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## **PERFORMANCE CRITERIA FOR CONCRETE AS A BARRIER FOR LEACHATE IN WASTE CONTAINMENT**

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### **ABSTRACT**

When cementitious materials are used for the containment of waste they act in two different, and sometimes conflicting, ways. The first is physical containment in which the waste is physically isolated from the environment. The second is chemical containment in which the water passing through the barrier is buffered to high pH thereby substantially reducing the solubility of many harmful species and promoting sorption onto the matrix. Chemical barriers have been extensively researched for nuclear waste containment and this paper reports on a major research programme in which the principles developed in the nuclear work have been applied to non-nuclear work.

The concrete in the current programme is used as a liner below a landfill for domestic waste. In this environment the performance criteria normally applied to structural concrete are often not relevant and other criteria apply. After one or two years of operation the landfill is normally capped and the strength of the concrete is not important. Similarly sulphate attack is harmless and frost resistance is not relevant. The performance criteria which are relevant are the permeability, diffusion coefficient and the chemical buffering capacity.

This paper reports on the evolution of the concept of the concrete barrier from nuclear to non-nuclear waste and the range of tests and site trials that are being carried out to demonstrate performance in a landfill barrier.

**KEYWORDS:** chemical buffering, concrete, permeability, diffusion, waste disposal

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## 1. INTRODUCTION

During the 1980's a very large research programme was carried out in the UK to develop designs for repositories for nuclear waste [1-3]. Three of the present authors (Claisse Atkinson and Tyrer) worked on this programme. 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.

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 [4]. 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.

The nuclear programme was stalled in the 1990's by the refusal of planning permission for the test facility at Sellafield in Cumbria. By this time, however, new legislation had been introduced which required containment for non-nuclear wastes (both domestic and industrial) in landfill. Technology to achieve this containment is now used on a massive scale throughout the world and it is normally based on clay systems supplemented with high-density polyethylene (HDPE) membranes.

Clay based liners were considered for the nuclear repository but concrete was chosen as the best option for the UK. Work is now in progress on a programme to use the results from the nuclear work to develop designs for cementitious barriers to give an economically and technically superior performance for non-nuclear waste. The programme started in 1998 and is being carried out in the UK by Imperial College and Coventry University.

In this paper the two different containment mechanisms are discussed, physical and chemical. Results from a numerical model are then presented which show the effectiveness of chemical containment. The laboratory and site work in the programme is described and initial results from the laboratory work are discussed.

## 2. PHYSICAL CONTAINMENT

### 2.1 Transport processes

There are numerous physical transport processes which could take place in the barrier, these include: Thermal migration, Electromigration, Osmosis, Electro-osmosis and Capillary suction. Initial analysis has, however, shown that these will not be significant and for the present work only advection and diffusion are considered:

## 2.2 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 [1]:

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

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 (the Flux,  $F$ ) is given by [2].

$$F = \varepsilon D \frac{dC_1}{dx} \text{ kg/m}^2/\text{s} \quad [2]$$

where  $\varepsilon$  is the porosity,  $D$  is the intrinsic diffusion coefficient and  $C_1$  is the ionic concentration in the pore fluid.

## 3. CHEMICAL CONTAINMENT

The physical transport processes are restricted by adsorption in which a linear isotherm is assumed, i.e. a fixed proportion of the ions in any part of the barrier are adsorbed onto the matrix and will not move. To describe these processes two different ionic concentrations must be defined:

$C_l \text{ kg/m}^3$  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  $\varepsilon C_l$  where  $\varepsilon$  is the porosity.

$C_s \text{ kg/m}^3$  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 as [3]

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

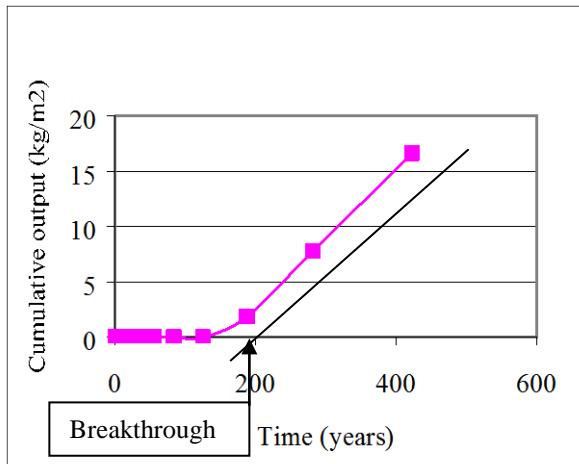
For many ions in a typical concrete  $\alpha$  is 1000 or higher. If the porosity is approximately 10% only one ion in  $10^4$  will be available for transport.

