Transport processes for harmful species through concrete barriers made with mineral wastes.

by

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ABSTRACT

Composite concrete barriers represent a good alternative to current landfill containment systems because they can be made using waste minerals and thus have a lower environmental and economic cost. They are also more robust when in place. This paper presents results from laboratory testing and site trials and modelling of a concrete barrier system. Laboratory trials to determine the effect of cracking on the composite barrier are also reported. It is concluded that cracking will not cause failure. The modelling of the transport processes provided a good prediction of the movement of some species in the site trials.

Keywords
transport properties, diffusion, permeability, waste management, modelling

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1. INTRODUCTION

1.1

When waste is deposited in landfill it is necessary to contain the leachate using a low permeability liner so that it may be treated before being discharged into the environment. The novel barrier system which is described in this paper has been developed because current liner systems based on high density polyethylene membranes and bentonite enhanced sand have a high economic and environmental cost and are easily damaged after installation.

It is of note that, while current policy requires that deposition in landfill should be minimised, very significant quantities of waste are still being landfilled and this will still form an important part of the waste industry for several decades.

The proposed composite barrier system has two layers of concrete with a layer of clay between them and is shown in figure 1. The principal intended benefits are:

- Low permeability combined with high cation exchange capacity to give improved containment.
- Composite construction to overcome problems with cracking
- Construction from waste materials which would otherwise go into landfills.
- A relatively hard concrete surface to permit operation of vehicles and to prevent damage from large items of waste compacted onto it.

This paper describes a programme in which transport properties of the barrier were measured in the laboratory and used to predict the performance of site trials. Four elements were studied: Na, K, Ca and S. These elements are not generally of concern in the environment but it was not possible to use toxic elements such as Hg because the work involved large quantities of leachate in field-scale trials.

The key transport processes are discussed and the basis of the modelling is described. Results from laboratory and site trials are then presented and compared with the theoretical predictions. The results from the initial period of the site trials have been

reported in a previous paper (1). The results for the final 2 years of the trial and further laboratory testing including cracked samples are included in this paper.

1.2 Research Significance

The transport processes in concrete are key determinants of the durability of concrete structures. In particular the transport of chlorides in reinforced structures is the main cause of corrosion. This paper describes the analysis of transport in a waste containment barrier but the same processes are equally relevant to durability calculations (2).

1.3 Previous research

During the 1980's a very large research programme was carried out in the UK to develop designs for repositories for nuclear waste (3,4). The design which was developed to the greatest extent was the repository for intermediate and low level waste.   This repository was required to have a predictable performance in a deep saturated geological environment over a timescale of up to a million years. The design essentially involved placing the waste in concrete containers and placing these containers in an excavated underground cavern. This cavern was then to be backfilled with a relatively soft cementitious grout (5).

One of the achievements of the nuclear programme was to analyse and define the performance which was actually required of the concrete when used for this application. This performance requirement is quite different from the requirements for concrete in normal construction and lead to the development of some very unusual concrete mixes.

The barrier design uses conventional engineering materials but its method of operation is far from conventional for an engineering structure because it is essentially sacrificial (6). The main function of the barrier is to condition the chemistry of the repository to high pH by dissolving alkalis in the groundwater. The alkalis are free sodium, potassium and lime and subsequently the calcium silicate hydrate which forms the structure of the hardened cement. At the high pH values the harmful species from the waste which are permeating through the barrier are adsorbed onto the cement matrix and immobilized. Clay based liners were considered for the nuclear

repository but concrete was chosen as the best option for the UK. The nuclear programme was stalled in the 1990's by the refusal of planning permission for the test facility at Sellafield in Cumbria.

2 TRANSPORT PROCESSES IN A COMPOSITE CONCRETE BARRIER

2.1 Advection

In this process the pressure of the leachate head causes water flow which carries dissolved ions through the barrier. The rate of transport through the barrier will be determined by the coefficient of permeability k which has the units of m/s and is defined from equation 1 (7):

$$V = \frac{k(h_1 - h_2)}{x} \quad m/s \qquad [1]$$

where V is the Darcy velocity of the fluid flowing through a thickness x (m) with pressure heads h_1 and h_2 (m) on each side.

2.2 Diffusion

In this process the dissolved ions move through the water at a rate determined by the concentration gradient. The flow per second per unit cross sectional area of a porous solid (the Flux, F) is given by equation 2 (7).

$$F = \varepsilon D \frac{dC_l}{dx} \quad kg/m^2/s \qquad [2]$$

where ε is the porosity, D is the intrinsic diffusion coefficient and C_l is the ionic concentration in the pore fluid.

2.3 Adsorption

By far the most effective containment mechanism will be chemical containment. Many harmful species are immobilised by the chemistry of the barrier. In the modelling this has been represented by a linear adsorption isotherm which is measured as a capacity factor for each element in the leachate in each layer of the barrier. To describe these processes two different ionic concentrations must be defined:

C_l kg/m^3 is the concentration of ions per unit volume of liquid in the pores. These ions will pass through the barrier under the influence of the physical transport processes. The concentration per unit volume of the solid will be εC_l where ε is the porosity.

C_s kg/m^3 is the total concentration (including adsorbed ions) per unit volume of the solid. The ions which are adsorbed onto the solid will not move. The capacity factor is defined in equation 3 (8).

$$\alpha = \frac{C_s}{C_l} \qquad\qquad [3]$$

Note that we may calculate $k = \dfrac{Concentration\ in\ solid}{Concentration\ in\ liquid} = \dfrac{Cs - \varepsilon Cl}{Cl} = \alpha - \varepsilon$ [4]

2.4 The Computer Model

A computer model has been written to simulate the transport processes (1). This model was used both to obtain transport properties from the laboratory results and to predict the results from the site trials. In each application the calculations are identical. The only differences are in the exact output given and the length of time that a run simulates. The model is based on physical transport processes (diffusion and advection) with linear adsorption and assumes that the barrier layers are homogeneous and saturated at the start of the trial.

