

PERFORMANCE TESTING OF A SELF-FORMING & SELF-REPAIRING SEAL

P.A.J.P. Cnubben
A. van Zomeren

Revisions		
A		
B		
Made by:	Approved:	ECN-Clean Fossil fuels
A. van Zomeren	R.N.J. Comans	
Checked by:	Issued:	
G.D. Roskam	C.A.M. van der Kleijn	

Acknowledgement/Preface

This research is conducted with financial support of the Dong-Ah construction company under supervision of Dr. Jin Sung Ki.

CONTENTS

SUMMARY	5
1. INTRODUCTION	7
1.1 Background and rationale	7
1.2 Objectives	7
1.3 Envisioned results	7
2. MATERIALS AND METHODS	8
2.1 Matrix materials and reactant materials	8
2.2 Materials characterization	8
2.3 Grain size distribution	9
2.4 Preparation procedure of top and bottom layer recipes	12
2.5 Preparation procedure of permeability measurement	12
3. RESULTS AND DISCUSSION	14
3.1 Permeability and permeability development	14
3.2 Leakage of the SF&SR system	16
4. CONCLUSIONS	17

SUMMARY

A Self-Forming & Self-Repairing seal system (SF&SR) has been developed by the geotechnical research team from Dong-Ah Construction Industrial Co. Ltd. The system is to be applied as a top isolation for landfills. The seal system is based on the formation of a CaCO_3 and $\text{CaO}\cdot\text{SiO}_2$ precipitate in the pores on the interface of the two adjacent layers (top layer and bottom layer) of the SF&SR system. The SF&SR system consists of two layers placed upon each other, each layer has a specific composition. The reactive agents present in the top layer and bottom layer are transported to the interface of both layers to form a precipitate which in time will seal the pores on the interface. This will cause an increasing resistance to water percolation. A crack of the seal will result in restart of the precipitation reaction thereby demonstrating the self-repairing properties of the SF&SR system.

The permeability and permeability development with time of the SF&SR seal were measured using a permeability measurement instrument based on the falling head method. The starting permeability of the SF&SR system was low ($1.5 \cdot 10^{-10}$ and $1.8 \cdot 10^{-10}$ m/s) and transport of the reactants towards the precipitation zone was probably controlled by diffusion rather than by percolation processes due to this low permeability. This has also resulted in a somewhat slower formation of the SF&SR seal than in situations where the reactants were transported by percolation.

After cracking the seal, the permeability increased to a value of approximately the starting permeability. In the period after cracking the seal the permeability was monitored again and it was clear that the system has self-repairing properties. The permeability decreased again with approximately the same speed as before cracking the seal.

1. INTRODUCTION

1.1 Background and rationale

The geotechnical research team from Dong-Ah Construction Industrial Co. Ltd. has developed a Self-Forming & Self-Repairing seal system (SF&SR) to be applied as a top isolation for landfills. The seal system is based on the formation of a CaCO_3 and CaO.SiO_2 precipitate in the pores on the interface of the two adjacent layers (top layer and bottom layer) of the SF&SR system. The SF&SR system consists of two layers placed upon each other, each layer has a specific composition. The reactive agents present in the top layer and bottom layer are transported to the interface of both layers to form a precipitate which in time will seal the pores on the interface. The precipitation reaction in the pores on the interface of the top layer and bottom layer will result in an increasing resistance to water transport through the system, thereby forming an effective barrier to water infiltration, with self-forming and self-repairing properties. Due to its intrinsic properties the SF&SR system will form itself. A crack of the seal due to for instance mechanical stress will result in restart of the precipitation reaction thereby demonstrating the self-repairing properties of the SF&SR system.

1.2 Objectives

The aim of this project is to determine the initial permeability, the permeability development with time and the self-repairing ability of the SF&SR seal as defined by Dong-Ah.

1.3 Envisioned results

The envisioned results of the proposed research are:

- Determination of the permeability and consequent permeability development of the SF&SR recipe developed by Dong-Ah.
- Determination of the self-repairing properties of the SF&SR recipe, and the effect on the permeability development.

