional validation from field data. To check the occurrence of ageing phenomena in practise, Trisoplast barriers installed in the years 1995 and 1996 have been excavated and sampled in October 2001. All inspected barriers were homogeneous in respect to water content, density, thickness, plasticity and structure. No indications for desiccation, crack formation or other aging processes were found by visual or microscopic inspection.

Further research is done by Melchior + Wittpohl Ingenieurgesellschaft, Hamburg, Germany, to the long term effect of cyclic drying and rewetting on mineral barriers.

2.3 Effect of temperature

Temperature effect on mineral barriers result in (1) change of the viscosity of the liquid and (2) aggregate formation during freezing and thawing cycles, which occurs in clay soils under natural conditions. From experiments conducted by Weitz and Boels (1994) it can be concluded that prolonged freezing and thawing cycles do not have a lasting effect on bentonite containing barriers. This conclusion is supported by Kraus et al. (1997).

Also prolonged exposure to elevated temperature did not affect the permeability (Weitz et al. 1997).

2.4 Differential settlements / strain

In practice differential settlements cause strain up to an average of about 2-5% (Boels and Van de Wal, 1999). To evaluate the effects of bi-axial strain, a special test equipment was developed by Boels (Boels and Schreiber, 1999; Boels & Van de Wal, 1999), which allows samples to be subjected to bi-axial strain of up to 10% and loads up to 250 kPa. Tests on the behaviour of different mineral barrier materials have been executed for saturated and unsaturated conditions (moisture content equal to the installation moisture content) and a load of 25 kPa, representing conditions typically found in landfill caps (see table 1).

Table 1 Effect of bi-axial strain on permeability of Trisoplast®

| Strain (%) | Permeability ($\times 10^{-10}$ m.s ⁻¹) | |
|------------|--|-------------|
| | Saturated | Unsaturated |
| 0.0 | 0.06 | |
| 1.0 | 0.09 | |
| 2.0 | 0.11 | |
| 2.5 | | |
| 3.0 | 0.16 | |
| 5.0 | 0.23 | |
| 7.5 | 0.21 | 0.60 |
| 10.0 | 0.21 | 0.37 |

Unsaturated = moisture content during installation

These experiments show that strain effects are small. These results are in sharp contrast with experiments of Edelmann, 2000, executed with materials with low clay content. This author reports significant increased permeability which could be attributed to fissures. It should be noted, however, that this author did not apply any load to the barrier during straining, which is not realistic and explains for a great deal the different results.

Effects of negative strains in mineral barriers are not expected when this negative strain is less than -4% (Egloffstein et al., 1996)

2.5 Susceptibility to dissolved chemicals

Two major effects with mineral barriers can be attributed to dissolved chemicals: (1) the potential deterioration of the clay crystal structure (high or low pH) and (2) the increase of the effective porosity (high salinity, high calcium / sodium ratio, or low dielectric constant of the liquid). Experiments with high and low acidity (pH 10 respectively 1.5) show no adverse effects. Experiments with a range of different calcium-sodium ratio were executed in a laboratory experiment and described in this report (see 3.4.3). The effects of moderate total salt concentrations is more or less in agreement with the theoretical expected effect as outlined by Boels et al (1993), Boels and Van der Wal (1999), Boels (2001a & b) and experiments of McNeal et al, (1966), Quirk and Schofield (1955), Reeve and Tamaddoni (1965). But in contrast with this expectation, the

permeability of mineral barriers for pure seawater appears to depend on the saturation degree prior to exposure to this liquid. The permeability of TRISOPLAST® increased in a period of 640 days from 2.5 till 9.0 x10⁻¹¹ m/s when initially TRISOPLAST® was completely saturated with fresh water. When, however, the initial saturation degree was about 10%, the permeability increased to about 35 x10⁻¹¹ m/s (see table 2). According to Didier and Comeaga (1997), GCLs (Geo-synthetic Clay Liners) behave in a similar but a much more pronounced way. These authors report a permeability of a GCL for leachate (sum anions + cations ~268 meq/l, SAR = 0.52) of 4 – 6 10⁻¹¹ m/s when the samples were completely saturated prior to the measurements. When the samples were saturated to a degree of 80%, prior to exposure to leachate, the permeability after 120 days increased to 5.5 10⁻⁹ m/s. A similar phenomenon was found when a liquid with a low dielectric constant was applied. Experiment with crude oil showed the effect of immiscible fluid flow in porous materials (Boels and Veerman, 1996), but this fluid did not affect the permeability of TRISOPLAST®.

An overview of permeability of Trisoplast® for different liquids is presented in table 2

Table 2 Permeability of Trisoplast® for different liquids (falling head, sample thickness 0,025 m)

| Dry bulk density | Liquid | PH | Permeability | Remarks |
|------------------|--------------|-----|--------------|-----------------|
| 1680 | Water + HCl | 1.5 | 0.89 | after 440 days |
| | Water + NaOH | 8.9 | 1.0 | after 440 days |
| | Water + NaOH | 10 | 0.9 | after 440 days |
| | Leachate | 7 | 3.7 | after 440 days |
| 1560 | Water + HCl | 1.5 | 1.23 | after 440 days |
| | Water + HCl | 3 | 1.48 | after 440 days |
| | Water | 5 | 1.5 | after 440 days |
| | Water + NaOH | 8.9 | 1.7 | after 440 days |
| | Water + NaOH | 10 | 1.05 | after 440 days |
| | Leachate | 7 | 4.3 | after 440 days |
| 1490 | Water | | 1.2 | after 200 days |
| | Water | | 1.0 | after 640 days |
| | Crude oil | | 0.3 | after 640 days |
| | Phenol | | 1.5 | after 196 days |
| | Diesel | | 16.0 | after 640 days |
| | Seawater | | 9.0 | After 640 days, |
| | Seawater | | 33.8 | 10% saturation |

2.6 Attenuation of contaminants

When applied below a waste body, a liner should prevent or at least attenuate the dispersion of contaminants. Attenuation from a single mineral liner is obtained from (1) a low percolation rate and (2) the distribution coefficient of contaminants in the soil solution. The distribution coefficient (ratio of adsorbed concentration over dissolved concentration) can be derived from an empirical relationship between adsorbed and dissolved substances:

$$\text{Log}(Q_{me}) = A1 + A2 * \text{log}(CEC) + A3 * \text{pH} + A4 * \text{Log}(\text{clay}\%) + A5 * \text{Log}(\text{Org. carbon } \%) + A6 * \text{Log} [\text{Log}\{(a_{Me})/\text{sqrt}(a_{Ca})\}]$$

where:

| | | |
|-----------------|---------------------------------------|---------------------------|
| Q _{me} | = adsorbed metal in dry soil | (mol/kg) |
| CEC | = Cation exchange capacity | (eq/kg) |
| Clay% | = percentage particles < 2 micrometer | |
| Org. carbon% | = percentage organic carbon | (~ 0,56 * organic matter) |
| a _{Me} | = activity metal in soil solution | (mmol/l) |
| a _{Ca} | = activity calcium in soil solution | mol/l) |
| a | = activity coefficient | (< 1 ~ 0.013 * EC) |

The constants A1 through A6 are listed in table 3

Table 3 Coefficients in the empirical relationship for the distribution of dissolved and adsorbed substances

| Metal | Symbol | Atomic weight | Constants | | | | | |
|---------|--------|---------------|-----------|------|------|-------|-------|------|
| | | | A1 | A2 | A3 | A4 | A5 | A6 |
| Cadmium | Cd | 112.4 | -3.15 | 1 | 0.5 | -0.24 | 0 | 0.82 |
| Copper | Cu | 63.5 | -3.85 | 0.52 | 0.7 | -0.14 | 0.46 | 0.55 |
| Lead | Pb | 207.2 | -4.4 | 0.62 | 0.6 | 0 | 0.462 | 0.55 |
| Zinc | Zn | 65.4 | -3.42 | 1.3 | 0.75 | 0 | 0 | 0.75 |

From the empirical relationship the distribution coefficient (K_d) is calculated as the ratio absorbed substances (meq/kg) and the fluid concentration (meq/l). The retardation coefficient is calculated from: $R = 1 + K_d \times \text{dry bulk density} / \text{volumetric moisture content}$. The flux of substances equals the pore water flux divided by the retardation coefficient. The attenuation can be calculated given the chemical and physical characteristics of pore water and liner material and subsequently the breakthrough time.

Ranking the performances of different liners in a construction after the EC-guidelines on the basis of the emission attenuation, the following sequence was found: Trisoplast® > Clay (clay fraction 40%) > Sand-bentonite > clay (clay fraction 10%) > GCL.

3 Expected functional lifetime in relation to chemical compositions of pore water solutions in barriers

The functional lifetime of barrier materials in the Netherlands is defined as a time period between installation and the moment when the quality becomes less than prescribed in the landfill licence. The minimum quality is expressed as a leakage rate less or equal to 0.0001 m per day when a hydraulic head of 1-m water column is applied to the top of the barrier and free outflow prevails. In previous chapters it was explained that most physical and chemical phenomena in landfills would not seriously affect the permeability of Trisoplast®. A theoretical study of Boels and Van der Wal (1999), shows that changes of the ratio of mono- and bivalent cations absorbed to the clay complex of mineral barriers can significantly increase the permeability of these materials, although still a long functional lifetime of Trisoplast® was expected. In practise changing of an initially high into a low ratio of these ions (especially the sodium and calcium ions) is likely when Trisoplast® or any other mineral barrier is in intimate contact with a layer with relatively high calcium contents. Under such circumstances, molecular diffusion or transport by water flow causes exchange of bivalent cations between both layers. The rate of this process has a great influence on the functional lifetime. Relevant for the permeability of clay-based mineral barriers is the ratio of monovalent over bivalent cations, either absorbed to the clay complex or in the soil solution and the total electrolyte concentration of the soil solution. In general the most dominant ions are sodium, potassium, calcium and magnesium. Sometimes manganese plays a role.