The model works by repeated application of equations 1, 2 and 4 through time and space. The adsorption processes are assumed to reach equilibrium within each time step.

2.5 Cracking and other preferential flow paths.

Cracking could be caused by drying or thermal effects or the imposed stresses on the barrier and would permit transport regardless of the properties of the concrete. The solution to this is to use composite systems of concrete and clay and their performance has been demonstrated experimentally in the work described in this paper.

A possible cause of premature chemical failure is the formation of impermeable "boulder-like" pieces with preferential flow paths for water around them. These

boulders could develop impermeable surface layers through the formation of carbonates, chloroaluminates or magnesium compounds in a similar manner to that observed at the surface of existing concrete structures in hostile environments. If this occurred the alkaline buffering and sorption capacity of the interior of the boulders would be lost. In this way the total buffering and sorption capacity of the repository would be substantially reduced.

In the plans for nuclear waste it is envisaged that almost all of the cementitious material will be in the form of a soft grout (5). This material has been chosen to comply with various operational criteria including being readily pumpable into small spaces between the packages and having a low strength. These requirements have the effect that the formation of hard impermeable boulders will be strongly inhibited. For non-nuclear waste the strength of the concrete was also kept as low as possible.

2.6 Action of sulphates
Penetrating sulphates react with hardened concrete and produce reaction products which have a volume which is greater than the available pore space (6,7). In normal structures this cause expansion of the matrix which continues until the critical stress is developed and cracks form. It is of note, however, that in a deep nuclear waste emplacement the effect is harmless because there is no void space in the whole repository and all compressible wastes are supercompacted to save space. The entire system is also subject to long-term compression from the surrounding rock and this will prevent any expansion and the resulting compressive stresses may even be beneficial. For non-nuclear applications with typical waste emplacement at depths possibly greater than 20m the expansive stresses would be sufficient to cause cracking but the cracks would not open and would remain sealed with reaction product. In the event of any dissolution of this product the multi-layer barrier design described below would make the cracks fill with clay which would be extruded from the inter-layer.

3 THE NOVEL BARRIER DESIGN

The design concept of the novel composite landfill liners is to emplace a number of different layers, each of which compliments and enhances the behaviour of the others

(1,11). Each of the layers has different properties, so that any defects such as cracks, are likely to form at different locations in different layers, thus limiting the creation of connected pathways through the barrier. In the design considered in this work, three layers are envisaged as illustrated in figure 1. The clay-based hydraulic barrier is sandwiched between two layers of concrete. The clay may be a natural material or, for some applications, the concept of an "artificial pourable clay" made from a setting mix based on waste gypsum or ash has been developed.

In most current landfill designs the sided of the cells slope, typically at 30 degrees to the horizontal. This enables conventional machines to work and compact the mineral layers. In order to provide maximum deposition volume on restricted sites a number of vertically sided cells have been built, but the technology for this has been very expensive. The proposed novel barrier system would be well suited to this application because the two concrete layers could be built as conventional concrete walls and then the pourable clay placed between them as the waste level rose to contain the resulting fluid pressures.

4 LABORATORY TESTING

4.1 Mix Designs
The mix designs are shown in table 1. The designs were chosen to make use of available industrial wastes and give an adequate strength (5 MPa) and permeability (10^{-9} m/s) (5,6). Tests were carried out on samples taken from the mixes at the time of the pours for the site trials (12,13).

4.2 Diffusion Tests
 The diffusion cells were used to examine mass transport in reactive systems. An aggressive solution, simulating an acetogenic leachate typical of the early stages of landfill evolution (see table 2) was allowed to react with the sample, whilst concentration changes due both to diffusion and reactive transport were monitored in the cells.

This diffusion test is intended to measure both the diffusion coefficient and capacity factor of species partitioned between a solution and a porous sample. The basis of the

test is a divided cell with the sample in the centre. Artificial leachate is placed on one side and deionised water on the other. Chemical analysis is used to track changes with time on each side (figure 2) (13).

The capacity factors and diffusion coefficients were calculated from the diffusion tests on the mixes used for the site cell using an optimisation routine in the computer model.

The modelled input – output and experimentally measured (real) input- output concentrations of the diffusion cell are plotted for two typical examples in figures 3 and 4. The results show that, for the limited data used, the model optimisation gave a very good agreement between the modelled values and the experimental values. This was achieved by the progressive changing of the capacity factor and diffusion coefficient by the optimisation routine. In figure 3 increasing either parameter increases the transport into the sample and will thus increase the rate of decline of the input concentration. The output concentration will, however, change more if the diffusion is increased but the adsorption is decreased. In figure 4 a high initial concentration in the sample (measured by pore fluid expression and input into the model) gives a rising concentration on both sides of the sample.

The derived results for diffusion and adsorption are in Table 3.

4.3 Permeability Tests

The permeabilities of the specimens were determined using a continuous high-pressure through flow experiment. A solution was eluted through the materials at pressures up to 10 MPa depending on the compressive strength of the particular specimen (12). The apparatus measured both the flow and pressure drop across the samples. Measurements were made after one sample volume of liquid had passed through the concrete or mortar specimens. Assuming an average permeability of 10^{-9} and a maximum leachate head of 1 m above the liner, this corresponds to 16 years of exposure in service. The results are in Table 4.

4.4 Pore Fluid Concentrations

Samples of pore fluid were expressed under pressure from specimens of the different mixes using a pore press similar to an OPI-CAD cell (17). The concentrations were obtained from them using ICP analysis.