2. MATERIALS AND METHODS

This chapter describes the basic properties of the applied matrix and reactant materials. The resulting properties of the recipes for the top and bottom matrix as prepared from the matrix and reactant materials, as well as the recipe preparation procedure and processing characteristics are reported. Finally the permeability development of the mixture is measured and monitored and the self-repairing properties are tested.

2.1 Matrix materials and reactant materials

Reactant material mixtures

The reactant materials were provided by the Geochemical Research Team of the Research Development Center of Dong-Ah Industrial Construction Co. Ltd. The two reactant material mixtures were named **CT** (Top layer component mixture) and **CB** (Bottom layer component mixture). Both CT and CB were prefabricated by mixing the pure CT and CB reactants with fine sea sand at some known weight ratio.

Matrix materials

No matrix materials were provided, in consultation with Dong-Ah a suitable matrix material was defined. In this case, after consultation, a clayey like type of soil was used for the experiments.

Top and bottom layer recipes

The Top and Bottom layer recipes used in the Self Forming and Self Repairing recipe have been prepared using a composition ratio of 10 mass % of the reactant mixture (CT or CB reactant mixture) and 90 mass % matrix material (clayey sand) on dry mass basis.

2.2 Materials characterization

In order to design a proper SF&SR top and bottom layer recipe, some characteristic properties of the reactant materials, the matrix material and the resulting top and bottom layer mixtures were characterized. The measured properties were the pH-value, moisture content, soluble components, grain size distribution of the reactant mixtures (CT and CB) and of the matrix material. Another important property in application of the top and bottom layer mixtures under field conditions is the optimum density. The optimum density of the material mixtures have been determined using the Proctor test method.

Basic characteristics

In table 2.1 the characteristic properties of both the CT and CB reactant mixtures are given. The pH and conductivity were measured at a liquid to solid ratio (L/S) of 10 l/kg after 15 and 60 minutes in a suspension with demineralized water. The dry matter content and moisture content were determined after drying the mixtures for 24 hr at a temperature of 105 °C. The amount of solubles (soluble components) in the mixtures was determined by suspending CT and CB in demineralized water at L/S=10 l/kg for 60 min, filtering the suspension, drying the retentate at 105 °C during 24 hr and back weighing the dried material.

Table 2.1 *Characteristic properties of CT and CB component mixture*

Property	Remarks	Unit	CT component mixture	CB component mixture
pH	L/S=10*, 15 min	[- log H ⁺]	10.23	12.54
pH	L/S=10*, 60 min	[- log H ⁺]	10.23	12.55
Dry matter		[% m/m]	88.08	99.91
Moisture		[% m/m]	11.92	0.09
Conductivity	L/S=10*, 15 min	[mS/cm]	9.73	9.45
Conductivity	L/S=10*, 60 min	[mS/cm]	9.69	9.6
Solubles		[% m/m]	18.75	0**

* The pH and conductivity of the mixture are measured in suspension at a liquid to solid ratio (L/S) of 10 l/kg after 15 and 60 minutes.

** The amount of solubles in this sample was too low to be measured gravimetrically. It is for sure that a small fraction of the CB component is soluble.

2.3 Grain size distribution

The grain size distribution was measured after drying the materials under investigation at 105 °C during 24 hr. After drying, the materials are sieved using a sieving tower with 8 different grain size classes ($d > 4$ mm, $2 < d < 4$ mm, $1 < d < 2$ mm, $0.71 < d < 1$ mm, $0.5 < d < 0.71$ mm, $0.2 < d < 0.5$ mm, $0.1 < d < 0.2$ mm and $0.1 < d$ mm).

For CT and CB the grain size distribution was measured for two situations (1) after drying and (2) after suspension in water (L/S=10 l/kg) followed by filtering and drying of the retentate. In this way information is obtained about the distribution behavior of the soluble components in the reactant mixtures. The grain size distribution of the clayey sand matrix was measured after drying (24 hr, 105 °C). The grain size distribution of the CT and CB reactant mixtures as well as the grain size distribution of the used clayey soil are depicted in the three figures below (figures 2.1, 2.2 and 2.3).