## 4. LIMITS ON THE PERFORMANCE OF THE BARRIER

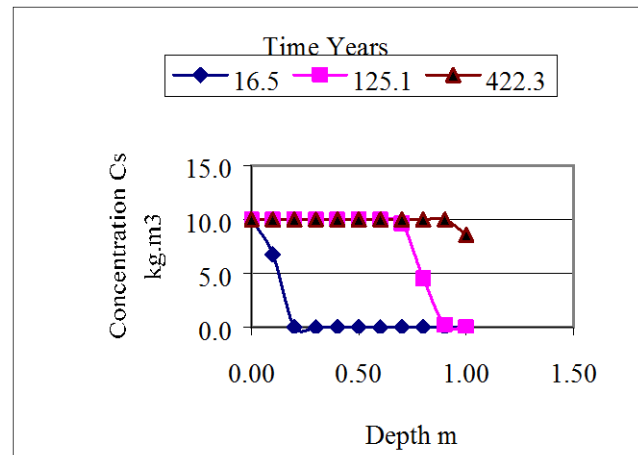
### 4.1 The transport processes

Figs. [1-5] show the influence of the physical and chemical properties of the concrete on the transport through the barrier. They were produced with a numerical model which applies equations [1-3]. Figs. [1] and [2] are for the base case which is a 1m head of leachate on a 1m

thick concrete barrier with a permeability of  $10^{-9}$  m/s, a diffusion coefficient of  $5 \times 10^{-10}$  m<sup>2</sup>/s and a capacity factor of 10 (typical values for concrete). Fig. 1 shows the output of ions from the base of the barrier against time. The breakthrough time (158 years) is obtained by extrapolating the linear part of the curve back to the axis as shown. Fig. 2 shows the concentration across the barrier at different times. It may be seen that in the steady (final) state the concentration is at its maximum level throughout the barrier.

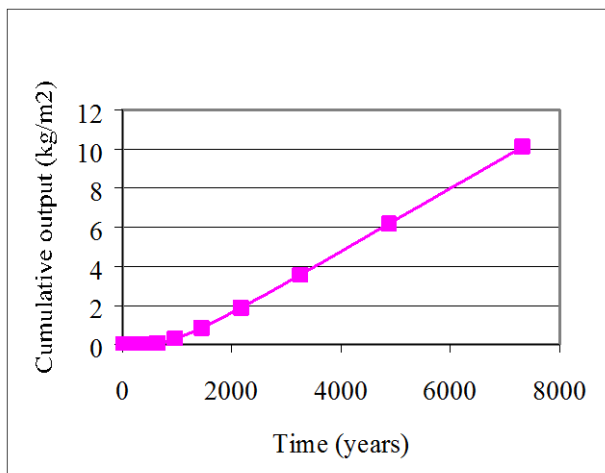


**Fig. [1] Base case output**

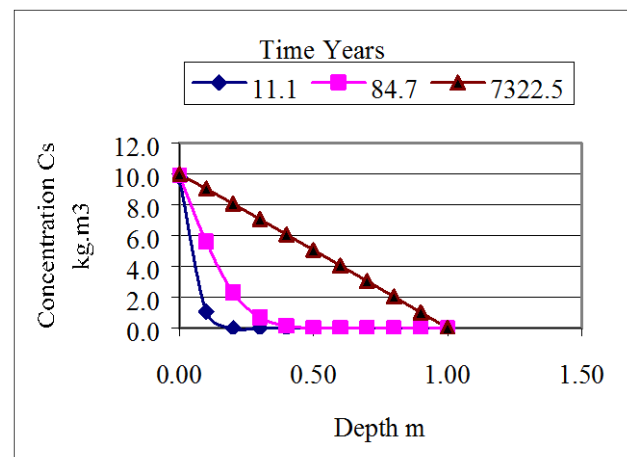


**Fig. [2] Final concentrations for base case**

Fig. [3] shows the situation with the permeability reduced to  $10^{-12}$  m/s (typical value for a very good concrete). The breakthrough time has only increased to 1049 years for the three order of magnitude decrease in the permeability. The reason for this can be seen in Fig. 4 where the steady state concentration can be seen to show a linear decrease across the sample indicating diffusion control.