4.5 Cracked Samples

In addition to the basic transport measurements the most important design consideration was the effect of cracks. These were investigated using the cell which was used for permeability measurements.

Two sets of different tests were carried out to check the self-sealing property of the multi layer barriers. In the first set of test a 100 mm diameter mortar disc 33.4 mm thick was cracked by applying small loads using a compression test machine. Fibre Reinforcement was used in the sample to prevent it from falling apart. The cracks induced in the disc were clearly visible by naked eye and were measured between 0.5 mm and 1.2mm wide (see figure 5). A 55 mm thick metal spacer ring packed with clay from the trial site was placed on the top of the cracked disc. A two-layer test was then carried out on the high pressure cell apparatus using the synthetic leachate with Fluorescein dye. After running the high pressure cell for about an hour at 95 bar pressure, liquid started to leach out showing the fluorescein colour. When left for another 5 hours at 95 bar the leaching stopped. About 0.8 sample volumes of liquid were collected during this period. No more leaching was observed when the cell was continuously run at the same pressure for another 24 hours (see figure 6).

In the second set of tests hair line cracks (about 0.2 mm wide) were induced in mortar samples (similar to cell number 2 bottom layer mix) and clay (passing 5 mm mesh sieve) was packed inside the sample (see figure 7). A mortar disc (similar to the cell number 2 top layer mix) was then placed on top of the clay and cracked mortar (Fig. 8). The thicknesses of the three layers were proportional to those in the site trial cells. Running this multi layered sample at 100 bars for 3 days showed no leaching from it at all. After dismantling it was noticed that the top mortar had disintegrated under the high-pressure leachate flow but no trace of leachate could be found in the bottom cracked mortar. These experiments therefore indicated that, under the expected high saturation pressures, the clay would effectively seal cracks in the concrete.

5 SITE TRIALS

5.1 Objectives

Three cells were constructed on a licensed landfill operation site at Risley, Cheshire UK with different cementitious composite mineral waste materials (11). This landfill site receives both domestic and industrial waste. The results from cells 2 and 3 are reported here (cell 1 was dismantled early due to site requirements and is not reported).

The purpose of the cells was as follows.

- To provide validation data for the modelling of the performance of the barriers in service.
- To demonstrate a construction method.
- To demonstrate that the novel mixes can be made in industrial quantities (150 tonnes of concrete were used in the three test cells).
- To provided samples of real site batched concrete for laboratory testing.

5.2 Layout and construction methods of the cells:

A typical test cell is shown schematically in Figure 9. The barriers were made up of two layers of concrete with a layer of clay between them. These inverted pyramid shape cells measured 8 metres wide and contained waste to a maximum depth of 1.1 metres. The slopes of the cells were 30° and the cells contained 5.4 m^3 of waste. Table 5 gives the dimensions and volume of each layer in the test cells. The excavation was carried out with an excavator which was also used to place the concrete and mortar mixes designed for the different cells. The concrete layers were placed and levelled by the excavator. The compaction of concrete layers was carried out by two poker vibrators and the compaction of clay layer was carried out using the outside surface of the excavator's bucket.

5.3 Observations from the construction

During the construction of cell numbers 2 and 3 the mix proportions actually used were different to those designed in laboratory due to inaccurate weighing of different materials and partial hydration of CKD while stored at the plant. The mixes actually made were tested and showed higher permeabilities than the mixes initially designed in the laboratory.

5.4 Emplacement of waste and leachate

Shredded waste was used due to size and shape constraints of the cells. It was placed and compacted up to the top level of the test cells. A leachate which was the most aggressive solution found in the landfill was obtained from the treatment plant and the cells were filled 100 mm below the top giving a 1 m head at the deepest point. The cells were covered with a tarpaulin cover to prevent rainwater ingress and contain odour.

5.5 Instrumentation and sampling

Two types of sampling lines were used between the layers of the cell liners using 3 mm plastic tubes in both. In one type the ends of the 3 mm plastic tubes were glued inside porous stone discs of 60 mm diameter. In the other type the layer was drilled and the 3 mm plastic tubes ends were sealed in place in the set concrete with sponge around the end of the line. The sampling lines were placed as an array in the various liner materials and levels. Liquid samples were obtained by applying a vacuum to the lines.

5.6 Modelling transport in the tests cells

Cell 3 needed to be refilled after 12 months. The reason for this was inadequate compaction of the clay layer leading to an increased permeability. The effective indicated premeability was calculated as follows:

Total Volume of the leachate leaked $\cong 4.54$ m^3

Surface area of pyramid in bottom of clay layer $= 25.3$ m^2

Thus indicated permeability $k = 5.2 \times 10^{-9}$ m/s

This corresponds to nearly the same permeability as a Bentonite Enhanced Sand liner and indicates satisfactory performance even when very poor construction practice was

evident; but it did affect the modelling considerably. The permeability calculated from these site observations was therefore used for the clay layer in the model.

6 RESULTS AND DISCUSSION.

6.1 Comparison between model and observations

The initial concentrations of different elements in site leachate and in the pore pressed solutions from the barrier components are in table 6. These were used in the model together with the capacity factors and diffusion coefficients obtained from the diffusion tests. The comparisons of the modelling results and the site observations are shown in figures 10 to 15 for Ca, Na and K for cells 2 and 3 respectively.   On these graphs error bars are shown between the 10th and 90th percentiles from probability calculations at ages of two and four years (in figures 13,14 and 15 these have been offset slightly for clarity). The observed concentrations which are shown on the graph are based on the average from up to four different samples taken in different parts of the cells. For some of these a considerable spread of results was recorded.