Interpretation of the grain size distribution

Looking at the grain size distribution of CT (figure 2.1) under dry conditions, about 75% of the mixture is smaller than 0.1 mm. Approximately 17% has a grain size between 0.1 mm and 0.2 mm and about 5 % is between 0.2 mm and 0.5 mm. After leaching and drying of the mixture it can clearly be seen that the amount of grains smaller than a size of 0.1 mm has decreased to 50% and the grains between 0.1 mm and 0.2 mm have decreased to about 8%. However the mass percentage of the grains with a dimension larger than 0.5 mm has increased significantly. This feature will be of major importance when mixing the components with the matrix material and adding the water to reach the desired moisture content. When leached, about 19% of the mass will be solubilized (see table 2.1). However after filtering and drying some coagulation of small sized grains has occurred resulting in larger grains, as can clearly be seen in figure 2.1.

In practice the matrix materials used are moist. When mixing the moist matrix material with the 'dry' reactant mixture (or separate materials) the reactant will coat to the surface of the larger matrix material grains. In this way a large part of the reactants needed will be inhomogeneously distributed in the mixture causing a less efficient recipe. In the laboratory, the materials are therefore mixed dry to obtain a homogeneous mixture. Water is added after thoroughly mixing to avoid the described coating of the larger particles in the mixture.

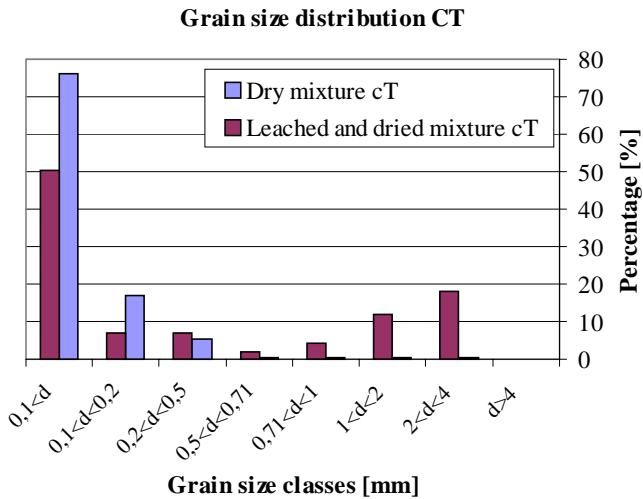


Figure 2.1 Grain size distribution of CT

For CB the grain size distribution under dry and leached/dried conditions are equal, as is shown in figure 2.2. No solubilization of reactant material is observed (also see table 2.1). However when mixed together with moist matrix materials, as will be the case in practice, some immobilization of required reactant material can be expected similar to the situation for CT.

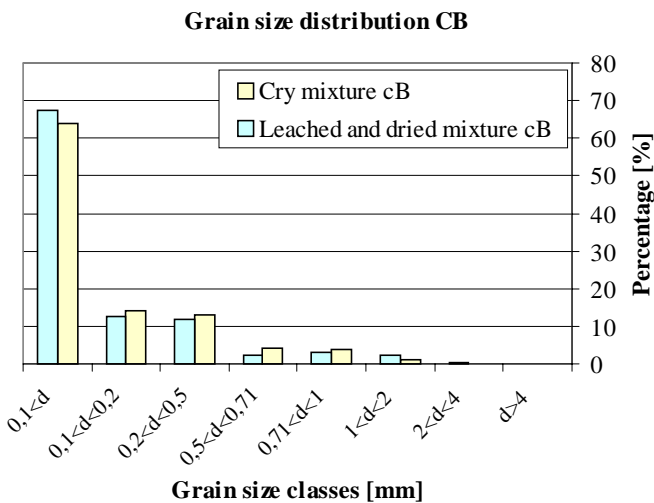


Figure 2.2 Grain size distribution of CB

The mixing behavior of CT and CB with the chosen matrix material is of major importance for the performance of the SF&SR seal under real size conditions. It is stressed that this aspect needs to be studied more thoroughly in a later stage of the investigations. Especially when a pilot scale test is aimed at.