The theoretical background of the lifetime approach has been basically derived from Lagerwerf et al. (1969) and is described by Boels and Van de Wal (1999) and more extensively by Boels (2001).

Laboratory experiments have been conducted to evaluate the effects of the composition of soil solutes on the permeability of Trisoplast®. The measuring programme includes:

- Actual composition of liquid in test samples;
- Cation exchange capacity;
- Total surface of the minerals;
- Effect of liquid composition on the potential swelling capacity;
- Effects of liquid composition on the hydraulic permeability;
- The presence of recovery mechanisms to reconvert higher permeable barriers back to lower permeable barriers.

3.1 Methods

3.1.1 Permeability

The permeability was measured according to the falling head method. This implies the application of a certain water pressure to the bottom of a sample to allow an upward flow direction in order to prevent air to be trapped. An outflow at a fixed elevation was provided at the top of the sample. The hydraulic head in the burette (fig. 3) controls the applied water pressure.

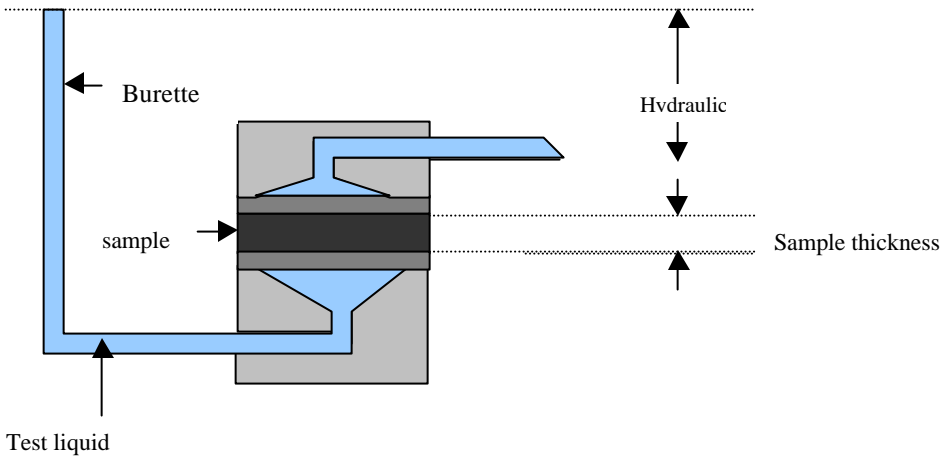


Fig 3 Laboratory set up for measuring the permeability according to the falling head method.

The average permeability during a time step is calculated from readings at two successive time moments:

$$K_{sat} = -\frac{l}{\Delta T} \frac{a_{burette}}{A_{sample}} \ln\left(\frac{h(T + \Delta T)}{h(T)}\right)$$

where:

| | |
|---------------|--|
| K_{sat} | permeability (m/s) |
| T | elapsed time during two readings (s) |
| $a_{burette}$ | area cross section burette (0.0000283 m ²) |
| A_{sample} | area cross section sample (0.00785 m ²) |
| $h(T)$ | level of liquid in burette at time moment T (max. 1 m) |
| $\ln(...)$ | natural logarithm (base e) |

Measurements were continued until a stable situation with no trends and limited variations was achieved. In case the permeability had already been determined flushing was provided to obtain a chemical composition in the sample equal to the composition of the applied liquid.

In these experiments a sample height of 0,015m was used.

3.1.2 Potential swelling capacity

The potential water absorption capacity had been determined according to the Enslin-methode. A quantity of oven dry mineral material, containing about 1 gram of clay, is pored on a coarse filter connected to a horizontally placed burette (fig. 4).

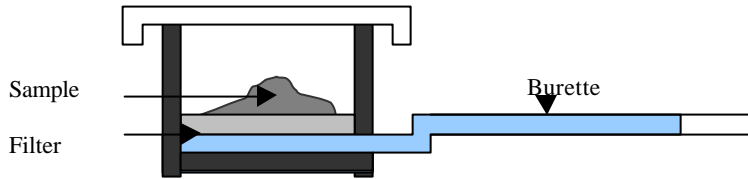


Fig. 4 Sketch of the set up for measuring the water absorption capacity after the Enslin method

Readings from the burette tell the absorbed quantity of water. The water absorption capacity is calculated as the quantity of absorbed water per gram of clay.

3.1.3 Total surface area of minerals

The principle of the determination of the surface area of minerals includes creating a monomolecular layer on the minerals. From a known weight of absorbed monomolecular layer of liquid per unit area of minerals, the total area is determined by removing the absorbed layer and measure the loss of weight. The method described by Klute (1986) based on ethyleneglycol has been applied. The weight of a monomolecular layer of this liquid per square metre amounts 0.00031 g per square meter.

3.1.4 Chemical composition of liquid in pores of the sample

At the end of the permeability measurements saturated samples are taken. Part of the liquids is removed by centrifuging. The collected liquid samples are diluted in a ratio of one to 5 or 10. The concentration of the parameters Na, K, Ca, Mg, Al, Fe and Mn is determined with an ICP (Thermo Jarrell Ash).

3.1.5 Cation exchange capacity

The principle of determining the cation exchange capacity (CEC) is that all absorbed cations are replaced by Ba-ions. Then a known quantity of Mg-chloride with a known concentration is applied to remove all Ba-ions. In the suspension the concentration of Mg has been determined. The difference between the applied Mg quantity and recovered Mg in the suspension was needed to replace all Ba-ions. From these data the CEC is calculated. The procedure described in NEN 5738 was used.

3.2 Test liquids

The permeability and the potential swell capacity are closely related to the ratio of absorbed mono- and bivalent cations. The ratio of absorbed cations at the clay surface corresponds to the ratio of these ions in the soil solution. The relationship between both ratios is expressed as the 'Sodium Adsorption Ratio (Bolt and Bruggewert, 1978) and can be expressed by the Gapon equation:

$$\frac{\Gamma_{Na}}{\Gamma_{Ca}} = K_{Gapon} * SAR = K_{Gapon} * \frac{Na}{\sqrt{\frac{Ca + Mg}{2}}}$$

where:

G concentration at absorption complex (eq/kg)
 K_{Gapon} Gapon factor
 Na and Ca concentration in liquid (eq/l)

The Gapon factor amounts about 0,5 for the majority of clay minerals and about 1 for montmorillonite. Above a certain SAR-value, the clay complex is almost completely occupied by sodium (and other monovalent ions). The composition range of the test includes a concentration where bentonite (montmorillonite) potentially is in a dispersed state (maximum swelling capacity) and a concentration where an aggregated state can be expected (no or very low swelling capacity). Rowell (1963) reports that montmorillonite starts to show swelling when the total concentration of the liquid is less than 100 meq/l and a dispersed state is found when the total concentration of all ions falls below 6 meq/l.

The used artificial test liquids in the experiments have a sum of cation and anion concentration range of 10 tot 500 meq/l and a range of $\{Na / (Ca + Na)\}$ ratios from 0.05 to 0.95 which corresponds with typical SAR-values from 0,05 to 4,25.

Table 4 Composition of liquids used in the tests

| Liquid | Total concentration of the liquid (meq/l) | Concentration (meq/l) | | SAR |
|--------|---|-----------------------|---------|-------|
| | | Sodium | Calcium | |
| 1 | 10 | 4.75 | 0.25 | 0.600 |
| 2 | 100 | 47.5 | 2.5 | 1.900 |
| 3 | 500 | 237.5 | 12.5 | 4.250 |
| 4 | 10 | 2.5 | 2.5 | 0.100 |
| 5 | 100 | 25 | 25.0 | 0.320 |
| 6 | 500 | 125 | 125.0 | 0.180 |
| 7 | 10 | 0.25 | 4.75 | 0.007 |
| 8 | 100 | 2.5 | 47.5 | 0.023 |
| 9 | 500 | 12.5 | 237.5 | 0.051 |

3.3 Materials and test procedures

Trisoplast® samples were installed at a dry bulk density of 1650 kg.m⁻³ using a manually operated press to obtain the prescribed thickness of the sample. The samples (18) were saturated with liquid number 1. After 3 x 24 hours the burettes were filled with liquid 1 and the permeability tests were continued until a permeability did not change significantly. Then the different fluid types were applied and measurements were continued. After completion of the measurements, test fluids of samples exposed to liquid 1 and 9 were changed to 9 and 1 respectively in order to investigate recovery mechanisms.

A second experiment was conducted with Trisoplast® samples (9) prepared with 10% tapwater, installed at a dry bulk density of 1650 kg.m⁻³ and directly exposed to the final liquid. Permeability has been tested with different test liquids. No further research on the composition of the pore fluid, CEC has been done.

To show the effect of the polymer, identical experiments were conducted with sand-bentonite mixtures with the same bentonite content as Trisoplast®. Of these samples the permeability was measured with tapwater first, followed by one of the nine test fluids. All data are compared with results of a mixture of sand and 7.5% bentonite, which is the reference material in the Netherlands for mineral barriers.