The results for cell 2 in figures 10, 11 and 12 lie within the error bars except for the high observed concentrations of sodium (1500 ppm) and potassium (almost 8000 ppm) at the bottom of the clay layer. Table 6 shows a very high initial concentration of sodium in the lower layer concrete and this could have been drawn into the sampling line from below by the applied vacuum. The observed potassium, however, rose significantly above the lower layer concentration after 3 years. The results for calcium and sodium make it unlikely that the pore fluid from the upper layer reached this level and it is clear that the leachate had not arrived because it only had a concentration of 5000 ppm. A possible explanation would be a reduction in pH of the upper surface of the lower concrete layer causing a reduction in the capacity factor and a resulting rise in to solution concentration.

The main differences between the model and the experimental observations in cell 3 were again at the bottom of the clay layer. The observed rise in calcium was not predicted but a predicted rise in potassium did not occur. The predicted potassium peak was caused by the high permeability value used in the model as a result of the observed drop in fluid level in the cell. This caused the model to be dominated by

advection causing the high potassium concentration from the top layer to permeate rapidly through the clay. The probable explanation of the discrepancy is that the observed higher permeability was caused by localised poor compaction of the clay and did not occur at the sampling points. This would also explain why the calcium concentration (which originated in the clay) did not drop as quickly as predicted.

It has been observed (16) that transport processes in landfills are associated with a high degree of uncertainty. The processes modelled in this paper used real landfill leachate which was undergoing biological reactions throughout the experiment and took place in a site environment with all of the associated uncertainty. The combination of these factors with the uncertainty associated with the stated assumptions in the model has given rise to some unexpected events. Nevertheless this is a useful exercise to indicate likely trends in a real environment. Long term results which will involve transport processes far closer to the steady state are actually likely to be more accurate.

7 CONCLUSIONS

The most significant properties of a concrete barrier material are the permeability, the diffusion coefficient and the capacity factor for adsorption. These may be measured in the laboratory and used in numerical modelling of the barrier.

The diffusion coefficient and capacity factor may both be obtained from diffusion tests by modelling the results.

Experimental work has indicated that minor cracking will be sealed by the clay. No barrier can withstand the effect of gross failure of the substrate. This would cause major cracks to form and should be avoided.

Predicting leachate transport in a real environment is very difficult but many of the trends were predicted and the long-term steady state flow should be easier to calculate.

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	Proportions Used	
	kg/m^3	% By mass
Composition of top layer mortar for cell No.2:		
Ferrosilicate slag sand (< 5 mm)	1575	65.9
Cement Kiln Dust – 60%	490	20.5
Lagoon Ash – 40%	325	13.6
Water	200	
Composition of top layer concrete for cell No.3:		
Ferrosilicate slag (< 150 mm to dust)	0	
Limestone (<20 mm)	715	29.8
Ferrosilicate slag sand (< 5 mm)	1105	46
Cement Kiln Dust – 60%	340	14.2
Lagoon Ash – 40%	240	10
Water	220	
Composition of lower layer concrete for cell No.2:		
Chrome Alumina Slag (< 40 mm)	1175	49.6
Chrome Alumina Slag (< 5 mm)	720	30.4
Green sand	100	4.2
Cement Kiln Dust – 60%	165	7
T1Sodium sulphate Solution (lt)	165	
Composition of lower layer concrete for cell No.3:		
Chrome Alumina Slag (< 40 mm)	1175	50.3
Chrome Alumina Slag (< 5 mm)	720	30.8
Green sand	110	4.7
Portland Cement – 5.2% CEM1 42.5N to BSEN 197-1	25	1.1
Cement Kiln Dust – 69.8%	185	7.9
Lagoon Ash – 25%	120	5.2
Water	240	

Table.1: Composition of mixes.

2.043 g	Concentrated Sulphuric acid
4.48 g	Acetic acid
1.897 g	Potassium chloride
7.755 g	Calcium acetate
1.186 g	Ammonium chloride
0.91 g	Sodium chloride
2.588 g	Sodium hydroxide

Table 2 *Composition of synthetic leachate, per litre of solution (pH=5.1)*

Chemical Element	Top layer mix Cell 2 & 3 (Porosity = 12%)		Bottom layer mix Cell 2 (Porosity = 9%)		Bottom layer mix Cell 3 (Porosity = 9%)	
	α	D	α	D	α	D
Ca	7.74	3.7×10^{-10}	0.5	1.77×10^{-10}	0.09	1.35×10^{-10}
Na	0.43	1.19×10^{-10}	0.09	10^{-14}	1	4.58×10^{-13}
K	0.86	1.07×10^{-10}	1.02	1.71×10^{-12}	1	6.73×10^{-12}
S	1	2.07×10^{-12}	1.37	10^{-14}	27.67	2.93×10^{-14}

Table 3: Capacity factor (**α)** and D (diffusion coefficient) values for the four major elements in the trial cells.

	7 days strength (MPa)	28 days strength (MPa)	Intrinsic permeability to water @ 28 days (m/s)	Intrinsic permeability to leachate @ 28 days (m/s)
Cell 2 top	1.1	1.7	4.5×10^{-9}	5×10^{-9}
Cell 2 base	4.4	6.9	2.3×10^{-9}	4.5×10^{-9}
Cell 3 top	0.9	1.3	1.2×10^{-8}	7.5×10^{-9}
Cell 3 base	2.8	6.0	1.2×10^{-8}	6.2×10^{-9}

Table 4: *Characteristics of the mixes used in the site trial cells.*

	Thickness (m)	Depth to lowest point (m)	Width (m)	Volume (m^3)
Waste	-	1.1	3.84	5.4
Upper Concrete	0.2	1.33	4.65	4.16
Clay	0.5	1.9	6.66	18.61
Lower Concrete	0.3	2.25	7.87	18.28