In figure 2.3 the grain size distribution of the matrix material is depicted. As can be seen in the graph the matrix can be characterized as clayey sand. The matrix material predominantly consists of particles smaller than 0.5 mm. The distribution is such that the mixture of the matrix material with the CT or CB reactant mixture will possess a very dense structure under dry conditions. However the interaction between the matrix material when moist (under practical conditions) will be such that the ‘fines’ from the reactant mixtures will coat the larger particles of the matrix material. This phenomenon will ‘immobilize’ a large portion of the reactant materials.

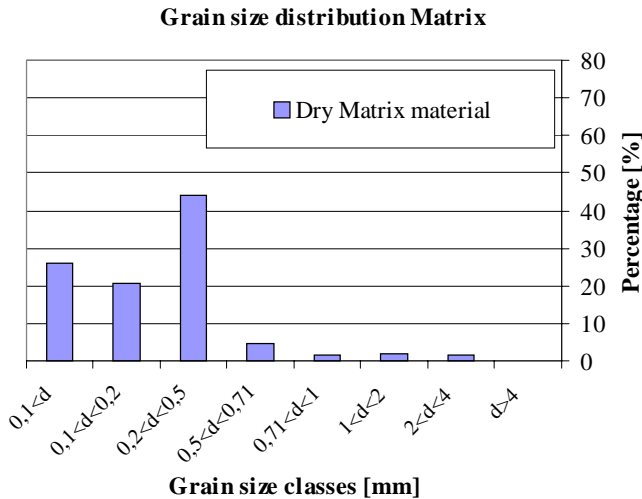


Figure 2.3 *Grain size distribution of Matrix material*

Optimum density

If the pores of a material are totally filled with water further compaction of the material is barely possible. The maximum compaction of soils will be reached if all remaining air is removed from the soil. In this situation the pores will be filled with water. (The limit value for the dry density given as a function of the moisture content is defined by the saturation curve of a material). The moisture content at which the soil is fully compacted is of major importance for the final density to be reached. The possibility to compact a sealant is checked on laboratory scale using the Proctor test. This test determines the moisture content at which maximum compaction is reached after application of a standard amount of energy to the sample.

The Proctor moisture content is determined using the following procedure: A mixture of 10% CT (top reactant mixture) and 90% matrix material is prepared by mixing the pre-dried materials using a Hobart mixer. Because the Proctor moisture content is expected to be about 20%, sufficient water is added whilst mixing to reach this moisture content. A Proctor specimen to determine the optimum density is prepared by compacting the ‘moistened’ mixture in a Proctor mold (dimension; diameter 0.1016 m, height 0.1164 m) equipped with an extension collar (dimension; diameter 0.1016 m, height 0.060 m). The test procedure in short, is as follows: The mold is filled with five layers of test material. The layers are of equal thickness. Each layer is compacted separately by application of 25 uniformly distributed blows from a rammer, with specified dimension (diameter 0.0508 m) and weight (2.5 kg). The rammer is dropped free from a height of 0.305 m. After the 25 blows have been applied to the layer the next layer is applied on top of the compacted layer. This is repeated until the five layers have been applied and compacted. After compaction, the extension collar is removed and the compacted soil is trimmed carefully. The mold is weighed so that the wet density is known and a sub sample for determination of the moisture content is taken. This procedure is repeated by increasing the moisture content of the material sample with an increment of the moisture content of about 1-2 % until the optimum density (Proctor density) is reached.

The optimum density for the mixtures using the Proctor test protocol is reached at a moisture content of 23% and a Dry density of 1577 kg/m³ and a Wet density of 1938 kg/m³. The results of the Proctor measurement are presented in figure 2.4. The results are regarded to be representative for both mixtures.