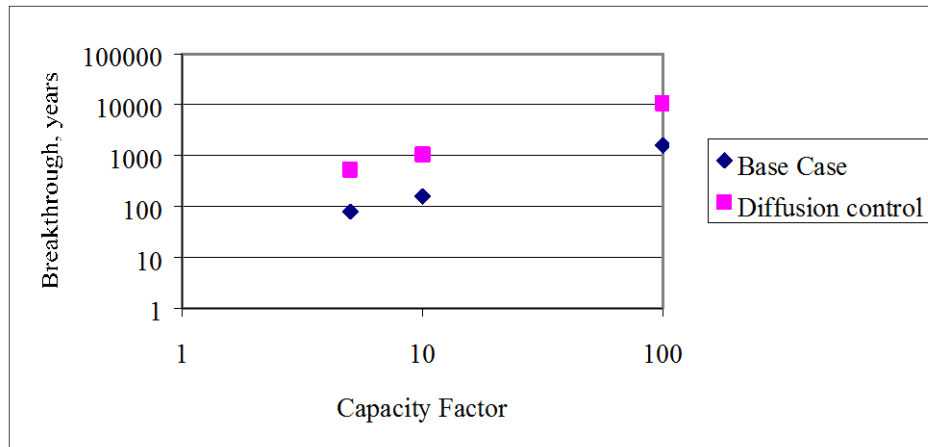


**Fig. [3] Diffusion control output**



**Fig. [4] Final concentrations for Diffusion control**

Fig. [5] shows the effect of changing the capacity factor for each of the examples shown above. It may be seen that this increases the breakthrough time far more effectively than reducing the permeability because it increases linearly with the capacity factor.



**Fig. 5 Influence of capacity factor**

#### 4.2 Cracking

This is the obvious mode of physical failure which has been the reason why concrete barriers have not been common in the past. Cracking could be caused by drying or thermal effects or the imposed stresses on the barrier. The problem is overcome by using composite barriers which are discussed below

#### 4.3 High pH "boulder" formation

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. 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 will also be kept as low as possible.

#### 4.3 Action of sulphates.

Sulphates react with hardened concrete and cause expansion of the matrix which leads to significant loss of strength. This effect may be prevented by the use of sulphate resisting cement or Blastfurnace Slag cement. It is of note, however, that in a deep nuclear repository the effect is harmless because the expansion is contained by the surrounding rock. For non-nuclear applications the containment pressures should also be sufficient with typical waste emplacement depths greater than 20m.

### 5. THE MULTI-LAYER BARRIER CONCEPT

In order to operate for a long time a chemical barrier depends on other barriers to limit the flow of groundwater through it. This is normally achieved for nuclear waste by positioning the

repository in a geology with a very low permeability. This option is not available for non-nuclear waste. It is also clear that a single cementitious barrier will not provide sufficient short-term containment to satisfy current regulations for a liner. Any design with this type of barrier must therefore use a composite system with several cementitious layers or layers of other materials.

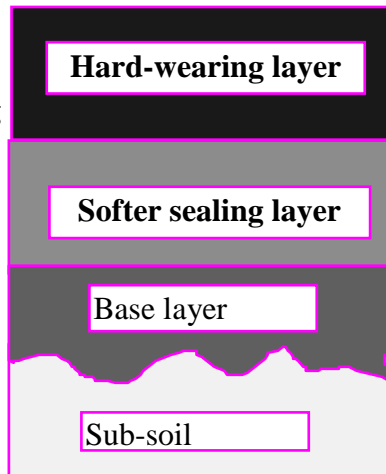
**Composition**

**Main Physical Function**

Concrete or mortar 0.2m thick with good mechanical properties during the operating phase

Non-swelling clay 0.5m thick

Concrete or mortar 0.3m thick with high alkaline buffering and low leaching



Mechanical support of vehicles during operational phase

Physical containment of leachate and crack sealing.

Base for sealing layer

**Fig. [6]. Schematic arrangement of Barrier**

Fig. [6] illustrates the type of multi-layer barrier which was used for the work described in this paper. Each layer has a specific function and enhances the performance of the others. The clay provides a physical seal and is protected from mechanical damage and extrusion by the concrete above and below. The upper concrete provides a hard working surface for vehicles to use during waste emplacement and eliminates the need to use selected waste near the liner (as required when an HDPE liner is used). The lower layer provides long-term physical and chemical containment. Once the waste has been placed the barrier will be subject to compression and the clay is expected to extrude into any cracks that form in the concrete. The possibility of light mesh reinforcement in the lower concrete layer has been considered for conditions where foundation conditions may lead to uneven settlement.