Table 5: *Dimensions and volume of each layer of test cells.*

		Cell 2			Cell3		
Elements	Site leachate	Top layer	Clay layer	Bottom layer	Top layer	Clay layer	Bottom layer
Ca	344	4	350	1214	4	350	14
Na	2300	450	450	10802	450	450	2157
K	4730	15193	300	1761	15193	300	761
S	770	2000	500	549	9294	500	50

Table 6 *Initial liquid concentrations used in models for site trials (mg/l).*

Composition		Main Physical Function
Alkali activated slag or pozzolan concrete containing spent foundry sand and metallurgical slag aggregate	Hard-wearing layer	Mechanical support of vehicles during operational phase and initial containment of leachate
Non-swelling clay or "artificial clay".	Softer sealing layer	Physical containment of leachate and crack sealing.
Concrete containing aggregate of larger particles of metallurgical slag and waste and spent foundry sand.	Base layer Sub-soil	Chemical conditioning of leachate to promote adsorption and physical containment with low permeability and diffusion coefficient. Base for sealing layer.
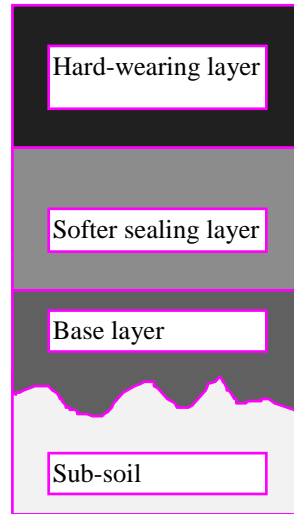
Figure 1: *The new composite landfill liner.*

Fig.2 *Diffusion cells.*
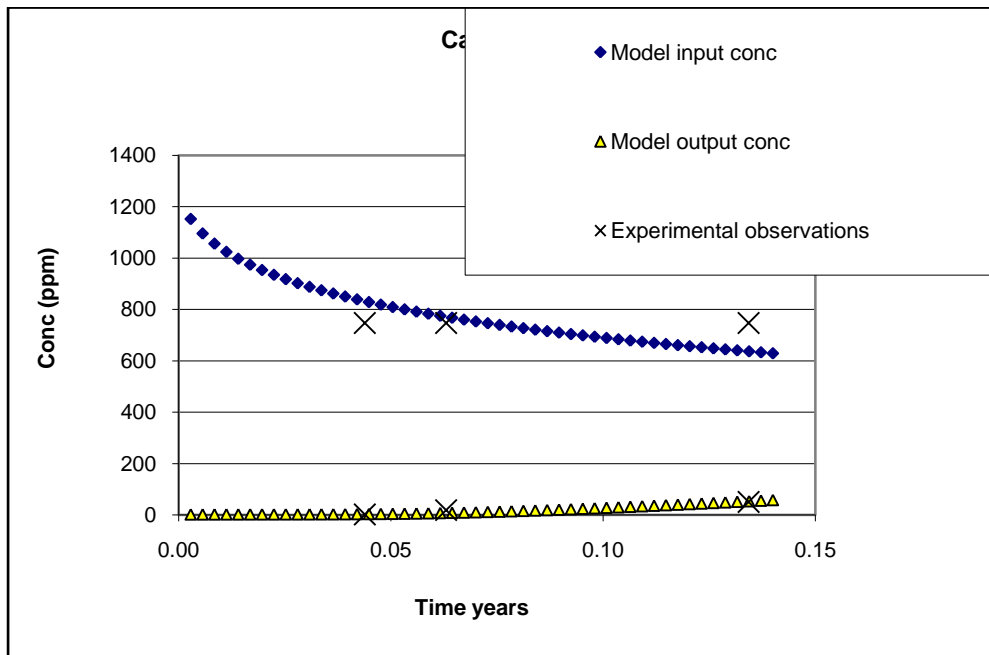
Figure 3 Concentrations from laboratory diffusion testing for calcium
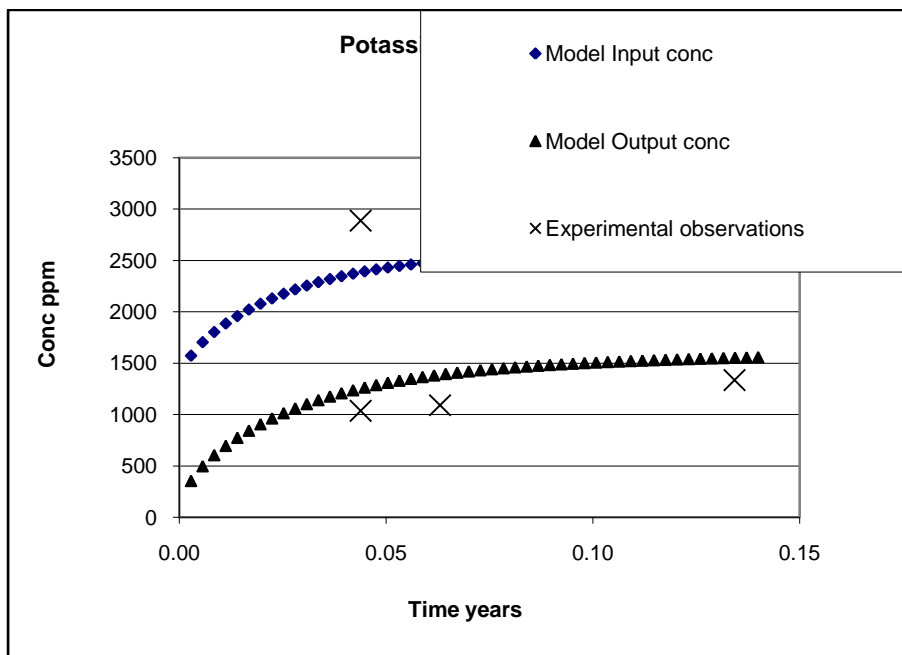
Figure 4 Concentrations from laboratory diffusion testing for potassium

Fig. 5: Narrow cracks induced in the F.R. mortar disc.