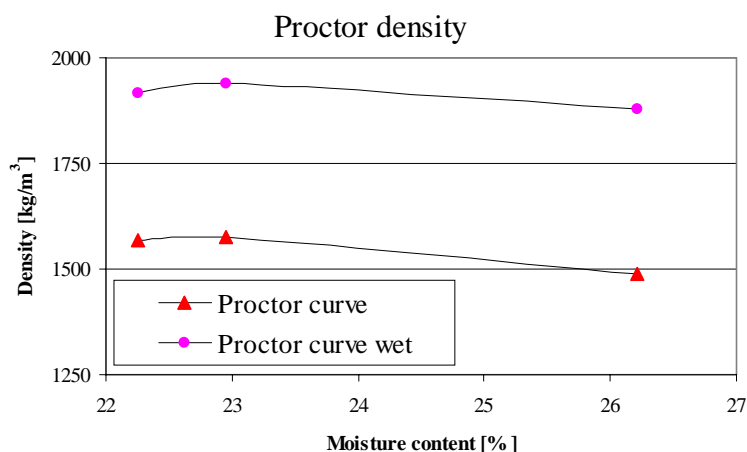


Figure 2.4 *Optimal density measurement for the matrix material (clayey soil)*

2.4 Preparation procedure of top and bottom layer recipes

The preparation is equal for both layers. All materials involved are dried (24hr, 105 °C) and subsequently mixed.

First the dry matrix material is put into the mixing bowl of the Hobart mixer (type Hobart N50, mixing bowl volume 4.5 l and diameter 0.20 m) and either the top (CT) or bottom reactant material (CB) is added slowly to the matrix material, whilst mixing (rotational frequency during mixing 120-180 rpm, mixing arm radius 0.14 m). After the reactant material (CT or CB) is added to the matrix the materials are mixed intensively during 1 minute until the mixture is thoroughly homogenized. Subsequently a specified amount of water is added slowly during mixing until the predetermined Proctor moisture content is reached. The mixing procedure is continued for 5 minutes in order to reach maximum homogenization. The mixed top and bottom layer recipes are removed from the mixing bowl and placed opposite to each other for the permeability measurement.

2.5 Preparation procedure of permeability measurement

The permeability and permeability development with time of the SF&SR seal are monitored using the falling head method. The SF&SR system consists of a top and bottom layer recipe which are placed opposite to each other in a specially designed and fabricated permeability measurement cylinder. After application of the two recipe layers the self forming and self repairing reactions start. Connecting the filled permeability cell to the permeability measurement instrument enables monitoring of the permeability and permeability development with time.

The measuring cylinder consists of two concentric perspex tubes placed inside each other. The inner tube is perforated. A flexible latex membrane is applied on the inside of the inner tube. After application of the bottom recipe layer in the inner perspex tube the bottom recipe layer is placed opposite to the top recipe layer. Both layers are mildly compressed in order to avoid formation of air and or water lenses. The layers are fixed inside the inner tube using a permeable glass frit at the top end and bottom end of the tube. The now prepared inner tube is mounted within the outer tube using a perspex top cover and a perspex bottom cover provided with rubber sealing rings. The covers are connected to the inlet and outlet of the falling head measuring unit (permeability measurement instrument). Water pressure is applied via the bottom of the cylinder. This design requires that the top recipe layer is situated beneath the bottom recipe layer. This 'reversed' measurement situation enables remaining air bubbles in the

mixture to be removed effectively during the saturation phase of the permeability measurement. The water is transported through the SF&SR seal system using hydraulic pressure (in these experiments 21 psi) as the driving force. At the top end of the cylinder water that passed through the SF&SR system is drained to the outlet. The outer cylinder is connected to a confining pressure unit, a pressure of 27 psi was applied in the experiments. Via this connection a confining pressure is applied radial to the perforated inner tubing and the flexible latex membrane. By applying a confining pressure perpendicular to the direction of flow in the inner cylinder (with the SF&SR system) leakage of water in radial direction is eliminated. Thereby only axial water transport is enabled and contribution of side effects (leakage) on the permeability measurement is eliminated. After saturation of the SF&SR system the permeability measurement can be started. Measurements of the inlet- and outlet flow of water at a certain axial pressure (in our case 21 psi) through the SF&SR system enables the calculation of the permeability of the SF&SR seal at any point in the measurement interval. Measurements are done by taking the readings of the water volume that passed the cylinder at different times. Due to the low permeability of the system, one permeability measurement was performed in a timeperiod of about one week.

Permeability measurements are performed at selected time intervals. In this way the permeability development can be monitored closely. Because the measurements are performed over longer periods of time the end permeability can be determined in a precise way. After the permeability has dropped at least one order of magnitude (compared to the initial permeability) the self repairing properties are studied. This will be done by cracking the sealing layer. A cracked sealing layer will have minimum resistance to water transport and the permeability will increase. Due to the self repairing properties of the SF&SR system the sealing layer will form again. Consequently the permeability will decrease demonstrating the self repair.