3.4 Results

3.4.1 Composition pore fluid and CEC

GID advised to saturate Trisoplast samples with a liquid containing a low concentration of sodium and calcium to promote the chemical binding process between the polymer and the bentonite of the fresh mixture. Therefore the samples of the first test were saturated with liquid 1. Also the reference permeability of all samples in this test were measured with test liquid 1. The samples were prepared with a bentonite-polymer mixture, provided by GID and commercially acquired sand. Chemical analysis of the collected outflow during the permeability measurements in the period 2 –12 days after the start of the measurements showed that the sodium content was very high (390 to 1190 meq/l) and very much higher than of the applied test fluid 1. Analysis of the sand used to prepare the Trisoplast samples showed that it concerned poorly washed sea sand. The reference permeability measurements have been prolonged as to leach as much sodium as possible. When the permeabilities did not show significant trends, the different test fluids were applied. At the end of the permeability measurements, the pore water composition of the samples was determined (table 5). The concentration of calcium complies more or less with the concentration in the test liquid whereas the sodium concentration in the pore liquid significantly exceeds the concentration in the test fluid by about 100 meq/l (table 5, fig. 4 A and B).

Table 5 Composition of fluid extracted from the pores, Sodium Adsorption Ratio and Cation Exchange Capacity of Trisoplast®

| Sample | Composition pore fluid (meq/l) | | | | CEC (meq/kg) |
|--------|--------------------------------|------|-------|------|--------------|
| | Na | K | Ca | Mg | |
| Tp1,1 | 187.3 | 2.9 | 11.58 | 4.64 | 107 |
| Tp3,1 | 338.9 | 1.24 | 14.81 | 5.65 | 107 |
| Tp6,1 | 293.6 | 1.33 | 92.87 | 8.16 | 110 |
| Tp9,1 | 156.8 | 1.05 | 182.1 | 8.75 | 111 |

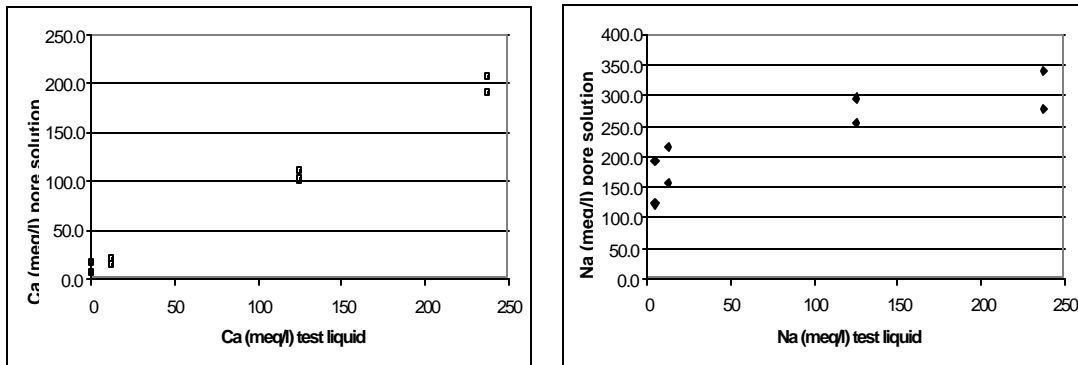


Fig. 5 Measured Ca and Na content in pore fluid (first test) related to composition of applied test fluid

The measured cation exchange capacity does not show significant variation. As the cation exchange capacity is mainly determined by the bentonite fraction in the sample it can be concluded that the composition of the used Trisoplast® samples was identical.

3.4.2 Potential water absorption capacity and internal surface area

The potential water absorption capacity has been determined after completion of the permeability measurements (annex 1). Prior to the swelling tests, samples have been oven dried and the chemical composition of the pore fluid has been determined. The swelling capacity tests were done with oven dry samples and demineralised water. During the water uptake, dilution of the present salts occurs, which has not been accounted for in the presentation of the results. The measured swelling capacity is the combined effect of water uptake by the granular skeleton of the sand and water uptake by swelling clay minerals as well as the polymer. The measured swelling capacity is reduced with the structural water uptake of the sand fraction. Table 6 shows the results.

Table 6 Potential swelling capacity of bentonite and polymer in Trisoplast® during 24 hour

| Sample | Dry bulk (kg/m ³) | Porosity (m ³ /m ³) | Moisture (m ³ /m ³) | Initial conc. (meq/l) | Internal (m ² /gr) | Water (g H ₂ O/g clay) |
|--------|----------------------------------|---|---|--------------------------|----------------------------------|--------------------------------------|
| Tp1,1 | 1650 | 0.38 | 0.31 | 207 | 19.0 | 7.24 |
| Tp3,1 | 1650 | 0.38 | 0.25 | 360 | 18.3 | 7.3 |
| Tp6,1 | 1650 | 0.38 | 0.24 | 396 | 22.3 | 3.15 |
| Tp9,1 | 1650 | 0.38 | 0.32 | 348 | 19.8 | 2.29 |

* reduced by the structural water uptake of the sand fraction

The measured internal surface seems not to change when the swelling capacity reduces. It can, however, not be ruled out that due to the determination method only the surface area of complete dispersed clay is measured, hiding the likely aggregation of clay platelets. The absorbed quantity of water expressed as a thickness of water layer on the area of minerals amounts 500 °A. This thickness is significantly higher than obtained from roentgen refraction methods or theoretical calculations (Bolt, 1955; Norrish, 1954; Schofield 1946 en Verwey en Overbeek, 1948). Rowell (1963) explains this discrepancy:

1. discontinuity in a system consisting of parallel oriented clay platelets, increasing the internal surface significantly;
2. the clay platelets constitute a spherical structure in which water is entrapped;
3. clay platelets connect inert crystals and form a spherical structure with entrapped water;
4. the last two explanations seem the most likely ones because the swelling capacity is significantly reduced when a certain total concentration has been exceeded, which can be attributed to a poor development of the spherical structure of clay and other minerals or even a collapse of this structure

Table 7 Relative water absorption capacity of three mineral barrier materials (g H₂O/g dry matter)

| Na+K (meq/l) | Ca+Mg (meq/l) | Sum anions+cations | SAR | Trisoplast | Sand-bentonite 13% | Sand-bentonite 7.5% |
|-----------------|------------------|-----------------------|------|------------|-----------------------|------------------------|
| 4.75 | 0.25 | 10 | 0.42 | 0.80 | | |
| 4.75 | 0.25 | 10 | 0.42 | 0.71 | | |
| 25 | 25 | 100 | 0.22 | 0.76 | | |
| 25 | 25 | 100 | 0.22 | 0.76 | | |
| 237.5 | 12.5 | 500 | 3.00 | 0.94 | | |
| 237.5 | 12.5 | 500 | 3.00 | 1.00 | | |
| 190 | 16 | 412 | 2.12 | 0.53 | | |
| 158 | 191 | 698 | 0.51 | 0.17 | | |
| 340 | 21 | 722 | 3.32 | 0.53 | | |
| 295 | 101 | 792 | 1.31 | 0.23 | | |
| 4.75 | 0.25 | 10 | 0.42 | | 0.78 | |
| 4.75 | 0.25 | 10 | 0.42 | | 0.84 | |
| 25 | 25 | 100 | 0.22 | | 0.81 | |
| 25 | 25 | 100 | 0.22 | | 0.88 | |
| 237.5 | 12.5 | 500 | 3.00 | | 1.00 | |
| 237.5 | 12.5 | 500 | 3.00 | | 0.84 | |
| 125 | 125 | 500 | 0.50 | | 0.68 | |
| 125 | 125 | 500 | 0.50 | | 0.59 | |
| 12.5 | 237.5 | 500 | 0.04 | | 0.51 | |
| 120.9 | 5.1 | 252 | 2.39 | | | 1.00 |
| 278.6 | 13.31 | 584 | 3.42 | | | 0.85 |
| 256.3 | 110.14 | 733 | 1.09 | | | 0.39 |
| 215.8 | 207.14 | 846 | 0.67 | | | 0.22 |

Measurement of all samples reveal that the potential swelling capacity is rather limited ($< 0.4 \times$ maximum swell) when the initial sum of cations and anions in the pore water exceeds about 500 meq/l. These values comply well with those reported by Rowell (1963). Below about 100 meq/l the effect of the liquid composition on swelling capacity is limited or can be ignored.

The Na/Ca ratio (through the SAR-value) too affects the swelling capacity as is shown in table 7. This table shows that the swelling capacity can be affected by the ratio of monovalent ions over bivalent ions, but effects of electrolyte concentration cannot be ruled out. A limited effect is observed from low SAR-values and limited electrolyte concentration, while at high concentrations effects on swelling capacity is limited when the SAR-value exceeds certain (high) values. A possible complication is that osmotic effects from elevated pore water electrolyte concentrations result in a water absorption capacity, which not fully can be attributed to the swelling behaviour of the clay minerals. In those cases the water absorption capacity of the clay minerals is overestimated. Further investigation of this phenomenon is recommended.

The combined effect of initial salt concentration of the pore liquid and the sodium adsorption ratio is evaluated through a regression analysis according to the model:

$$\text{Relative swelling capacity} = Z_1 \times (\text{sum cations} + \text{anions}) + Z_2 \times \text{SAR} + Z_3$$

Table 8 contains the parameters of this model for both Trisoplast® and sand-bentonite. The high value of R² shows that the model accurately describes this function.