Fig. 6: Cracked sample after clay has sealed the cracks.

Fig. 7: The set up of bottom layer mix and compacted clay with silicone rubber round the rim and inside wall to prevent ingress of leachate from rim and interfaces.
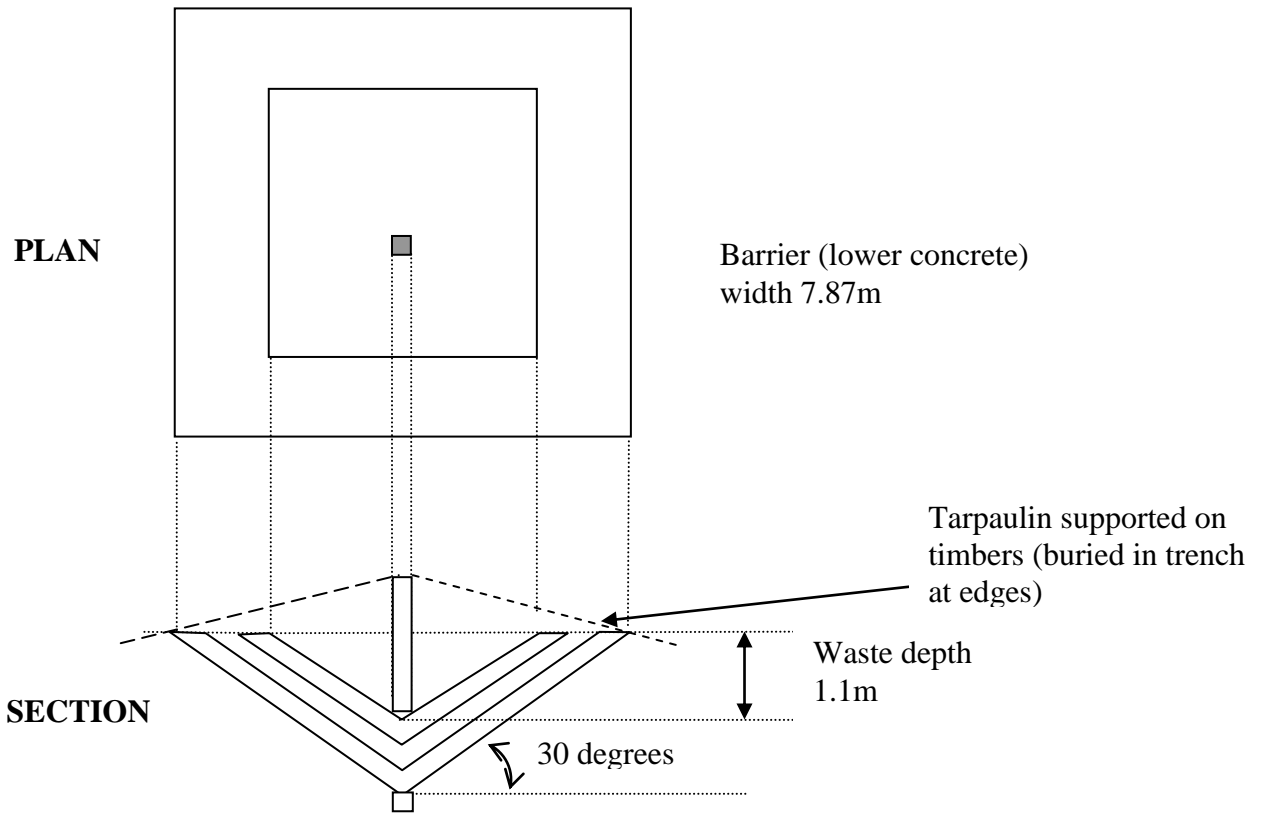
Fig. 8: Multi layer sample with partially compacted clay inside it.

PLAN

Barrier (lower concrete)
width 7.87m

Tarpaulin supported on
timbers (buried in trench
at edges)