At a formation time of 130 days the formed seal is cracked. Therefore the measuring cell is opened and the upper layer of reactant material is carefully removed until approximately 0.5 cm above the SF&SR seal. The seal is visible through the flexible latex membrane by a different color. Next, 5 holes (diameter 1.5 mm) are drilled through the SF&SR seal and the reactant material is put back into the measuring cell and the cell is closed again.

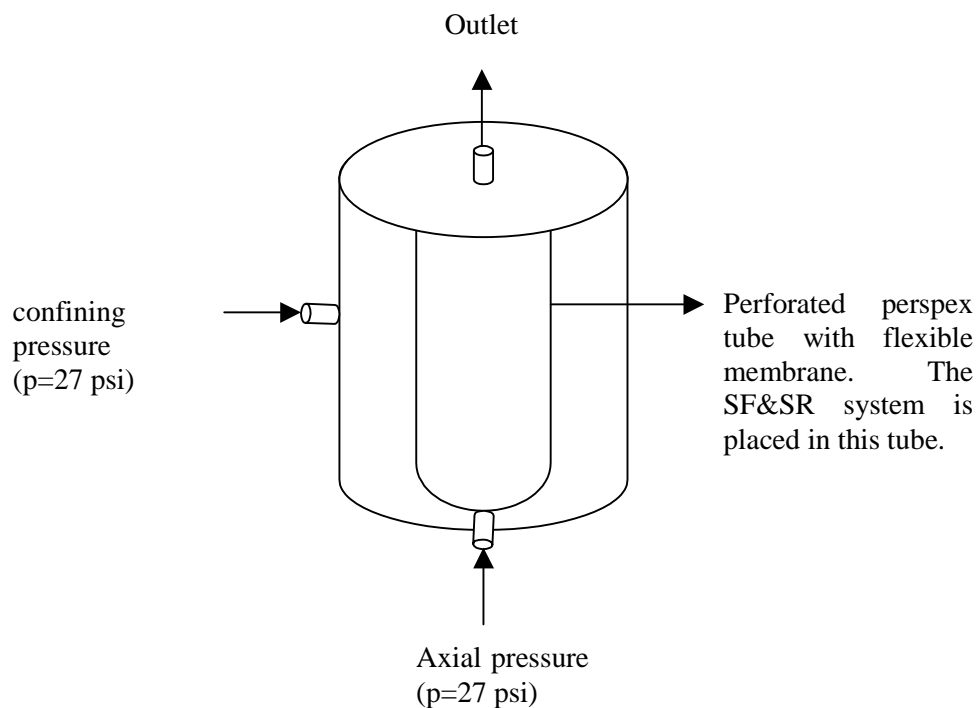


Figure 2.5 Schematic representation of the permeability measuring cylinder

3. RESULTS AND DISCUSSION

3.1 Permeability and permeability development

Due to its character the permeability of the SF&SR seal will decrease in time. The reactions forming the precipitate in the pores on the interface of the top and bottom SF&SR layer starts immediately. The precipitation reactions will continue until the pores are clogged, the sealing layer is formed and maximum resistance to water transport and consequently minimum permeability, is reached. The permeability measurements are performed in duplicate.

Permeability development

The permeability is also referred to as being the hydraulic conductivity. The permeability development of the described recipes is presented in the graphs below (Figures 3.1 and 3.2). On the x-axis the time of formation (days) is presented whereas on the left y-axis the permeability (K_{10}) is given. The unit of permeability, K_{10} , is given in m/s. The index 10 refers to the reference temperature being 10 °C. The right y-axis indicates to the external water pressure applied to the system. The water pressure is given in cm H₂O column. When comparing the permeability of a conventional sealing with a SF&SR system one must bare in mind that the permeability of a SF&SR system is dependent on the time of formation passed from the initial measurement. The seal is formed by the reaction of the active components from both (or multiple) reactant layers. With conventional systems the permeability is a material constant. The formation of the SF&SR seal is continuing in time and it is of major importance to monitor this development over longer periods of time.