Table 8 Parameters of the regression model: $\text{relative swelling capacity} = Z_1 \times (\text{sum anions} + \text{cations}) + Z_2 \times \text{SAR} + Z_3$

| Material | Z ₁ | Z ₂ | Z ₃ | R ² |
|------------------------|----------------|----------------|----------------|----------------|
| Trisoplast® | -0.00098 | 0.1928 | 0.741 | 0.81 |
| Sand-bentonite 13% | -0.00051 | 0.1207 | 0.813 | 0.81 |
| Sand-bentonite 7.5% | -0.00089 | 0.1417 | 0.886 | 0.99 |

With this model the minimum SAR-value of the pore water has been calculated when at a certain total concentration the swelling capacity should not be affected. The results are shown in fig. 6. This figure shows that the relative water absorption capacity of the bentonite in different barrier materials does not show a significant difference. But the real swelling capacity of the polymer-bentonite at low pore water electrolyte concentrations amounts 10.3 gr water/ gram bentonite-polymer (samples TP1,2 and TP2,2) is higher than of pure bentonite: 8,9 gr/gr (samples SB19, SB 20, annex 1).

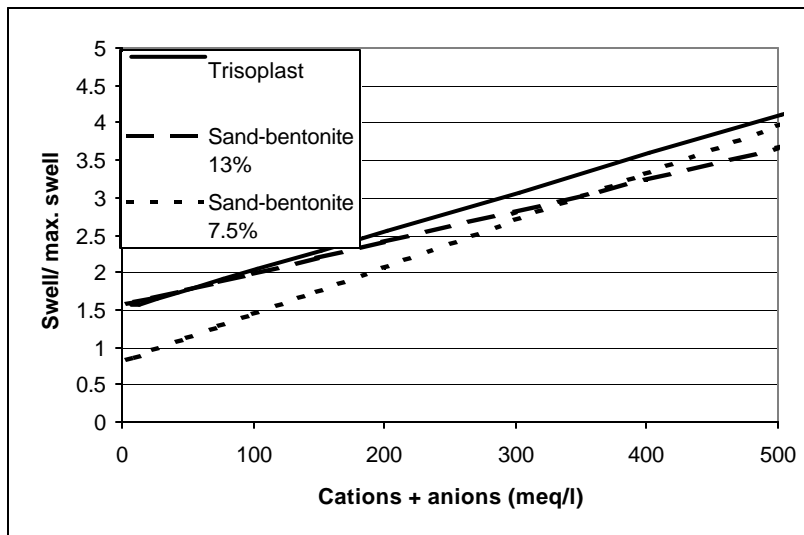


Fig. 6 Required minimum SAR-value to exclude effects on potential swelling capacity

Because of the osmotic effects have not been excluded in the above found relationship, no practical recommendations can be derived for maximum SAR values and values for the maximum sum of cations+anions.

3.4.3 Effect of fluid composition on hydraulic permeability

When the ratio of sodium over calcium concentration remains relatively high (eg. high SAR-values), the permeability is hardly affected by the salt concentration (sum anions+cations). A decrease of this ratio has no serious effects for Trisoplast® when the salt concentration remains low. Increasing of the salt concentration when the Na/(Na + Ca) ratio of the liquid amounts 0.5, has only a limited effect on the permeability of Trisoplast® (fig. 7).

Figure 7 shows the effect of total salt concentration of the pore fluid on the permeability of Trisoplast®, sand-bentonite 13% and sand-bentonite 7.5% bentonite. But the effects are not unique because the SAR-value has a significant influence too. As the permeability depends on the combined effect of salt concentration and the SAR value, relationship is defined through statistical model according to:

$$\text{Log}(K/K\text{-ref}) = A1 * (\text{sum anion+cation concentration}) + A2 * \text{SAR} + A3$$

where:

| | |
|-------------|--|
| K | actual permeability (m/s) |
| K-ref | reference permeability (m/s) (with tap water or low concentration and high SAR-value) |
| A1, ... , 3 | constants |

The constants have been derived from the observations (annex 1) and are presented in table 9. The accuracy of the model is rather high, given the high R2 values. For comparison also data of sand-bentonite are presented.

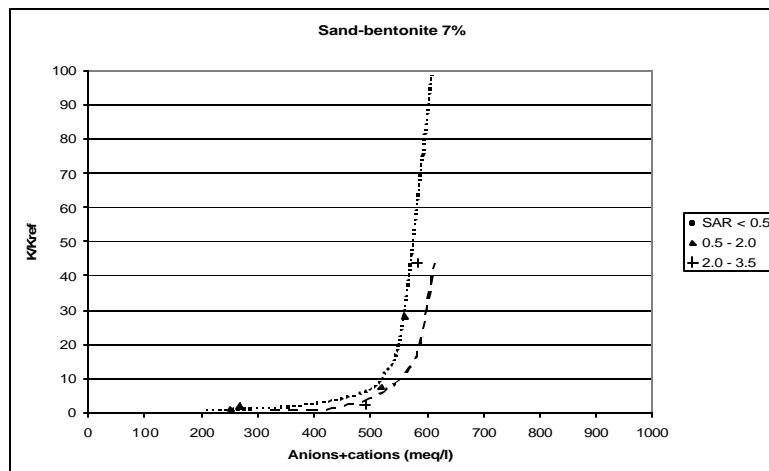
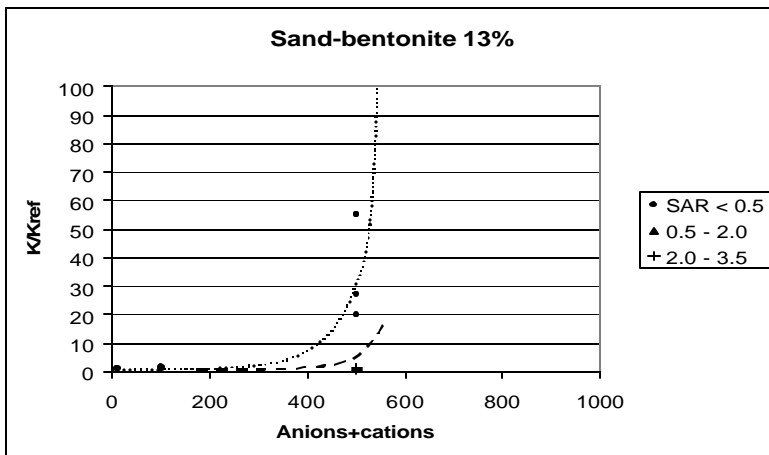
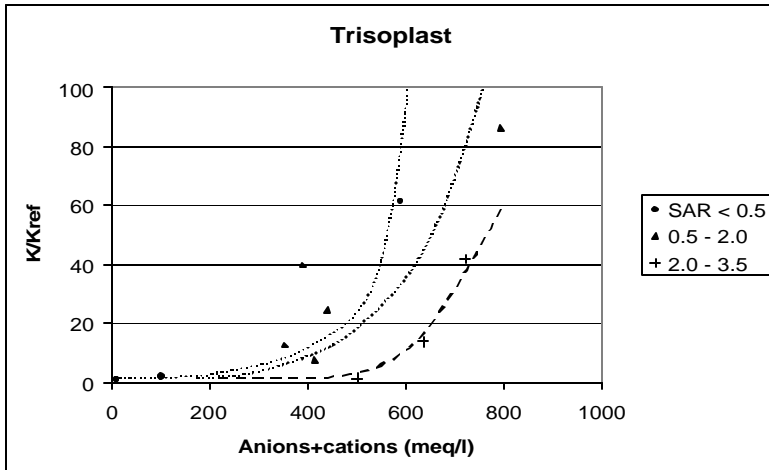


Fig. 7 Effect of total salt concentration and SAR on the permeability

Table 9 Constants in the regression equation:

$$\text{Log}(K/K\text{-ref}) = A_1 * (\text{sum cation+anion concentration}) + A_2 * \text{SAR} + A_3$$

| Material | A ₁ | A ₂ | A ₃ | R ² |
|---|----------------|----------------|----------------|----------------|
| Trisoplast®, all samples | 0.003551 | -0.46632 | 0.202783 | 0.85 |
| Trisoplast®, exposure at 44% saturation (tap water) | 0.00209 | -0.36287 | 0.16954 | 0.95 |
| Trisoplast®, exposure at 100% saturation | 0.00245 | -0.33951 | 0.748057 | 0.80 |
| Sand-bentonite 13%, exposure at 100% saturation | 0.003191 | -0.52304 | 0.436824 | 0.96 |
| Sand-bentonite 7.5%, exposure at 100% saturation | 0.00482 | -0.2374 | -0.39192 | 0.95 |

With the regression model the minimum required SAR-value for each salt concentration can be calculated when the barrier function has to comply with the requirements. The permeability depends on the thickness of the barrier and should have a value to guarantee a leakage rate less than 0.0001 m/d under standard conditions (1 m standing water layer on top of the barrier and free outflow). The standard thickness of Trisoplast® is 0.07m, of sand-bentonite 7.5%, 0.25 m. To evaluate the effect of the polymer, the thickness of the sand-bentonite barrier is fixed to 0.07 m.