Waste depth
1.1m

SECTION

30 degrees

Figure 9: Typical trail test cell layout.
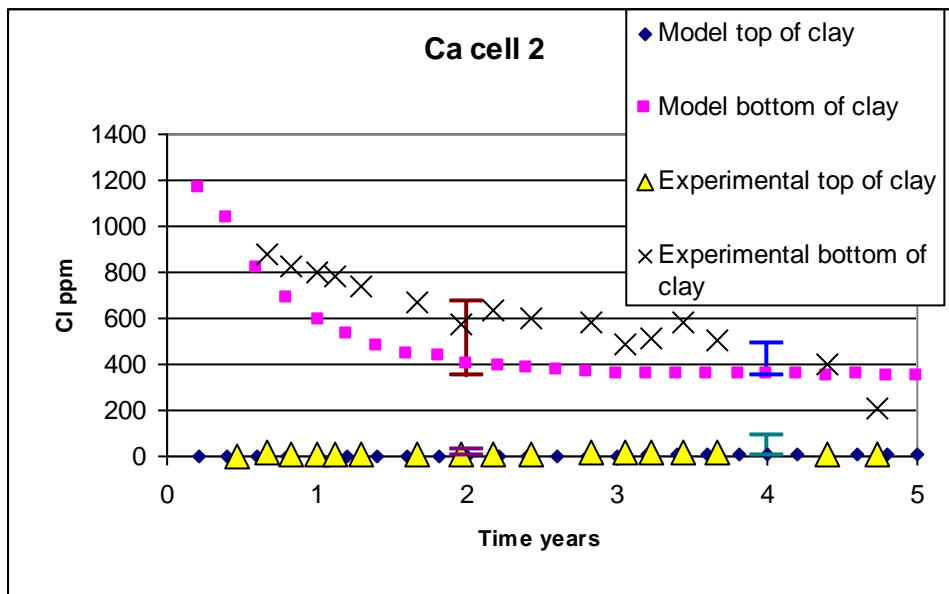
Figure 10. Concentrations of calcium in site trial cell 2
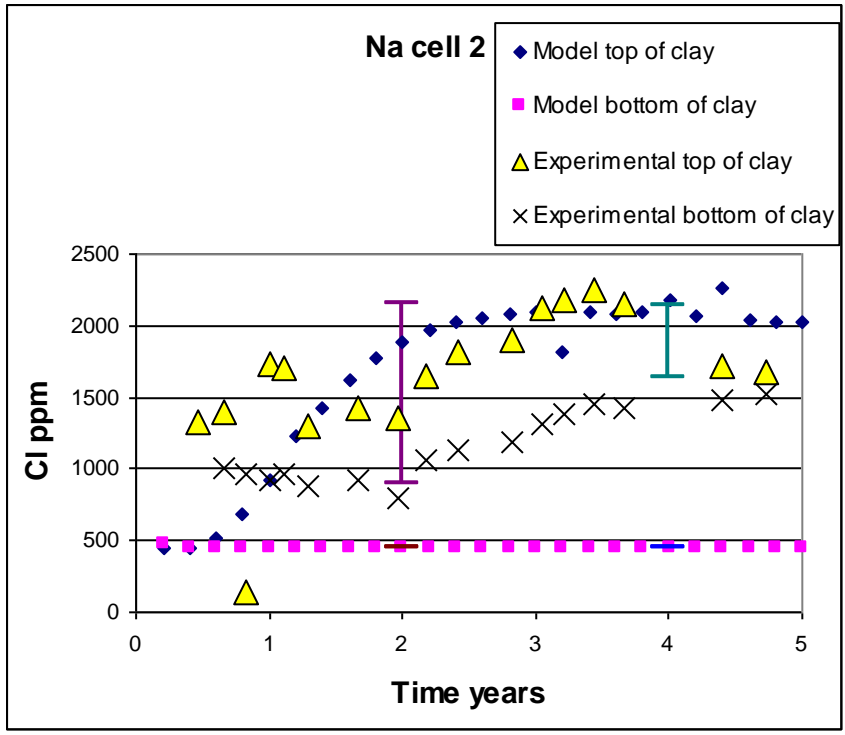
Figure 11. Concentrations of sodium in site trial cell 2
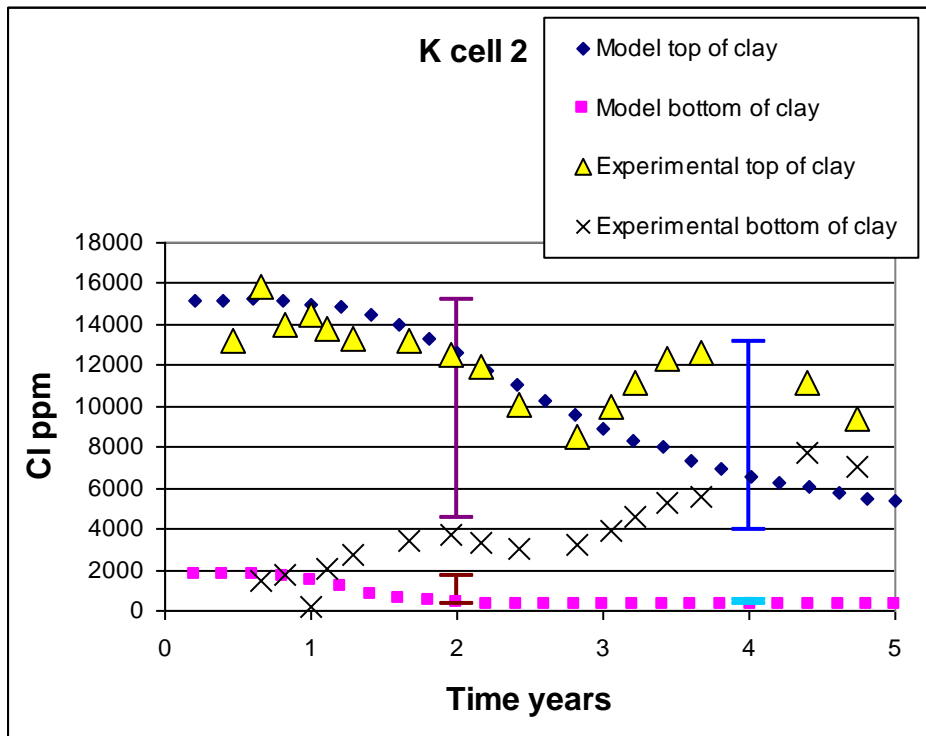
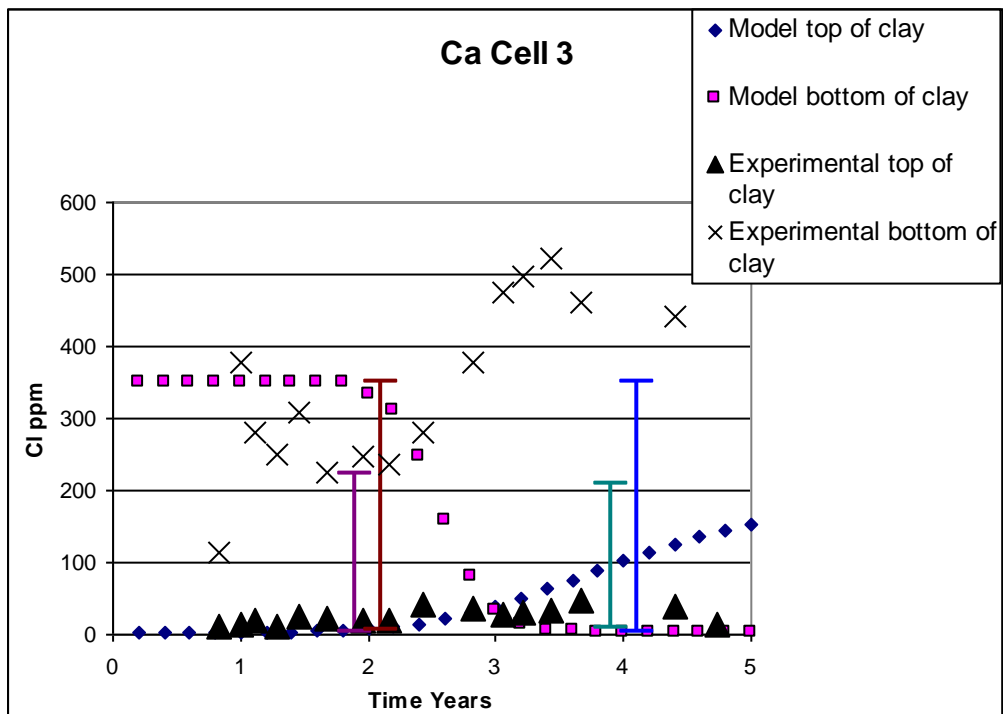Figure 12. Concentrations of sodium in site trial cell 2