As can be observed from both graphs (figure 3.1 and 3.2) the permeability is decreasing in time. Both cells are prepared simultaneously and behave more or less identical in the permeability measurements. After cracking of the SF&SR seal, one of the measuring cells was leaking and further measurements were not possible. Water from the confining compartment (radial pressure applied to the system, see chapter preparation procedure of permeability measurement) was leaking into the measuring compartment, thereby disturbing the permeability measurement. The exact numerical values for some selected permeabilities are given in the tables 3.1 and 3.2.

Clayey sand/CT 90/10 - Clayey sand/CB 90/10

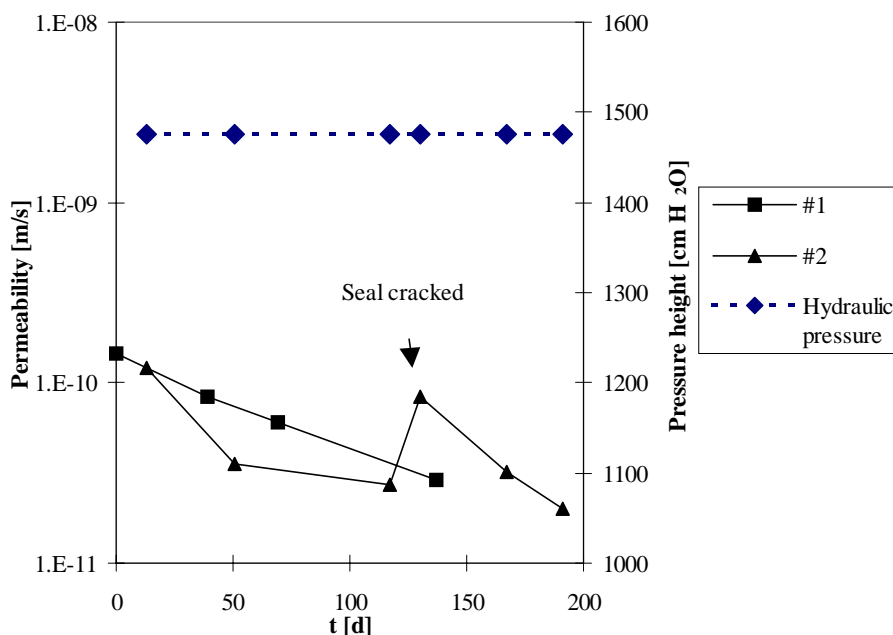


Figure 3.1 Permeability development of the two fabricated cells as a function of time. The first measuring cell was leaking after cracking of the seal and no further measurements could be performed on this cell.

Interpretation of the permeability development

The starting permeability is low with a level of approximately 10^{-10} m/s. This is due to the fact that the used soil type possesses some clayey features. The resulting sealing reactions, which are visually identified in the cells, are relatively slow (a precipitation layer is formed). After about 80-100 days the permeability has dropped over one order of magnitude (approximately 10^{-11} m/s). As can be seen from figure 3.2, the permeability increases after cracking of the seal to approximately the starting permeability of the SF&SR system. The self-repairing properties of the seal are clearly demonstrated from figure 3.2, the permeability decreases in about two months to the permeability level of before the cracking of the seal. In this period, diffusion and/or percolation processes in the system will take care of the supply of new reactant material towards the cracked seal. This will cause the permeability to decrease over time. The last measured datapoint in Figure 3.2 suggests that the permeability is at a minimum. Further measurements are needed to validate this finding.

The slow formation process compared to previous experiments performed at ECN (with other recipes) can be explained by the fact that the used matrix material possesses some clayey features and therefore the system has a relatively low starting permeability. Possibly, the low amount of solubles in the CB mixture can also play a role in the reaction time of the SF&SR system. The transport of the reactants towards the precipitation zone is then controlled by diffusion rather than by percolation processes. However the formation and self-repairing process of the recipe is demonstrated in this way, and the reached permeability levels with respect to Korean legislation are promising.