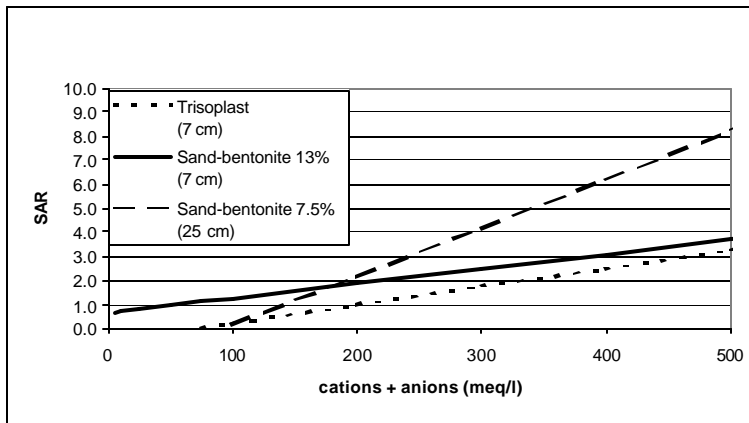


Fig. 8 Minimum SAR-value to meet Dutch legal barrier requirements with Trisoplast®, sand-bentonite 13% and sand-bentonite 7.5% barriers of respectively 7, 7 and 25 cm thickness

The required maximum permeability for these materials is: 0.8×10^{-10} m/s, 0.8×10^{-10} m/s and 2.31×10^{-10} m/s respectively. The reference permeability's (Kref) are 0.26×10^{-10} m/s, 0.60×10^{-10} m/s and 2.0×10^{-10} m/s; so ratio of K/Kref for these three materials may increase to 2.91, 1.26 and 1.16 for respectively Trisoplast®, sand-bentonite 13% and sand-bentonite 7.5%. Figure 8 shows that sand-bentonite 13% performs less than Trisoplast®, which demonstrates the positive effects of the added polymer. The polymer makes Trisoplast® less sensitive to elevated electrolyte (salt) concentrations of the pore water. Sand-bentonite 7.5% is more sensitive for higher salt concentration and requires higher SAR-values (high ratio of sodium over calcium concentration) than Trisoplast®.

3.4.4 Recovery mechanism

The potential swelling capacity experiments have shown that the fragile structure of bentonite and the polymer most likely collapses when the composition of the fluid changes from low to high Ca-contents simultaneously with increasing total salt concentrations. It has been questioned whether or not recovery of such a collapsed structure could be expected when fluids are applied with relatively high sodium concentrations and relatively low total salt concentration.

After completion of the permeability tests samples from series 1 (see annex 1) were selected that were exposed to a test liquid with a relatively high sodium concentration and a total salt concentration of 10 meq/l. This sample has been exposed to a test liquid with relatively high calcium and total salt concentration (95% Ca and 500 meq/l). After several displacements of the pore volume the initial liquid (95% Na and 10 meq/l) was again applied.

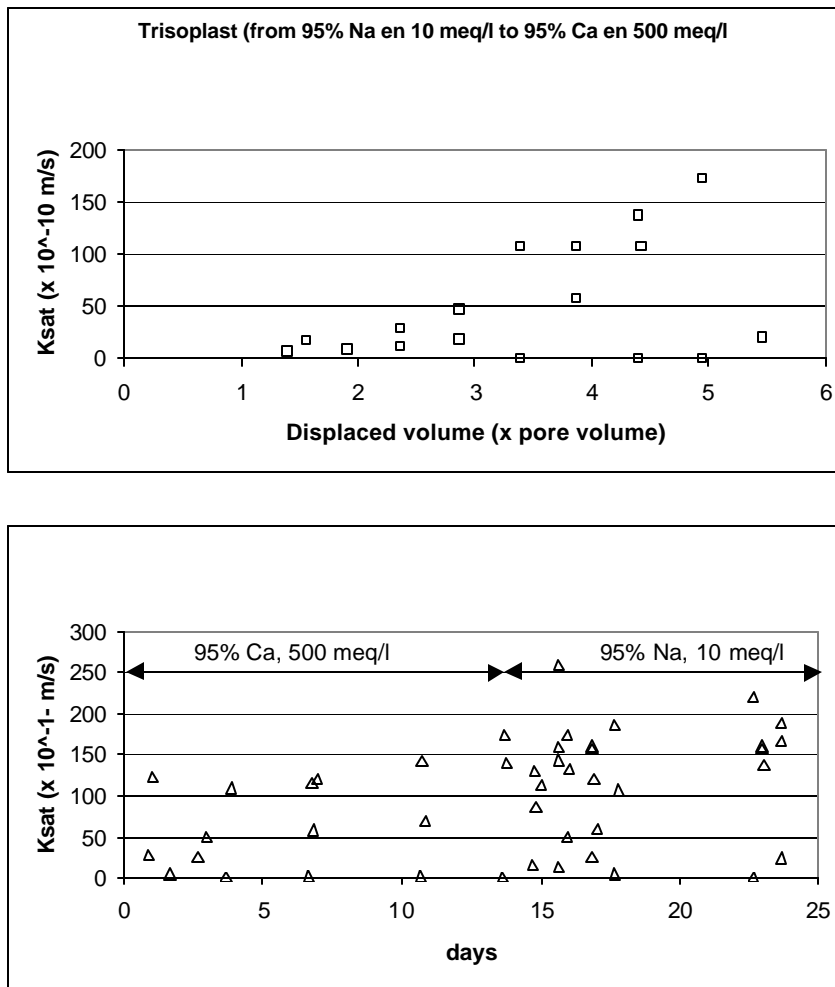


Fig. 9 Recovery test of Trisoplast® first exposed to liquid with high Ca-concentration followed by high Na-concentration

This experiment shows a sharp increase of the permeability to more than $100 \times 10^{-10} \text{ m.s}^{-1}$ when a liquid with a $\text{Ca}/(\text{Ca} + \text{Na})$ ratio of 0,95 and a total salinity of 500 meq/l was applied. Exposing the same sample to a liquid with a $\text{Na}/(\text{Na} + \text{Ca})$ ratio of 0.95 did not recover the originally low permeability. Obviously the spherical structure of clay minerals and polymer has irreversibly collapsed.

The consequence of this observation is that when a barrier has been replaced after its permeability was significantly increased by cation exchange processes, the barrier material should not be reused for constructing a new barrier.

This conclusions is preliminary because the samples were initially prepared with insufficient washed sea sand and the polymer had to develop in an environment with high salt content. It cannot be excluded that the development of the polymer under such circumstances is less than optimal.

3.4.5 Behaviour of Trisoplast® with sea-water

A separate experiment was conducted with seawater. Previous experiments showed a limited effect of seawater on the permeability. These were repeated. First the permeability was measured with tap water prior to exposure to seawater. The permeability after a period of 112 days with tap water, the permeability was $0.3 \times 10^{-10} \text{ m/s}$. Then seawater was exposed and after 30 days measurements were resumed (see fig. 10)

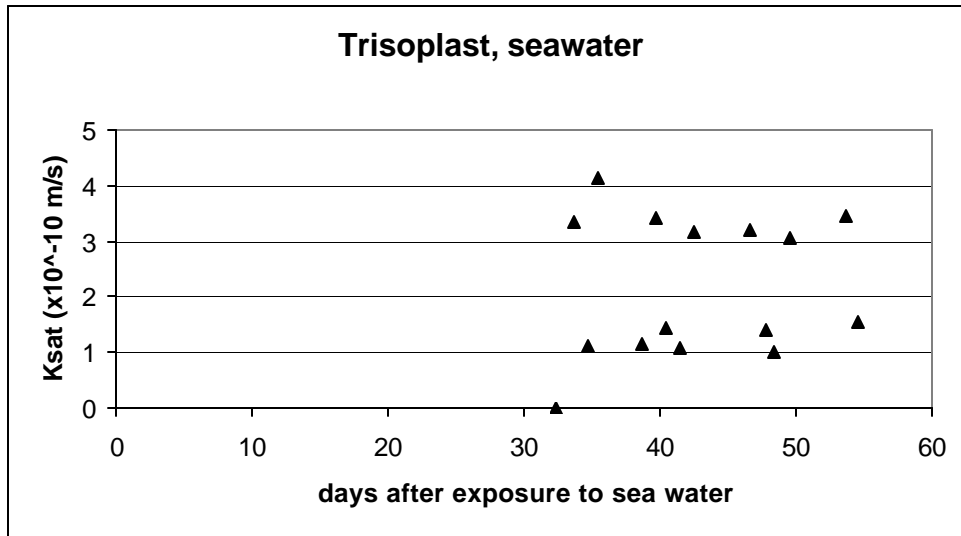


Fig. 10 Permeability of Trisoplast exposed to sea water

The results of the observations are summarized in table 10. This table shows that the (apparent) permeability is related to the hydraulic gradient.