Figure 13. Concentrations of calcium in site trial cell 3
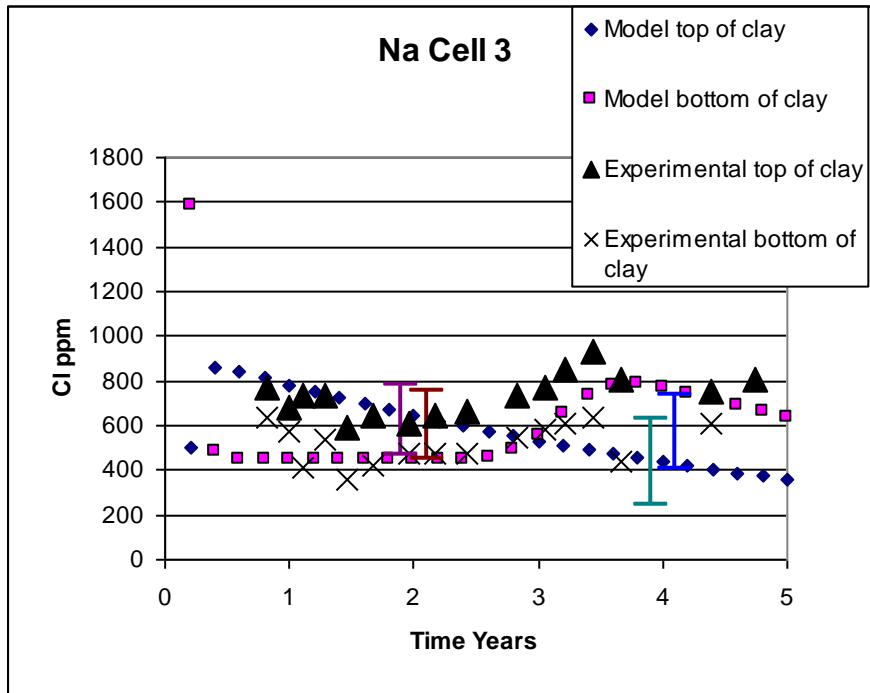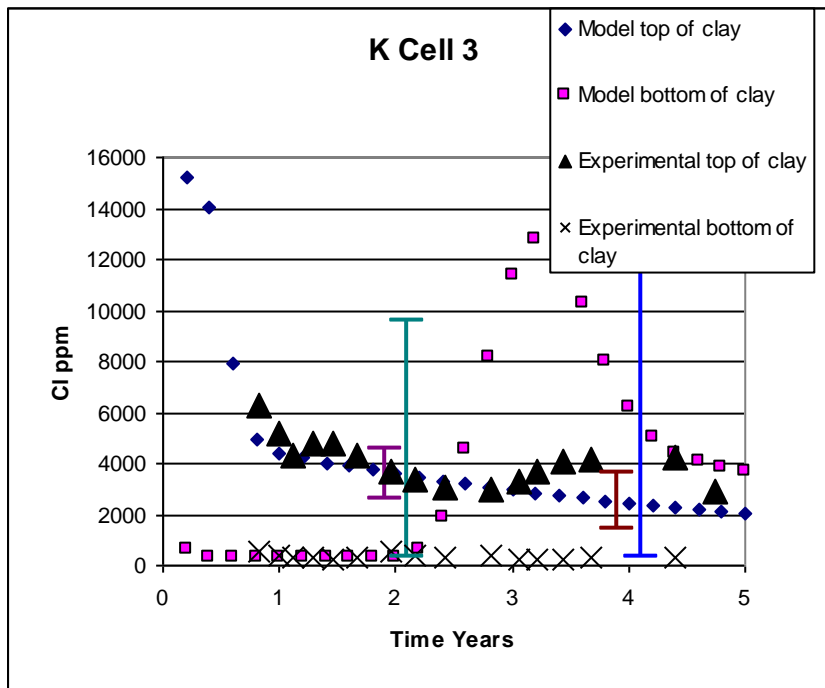

Figure 14. Concentrations of sodium in site trial cell 3

Figure 15. Concentrations of potassium in site trial cell 3