3.2 Leakage of the SF&SR system

The resulting leakage of the tested SF&SR-systems are compared with the leakage requirements set for the top cover of a waste disposal in The Netherlands. There are two types of seal types, the first type of seal is a mineral type of seal (for instance bentonite), the second type of seal is a combination of a mineral seal with a plastic seal. Both have different leakage requirements which are 0.1 mm/day for the mineral seal and 0.025 mm/day for the combined seal type at a pressure of 1 meter water column (on year basis this is equivalent to respectively 20 mm/year and 5 mm/year with an effective year of 200 days).

Calculation of the leakage

The leakage, v [mm/day] is calculated using the measured permeability according to the formula $v = K * I$. The term I is the ratio of the pressure height of the water column (P_{H_2O})

and the height of the measuring cell (H_{cel}) according to $I = \frac{P_{H_2O}}{H_{cel}}$, for a water column of 1

meter. For higher water column pressures a correction ($dH = \frac{I * H_{cel}}{100}$) must be performed by

dividing the leakage with dH . Eventually, this results in: $v = \frac{K_{10} * 100}{H_{cel}} * 86400 * 1000$

[mm/day] for the calculation of the leakage from the measured permeability for a situation with a water column pressure of 1 meter height.

The calculated leakage data from the measured permeability results are given in the tables 3.1 and 3.2. Calculation of the measured permeabilities into leakage data and comparison with the Dutch standards shows that the leakage is below the required leakage level of a mineral top cover (0.1 mm/day) and a combined top cover (0.025 mm/day).

Table 3.1 Numerical data on permeability and leakage in Cell 33

Time	P_{H_2O}	$I = P_{H_2O} / H_{cel}$	K_{10}	V_{10}
[d]	[cm H ₂ O]	[-]	[m/s]	[mm/day]
0	1476	97,75	$1,45 \times 10^{-10}$	0,08
39	1476	97,75	$8,44 \times 10^{-11}$	0,05
69	1476	97,75	$6,04 \times 10^{-11}$	0,03
137	1476	97,75	$2,89 \times 10^{-11}$	0,02

With H_{cel} 15.1 cm

Table 3.2 Numerical data on permeability and leakage in Cell 34

Time	P_{H_2O}	$I = P_{H_2O} / H_{cel}$	K_{10}	V_{10}
[d]	[cm H ₂ O]	[-]	[m/s]	[mm/day]
13	1476	97,75	$1,20 \times 10^{-10}$	0,07
51	1476	97,75	$3,56 \times 10^{-11}$	0,02
117	1476	97,75	$2,72 \times 10^{-11}$	0,02
130	1476	97,75	$8,39 \times 10^{-11}$	0,05
167	1476	97,75	$3,22 \times 10^{-11}$	0,02
191	1476	97,75	$2,09 \times 10^{-11}$	0,01
226	1476	97,75	$2,75 \times 10^{-11}$	0,02

With H_{cel} 15.1 cm

4. CONCLUSIONS

The geotechnical research team from Dong-Ah Construction Industrial Co. Ltd. has developed a SF&SR system to be applied as a top isolation for landfills. Permeability measurements are conducted on this SF&SR system, where a matrix of a clayey sandy soil is used in combination with the top and bottom reactants. In the experiments 90 m/m% of soil is mixed with 10 m/m% of reactant material for each layer.

The starting permeability of the SF&SR system was low ($1.5 \cdot 10^{-10}$ and $1.8 \cdot 10^{-10}$ m/s) and transport of the reactants towards the precipitation zone was probably controlled by diffusion rather than by percolation processes due to this low permeability. This has also resulted in a somewhat slower formation of the SF&SR seal than in situations where the reactants were transported by percolation.

The permeability of the SF&SR system was monitored over the whole period and it was found that the permeability decreased in time and that it showed an increase after cracking the seal. The permeability increased to a value of approximately the starting permeability. In the period after cracking the seal, the permeability was monitored again and it was clear that the system has self-repairing properties. The permeability decreased again with approximately the same speed as before cracking the seal, the last datapoint suggests that the permeability is at a minimum. Further measurements are needed to validate this finding.

Under practical conditions the matrix material (soil) will be moist. This will cause the 'fines' from the reactant materials to coat onto larger particles of the matrix material. This phenomenon will 'immobilize' a large portion of the reactant materials which can result in a lower performance of the system permeability development.