Table 10 Observed gradient, outflowrate and calculated permeability of trisoplast exposed to seawater after fully saturation with tap water

| Gradient | outflow rate (cm/d) | Permeability (*10 ⁻¹⁰ m/s) |
|----------|------------------------|--|
| 23.20785 | 0.020976 | 1.046 |
| 23.80824 | 0.023175 | 1.127 |
| 27.21383 | 0.000202 | 0.009 |
| 27.92213 | 0.038288 | 1.587 |
| 28.1237 | 0.02848 | 1.172 |
| 28.52975 | 0.029578 | 1.200 |
| 30.3998 | 0.037589 | 1.431 |
| 31.53819 | 0.04008 | 1.471 |
| 53.34794 | 0.156726 | 3.400 |
| 54.38882 | 0.146193 | 3.111 |
| 55.20344 | 0.168096 | 3.524 |
| 55.64265 | 0.157058 | 3.267 |
| 55.76032 | 0.155873 | 3.235 |
| 57.26007 | 0.172209 | 3.481 |
| 61.64646 | 0.22313 | 4.189 |

Seawater was applied to one side of the sample, while the other side contained tap water. Under such conditions the water fluxes are controlled by the combined hydraulic and osmotic gradients, provided the material behaves as a semi-permeable wall (Keijzer, 2000). In practise this behaviour is far from perfect. The effective water flux can be described by:

$$J_v = K_{sat} \frac{\Delta p - s \Delta p_o}{d}$$

where:

$$\Delta p_o = \frac{RT}{V_w} \ln \frac{a_w^{low}}{a_w^{high}}$$

- ? p osmotic pressure difference (Pa)
- ? p hydraulic pressure difference (Pa)
(i = ?p/d, when pressure difference is expressed in cm water column)
- R universal gas constant (8.31451 J/K/mol)
- T absolute temperature (Kelvin)
- V_w partial molar volume of water (m³.mol⁻¹)
- a_w activity of water at resp. low and high concentration side of a membrane

$$\ln a_w = -\frac{vmW_w}{1000} f$$

- W_m molar mass of solvent (kg/mol)
 v number of ions in which electrolyte dissociates
 m molality of solution (mol/kg)
 F electrolyte and temperature dependent parameter (~945)
 s reflection coefficient (< 1)
 d thickness of sample (m)

The real permeability of Trisoplast® for seawater is determined by plotting the hydraulic gradient against the outflow rate (fig. 11).

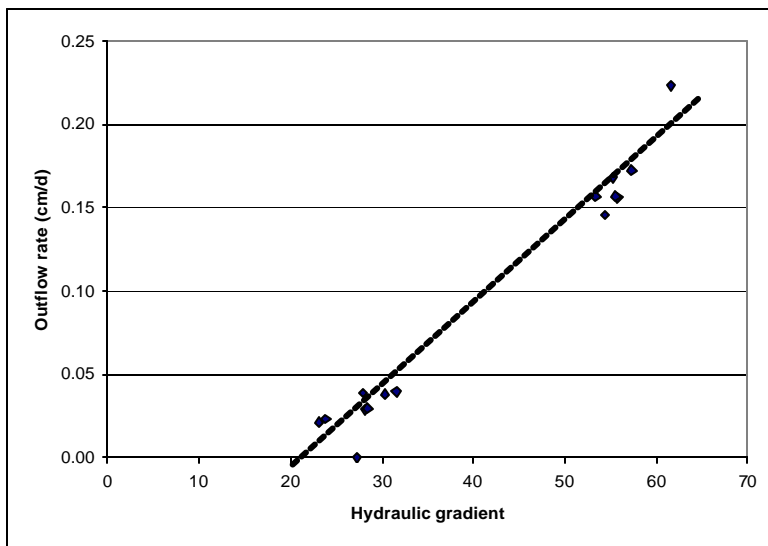


Fig. 11 Outflow rate gradient dependent in test with seawater

The linear regression line for the plotted data is calculated. The slope of this line equals the hydraulic permeability (= 0.00496 cm/d) and amounts 5.725×10^{-10} m/s. The intersection of the regression line with the outflow-rate axis (where $i = 0$ and outflow rate = -0.109271 cm/d) represents the parameter $[(K_{sat} \cdot p)/d]$. The thickness of the sample is 2.5 cm. Seawater contains about 35000 ppm salts, equivalent to a concentration of 0.60 mol/l. So $p \sim 2690$ cm water column. From these data the reflection coefficient has been calculated: $s = 0.0206$.

The characteristics of semi-permeability are attributed to the bentonite and polymer. Trisoplast® contains 13% bentonite by weight, and the total dry bulk density amounts 1650 kg/m^3 . The effective porosity of Trisoplast® in the pores of the sand skeleton is 0.82 ($= 1 - \{0.13 \times 1650 / 2650\} / \{1 - (1-0.13) 1650 / 2650\}$). The calculated reflection coefficient is comparable to the values determined by Keijzer (2000) for Wyoming bentonite with an effective porosity of 0.56 (0.03 when determined from measured fluxes and 0.015 from for pressure changes) and the

highest concentration of 0.1 mol/l. When, however the effective porosity's were less (about 0.65), and the highest concentration amounted 0.6 mol/l, the reflection coefficients this author reported were much lower : 0.0006 - 0.002.

This comparison gives the impression that possibly the polymer has a significant enhancing effect on the semi-permeable characteristics of bentonite. According to Keijzer, the highest concentration has a significant effect on the reflection coefficient. So the lower the concentration of the containment solution, the better the semi-permeability of bentonite is. If the same were true for the bentonite-polymer probably an ideal containment can be realised by keeping the hydraulic gradient in the barrier low. This can be realised by a high quality drainage system above base liners.

4 Calculation of the functional life time of Trisoplast®

4.1 Definition of barrier quality

The quality of a mineral barrier expresses its ability to reduce infiltration of excess of rainfall into the waste body. The quality will be expressed as a fraction of the initial capacity to reduce the infiltration rate directly after installation of the barrier. A crucial parameter for the quality of the barrier is the hydraulic resistance, R_h , which is defined as:

$$R_{h,t} = \int_D \frac{l}{K_{z,t}} dz$$

where:

- D barrier thickness
- $K_{z,t}$ permeability at depth z below the surface of the barrier at time t
- dz depth increment

And the quality of the barrier at a certain time moment is defined as:

$$Quality(t) = \frac{R_{h,t}}{R_{h,0}}$$

In this equation the hydraulic resistance directly after installing the barrier is denoted by $R_{h,0}$. The maximum functional lifetime has been reached when the quality is less than prescribed by the landfill licence. The acceptable quality reduction depends on the initial permeability and the maximum permeability to fulfil the landfill licence requirements. When the initial permeability of the barrier at the moment of installation is much lower than strictly required according to the licence, an increase of permeability does not violate the minimum protection level. When, however, the initial permeability is close to the prescribed one, a limited increase of the permeability of the barrier is sufficient to exceed the minimum protection level.

Table 11 Required permeability ($\times 10^{-10} m.s^{-1}$) directly after installation of the barrier to meet the Dutch quality standard after when the initial quality of the barrier has been reduced

| Material | Thickness (m) | Quality of the barrier (= 1, directly after installation) | | | | | | |
|----------------|---------------|---|------|------|------|------|------|-------|
| | | 1 | 0.8 | 0.6 | 0.4 | 0.2 | 0.1 | 0.01 |
| Trisoplast® | 0.08 | 0.86 | 0.69 | 0.51 | 0.34 | 0.17 | 0.09 | 0.009 |
| Sand-bentonite | 0.25 | 2.31 | 1.85 | 1.39 | 0.93 | 0.46 | 0.23 | 0.023 |
| GCL | 0.01 | 0.115 | 0.09 | 0.07 | 0.05 | 0.02 | 0.01 | 0.001 |

Table 11 gives an overview of the required average permeability of the barrier directly after installation to meet the minimum barrier quality according to legislation in the Netherlands, when the initial quality is reduced to a certain value.

The quality of a Trisoplast® barrier, for example, can safely be reduced till 0.2 (20% of the initial quality) when the permeability directly after installation of the barrier was equal to or less than $0.17 \times 10^{-10} \text{ m.s}^{-1}$.

In practice the permeability of barrier materials amounts about $0.1 \times 10^{-10} \text{ m.s}^{-1}$ for Trisoplast® $1.0 \times 10^{-10} \text{ m.s}^{-1}$ for mixtures of sand and 7.5% bentonite. For these materials minimum acceptable quality under Dutch legislation is 0.12 for Trisoplast® and 0.45 for sand-bentonite 7.5%.

4.2 Functional life time of Trisoplast® barrier

The calculation of the functional life time of Trisoplast® includes the simulation of two processes: (1) changing of the composition of the pore fluid in the barrier material by convection and diffusion and simultaneously by cation exchange (with model EPIDIM) and (2) calculation of the permeability of the barrier related to the composition of the fluid. The second step in the simulation includes the calculation of the hydraulic resistance of the barrier. The actual permeability is derived from 100 the regression model derived from laboratory measurements.

The simulation of changes of the chemical composition of pore fluid in barriers is based on a construction with a geomembrane overlaying a barrier (see also figure 1):

1. a geomembrane;
2. barrier with known initial pore fluid composition;
3. not obligatory sand layer (separation of barrier from aggressive sub layer);
4. a supporting layer (bottom ash) with known composition of pore fluid waste body

and boundary conditions:

1. a know and constant convective flux (= 0 when a geomembrane is present);
2. equilibrium between chemical composition of pore fluid and composition of the adsorption complex
3. constant moisture content in time and space
4. zero diffusion flux from the supporting layer to the waste

The displacement of dissolved matter is based on the combined convective and diffusive transport mechanism and triggers two processes in the barrier:

1. migrated calcium and carbonate ions are replenished from the precipitated quantity;
2. sodium ions are (partly) replenished by exchange of adsorbed sodium against calcium or vice versa

To describe the chemistry of a solution, a set of independent chemical components has been chosen that allow the build up of all the chemical species from this set. No component can be formed out of other components. In general terms, setting up an

inorganic geo-chemical model involves choosing N unknowns and N governing equations. Expressions for the concentrations of the species are formulated according to law of mass action. The chemical system is described by a set of N non-linear algebraic equations with N unknown variables. In a mathematical way, the system is completely determined. The set of equations is solved using a Newton-Raphson iterative technique. To account for a non-ideal behaviour of ions in solutions activity coefficients have been introduced and temperature effects are accounted for.

Cation exchange is the process whereby dissolved cations are exchanged for adsorbed cations fixed to a solid. Different cations compete for a fixed amount of exchange sites. It is assumed that the reactions are time-independent and reversible. The capacity of the adsorbents is fully occupied and the sub-system obeys to the electro-neutrality condition. A number of models are available to describe the exchange relations, each with its own properties. In the programme EPIDIM, which is used to calculate the evolution of the clay complex composition, three of the most well known models have been implemented: exchange relations according to Gaines-Thomas, Vanselow and Gapon. Definitions of the exchange equations can be found in (Rijtema et al.,1998).

Input parameters to calculate the rate of quality reduction of the barrier are presented in table 12.

Table 12 Input data for simulation of barrier lifetime

| Material | Dry bulk density (kg/l) | CEC (meq/kg) | CaCO ₃ (g/kg dry matter) | PH | Na + K (meq/l) | Ca + Mg (meq/l) |
|---------------------|-------------------------|--------------|-------------------------------------|-----|----------------|-----------------|
| Trisoplast® | 1.70 | 108 | 12.4 | 9.8 | 25.3 | 5.52 |
| Sand-bentonite 7.5% | 1.80 | 71 | 15.7 | 9.5 | 20.5 | 4.8 |
| Sand | 1.55 | 38 | 1.2 | 5.6 | 1.77 | 0.085 |
| Bottom ash | 1.50 | 75 | 55 | 8.1 | 151 | 67 |

The change of quality of a Trisoplast® and sand-bentonite (7.5%) barrier overlying a support layer with different compositions (see table 11) is calculated (figure 12 A through D).

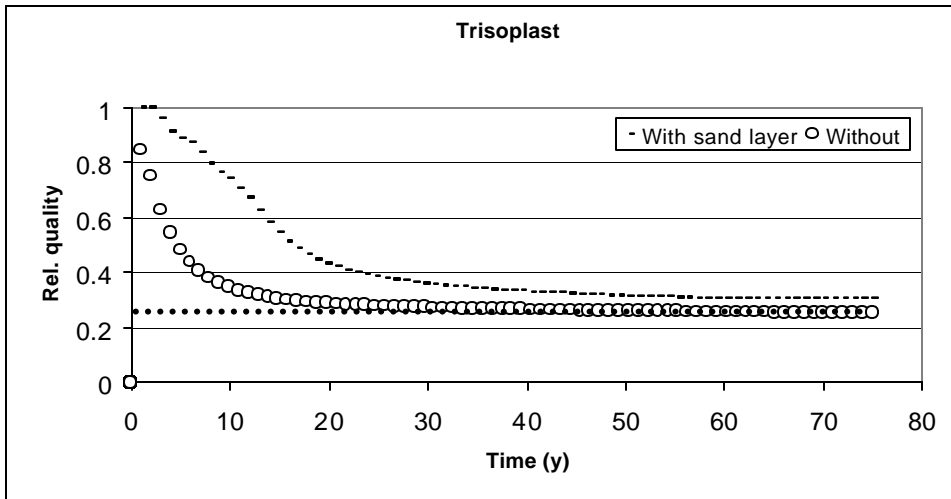


Fig. 12A Trisoplast® barrier directly on bottom ash or separated from this layer with sand; moisture conditions: pF2 ('field capacity')

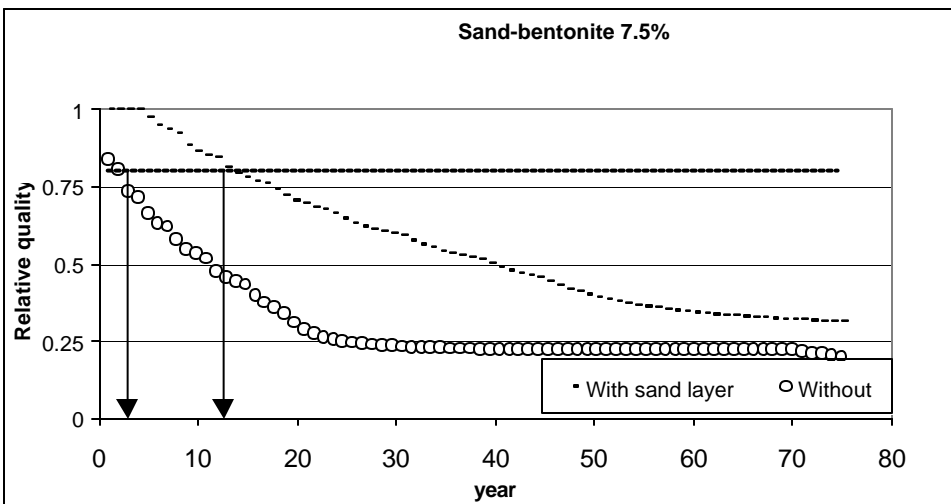


Fig. 12B Sand-bentonite 7.5% barrier directly on bottom ash or separated from this layer with sand; moisture conditions: pF2 ('field capacity'). Functional life time: ~3 years without sand layer and ~15 years with a sand-layer. When the relative quality is higher than 0.85, the barrier complies with legal requirements

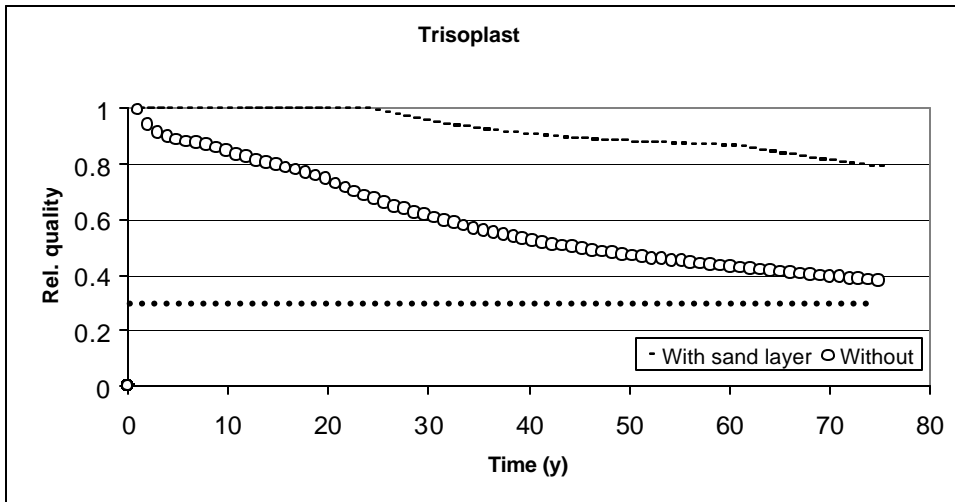


Fig. 12C Trisoplast® barrier directly on bottom ash or separated from this layer with sand; moisture conditions: pF3 ('relatively dry'). When the relative quality is higher than 0.3, the barrier complies with legal requirements

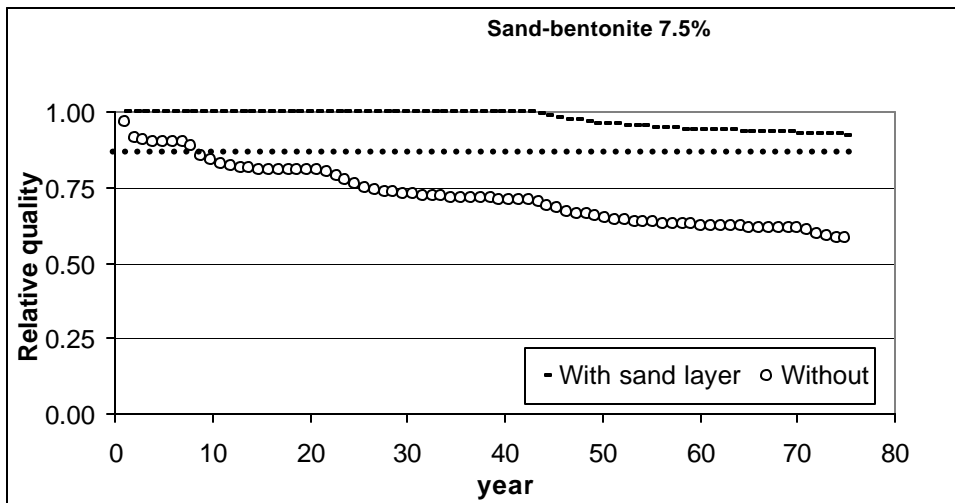


Fig. 12D Sand-bentonite 7.5% barrier directly on bottom ash or separated from this layer with sand; moisture conditions: pF3 ('relatively dry')

Calculations show that bottom ash has a negative effect on the permeability of barriers. The tolerance against this deterioration depends on the initial permeability of the barrier. The rate of deterioration depends on the magnitude of transport of dissolved matter, controlled by the diffusion coefficient. This parameter depends on the saturation degree. Moisture conditions corresponding with field capacity (relatively wet) are worse conditions than the more dry conditions, comparable with pF3 (pore water pressure -1000 mB). A sandy layer between the barrier and the bottom ash provides a good retardation of the transport of dissolved matter.

4.3 Additional measures to prevent quality reduction of Trisoplast®

When no or only a rather limited reduction is acceptable, ageing of the Trisoplast® barrier can be prevented effectively by amending of the top of the supporting layer with a Na-containing material. Calculations have been performed to evaluate the effect of an amendment of $\text{Na}_2\text{CO}_3 \cdot 2\text{H}_2\text{O}$. An example of the change of the quality of a Trisoplast® barrier overlaying a certain supporting layers is given. The composition of this layer has a significant effect on the quality of Trisoplast®, although the required minimum quality is not jeopardised (Fig. 13).

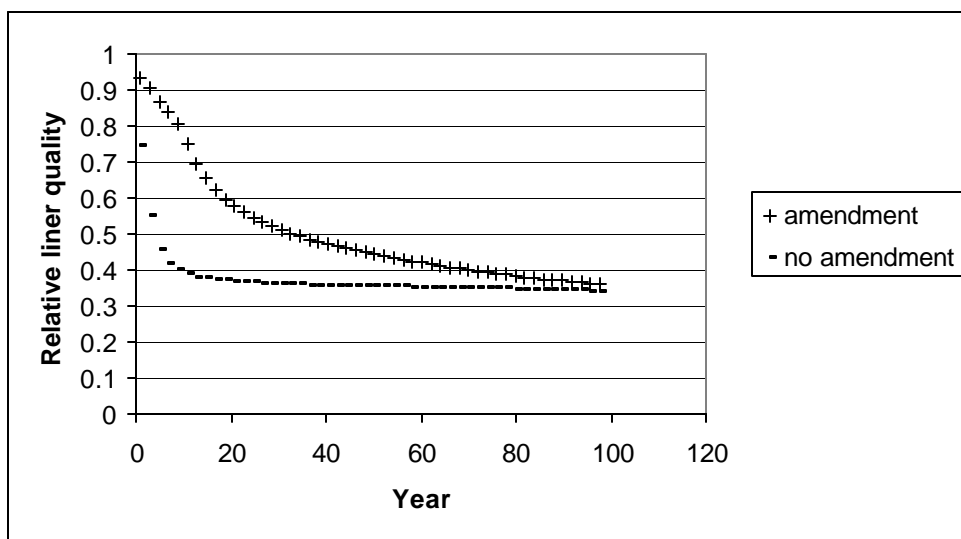


Fig. 12E Quality changes of a Trisoplast® barrier overlaying a certain reactive supporting layer. Sodium amendment applied to the supporting layer

To improve the quality, an amount of $2.7 \text{ kg.m}^{-2} \text{ Na}_2\text{CO}_3 \cdot 2\text{H}_2\text{O}$ has been applied and mixed in the top 0.2 m of the supporting layer. This amendment improves the quality significantly during the first 50 years, but after 100 years the effects are almost vanished. Instead of sodium carbonate, sodium chloride with an equivalent quantity of sodium may be applied.

4.4 How to apply the results

Laboratory analysis and transport simulations have shown that the performance of barriers can be adversely affected. The degree of deterioration and the pace at which this occurs depends on the chemical composition and the moisture conditions of layers adjacent to the barrier.

The following procedure is recommended for future designing of barriers.

1. chemical analysis of the near saturated extract of mineral materials which will come into close contact with barriers (after the method described by Boels and Breen, 2001a);

2. determination of the maximum increase of initial permeability for the situation where the pore water composition of the barrier becomes the same as in the adjacent layers. For this purpose the empirical relationships presented in this paper can be applied or laboratory permeability tests can be conducted with liquid of identical composition (sum cations + anions, SAR-value);
3. designing of the thickness of the barrier based on the maximum expected permeability as sub 2 has been determined (intrinsic safe design) when the expected life time is more than 50 years;
4. designing the thickness of the barrier based on the required initial performance and the needed functional life time (life time of the construction > 50 years).

When laboratory permeability tests are needed, the tests should be conducted by applying test liquids directly to samples. The saturation degree should be comparable to the moisture content during the installation of barriers.

4.5 Conclusions, discussion and further research

The results of the tested samples show that a standard construction of Trisoplast (thickness 0.07 M, bentonite content 13%) is less sensitive to cation exchange than a standard construction of sand-bentonite (thickness 0,25 m, bentonite content 7,5%). The safety factor is high enough to ensure its functionality during extreme long periods even under more severe conditions if the layer is installed directly on bottom ash. All permeability measurements of Trisoplast in series 1 were (by mistake) based on samples produced with badly washed sea sand. It is generally known that salts have an adverse effect on the permeability of mineral barriers, especially if salts are already in the dry mixture from the beginning. In the final stage of this research some comparing measurements with tap water were done on samples produced with normal sand (series 2). The permeability measured appeared to be a factor ten lower than of comparable measurements in series 1 (see Annex 1). The special properties of TRISOPLAST[®] are achieved by the addition of the polymer. The polymer has functional pendant groups, which form strongly sorptive bonding with the clay mineral bentonite, so that a practically irreversible net of polymer and bentonite is created. Bonding of the functional groups to the clay material is accomplished by hydrogen bridges at the clay mineral surfaces, anionic adsorption (especially at the breaking edges of the clay minerals or through polyvalent cations adsorbed on the clay mineral) and cationic adsorption in the clay mineral layers as well as specific adsorption (complex bonding). To build a good spherical structure of clay platelets and to build the network by interactions between the functional groups of the polymer and the clay mineral it is of importance that the bentonite swells very well so the surface of the clay platelets is open to reactions with the polymer. The existence of a high salt concentration already in the dry mixture can block the reactions. The use of sea sand in series 1 probably had a negative influence on the quality of the bentonite-polymer network. It is possible that the added value of the polymer was partly nullified by the initial presence of a high amount of salts in the sea-sand. For this it has been decided to repeat some measurements of series 1 with normal sand and to expand the measurements of series 2 to higher total concentrations and lower

SAR values. Further research is also done on the influence of the vertical load on the barrier, higher/lower bentonite-polymer content and thickness of the layer.

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Appendix 1 Summary of data

(permeability at 10 centi degree, average of duplicate samples. * = water absorption capacity not determined)

| Sample | Bulk density (kg/m ³) | Bentonite (%) | Porewater (meq/l) | | Water ads. cap. (g H ₂ O/g clay) | Permeability (x 10 ⁻¹⁰ m/s) |
|--|--------------------------------------|------------------|-------------------|--------|--|---|
| | | | Na+K | Ca+Mg | | |
| <i>Trisoplast, series 1, saturation and reference permeability with liquid 1 (insufficient washed sea sand used to prepare Trisoplast samples)</i> | | | | | | |
| TP1,1 | 1650 | 13 | 190 | 16 | 7.24 | 1.8834 |
| TP2,1* | 1650 | 13 | 300 | 18 | | 3.3142 |
| TP3,1 | 1650 | 13 | 340 | 21 | 7.30 | 10.147 |
| TP4,1* | 1650 | 13 | 155 | 21 | | 3.0514 |
| TP5,1* | 1650 | 13 | 155 | 65 | | 5.9568 |
| TP6,1 | 1650 | 13 | 295 | 101 | 3.15 | 20.7904 |
| TP7,1* | 1650 | 13 | 155 | 40 | | 9.5776 |
| TP8,1* | 1650 | 13 | 155 | 140 | | 14.8263 |
| TP9,1 | 1650 | 13 | 158 | 191 | 2.29 | 118.6031 |
| <i>Trisoplast, series 2, test liquid directly applied</i> | | | | | | |
| TP1,2 | 1600 | 13 | 4.75 | 0.25 | 10.95 | 0.2847 |
| TP2,2 | 1600 | 13 | 4.75 | 0.25 | 9.70 | 0.2409 |
| TP3,2 | 1600 | 13 | 25 | 25 | 10.40 | 0.4964 |
| TP4,2 | 1600 | 13 | 25 | 25 | 10.42 | 0.4599 |
| TP5,2 | 1600 | 13 | 237.5 | 12.5 | 12.97 | 0.3066 |
| TP6,2 | 1600 | 13 | 237.5 | 12.5 | 13.74 | 0.3358 |
| <i>Sand-bentonite 13%, start with tap water saturation</i> | | | | | | |
| SB 6,2 | 1650 | 13 | 237.5 | 12.5 | 10.95 | 0.6059 |
| SB 19 | 1650 | 13 | 4.75 | 0.25 | 8.57 | 0.584 |
| SB 20 | 1650 | 13 | 4.75 | 0.25 | 9.25 | 0.6205 |
| SB 21 | 1650 | 13 | 25 | 25 | 8.90 | 0.584 |
| SB 22 | 1650 | 13 | 25 | 25 | 9.61 | 0.803 |
| SB 23 | 1650 | 13 | 237.5 | 12.5 | 9.22 | 0.7811 |
| SB 28 | 1650 | 13 | 125 | 125 | 7.50 | 11.7019 |
| SB 33 | 1650 | 13 | 125 | 125 | 6.43 | 15.9067 |
| SB 34 | 1650 | 13 | 12.5 | 237.5 | 5.58 | 32.3171 |
| <i>Sand-bentonite 7.5%, start with tapwater saturation</i> | | | | | | |
| SB2 | 1650 | 7.5 | 120.9 | 5.1 | 6.65 | 2.8908 |
| SB4* | 1650 | 7.5 | 240 | 5 | | 6.9642 |
| SB6,1 | 1650 | 7.5 | 278.6 | 13.31 | 5.65 | 125.56 |
| SB8* | 1650 | 7.5 | 130 | 5 | | 4.8326 |
| SB10* | 1650 | 7.5 | 230 | 30 | | 22.2285 |
| SB12 | 1650 | 7.5 | 256.3 | 110.14 | 2.61 | 1198.66 |
| SB14* | 1650 | 7.5 | 125 | 9 | | 5.4385 |
| SB16* | 1650 | 7.5 | 230 | 50 | | 81.395 |
| SB18 | 1650 | 7.5 | 215.8 | 207.14 | 1.49 | 8119.06 |