Using a layer of bentonite in a concrete barrier might be possible but calcium bentonite reacts with the free lime in concrete and destroys both so the type of bentonite to be used would have to be carefully selected and tested.

**6. MATERIALS**

The conclusion from the design work on the concrete layers is that the material must satisfy the following performance requirements:

- A cube strength of 5 N/mm<sup>2</sup> is adequate. This will be sufficient to support vehicles during emplacement and to prevent large items in the waste penetrating the barrier during compaction.
- The strength requirement is only for emplacement. After 2 years little strength is required.
- Expansion of the barrier is harmless. In operation it will be subject to triaxial compression. Thus sulphate attack or unsound cements should be harmless.

- The permeability must remain low.
- Cracking is inevitable. What must be avoided are large cracks with large “boulders” between them. Small cracks will close due to the compression and will seal from mineral deposition (autogenous healing) or clay intrusion from the middle layer.
- Alkaline buffering is essential but this must only be available to the leachate permeating through the barrier. There can never be sufficient buffering for the entire waste load.

Careful consideration of these requirements revealed that the concrete could be made almost entirely from waste materials such as industrial slags and ashes. Most of these materials are unsuitable for normal structural concrete and therefore have no market value. Many of them also attract high disposal costs (and taxes) so the concrete may have “negative value”.

## 7. LABORATORY TESTING

Through flow testing was carried out on concrete samples at high pressure. Cylindrical samples approximately 30mm long with diameters of 55mm and 100mm were subjected to liquid pressures up to 14MPa on one of their flat surfaces. The test was carried out in a modified Hoek cell [5], as used in rock testing. In this cell the sample is contained in a membrane through which a confining oil pressure is applied on the curved surface. The confining pressure is kept above the applied liquid pressure. Measurements were made after one sample volume of liquid had passed through the concrete. Assuming an average permeability of  $10^{-9}$  and a maximum leachate head of 1m above the liner (as required in UK landfills) this corresponds to 16 years of exposure in service. Tests were carried out with water and also with a simulated leachate.

The high-pressure test is intended to measure:

- The permeability of the samples to water.
- The change in permeability in the presence of leachate.
- The adsorption of ions from the leachate by measuring eluent chemistry.
- The relationship between numbers of sample volumes passing and changes in permeability, strength and eluent chemistry (pH).
- The effect of different residence times in the sample by running the test at different pressures and/or sample thicknesses. This will determine whether the leachate achieves chemical equilibrium with the barrier. It will also check the sensitivity of the observed permeability to changes in pressure.
- The effect of cracking
- The performance of multi-layer systems by testing them in the cells
- The ability of the clay to seal cracks by testing multi-layer samples with cracked concrete.
- The availability of the buffering capacity of the barrier to react with the leachate (i.e. whether high pH “boulders” are forming) by permeating a dye through the samples

## 8. SITE TRIALS

### 8.1 Construction of the cells

Three site trials have been constructed. These consisted of miniature waste cells in the shape of inverted pyramids which contain waste to a depth of one metre and have sides at 30 degrees of slope, giving a total width of 8m. Each cell contained a total of 22m<sup>3</sup> of concrete



**Fig. [7]. Placing concrete in the site trial**

in the two layers. Fig. [7] shows cell 1 during construction. It is of note that the concrete in this picture contained only spent borax as its cementitious component – a material with no calcium or silica. After construction the cells were filled with shredded waste and leachate and kept covered to prevent rain ingress. The cells provide a basic check for leachate transport. Observations are made from the samples from vacuum lines and from dismantling. The advection is driven by a pressure head of 1m over a 1m thick barrier.

### 8. 2 Operation of the Vacuum Lines

The vacuum lines are fine bore flexible pipes which were cast in at each level of the barrier. On the end of the lines there is a sponge or a stone to form a void. If there is gas in adjacent pores or cracks etc. samples may be extracted easily with a vacuum. If there is no gas or other pathway the flow up the line must be from advection from a spherical region around the void. The velocity of flow up the pipe is given by [4]:

$$V = \frac{4khr_1}{r_0^2} \quad [4]$$

where:

k is the permeability ( $10^{-9}$  m/s)

h is the head of water corresponding to the pressure (m/s)

$r_1$  is the radius of the void (10mm)

$r_0$  is the bore radius of the pipe (1.5mm)

The outer radius of the integral is insignificant provided it is much greater than  $r_1$ . Using the values in brackets gives a flow of 640mm per hour.

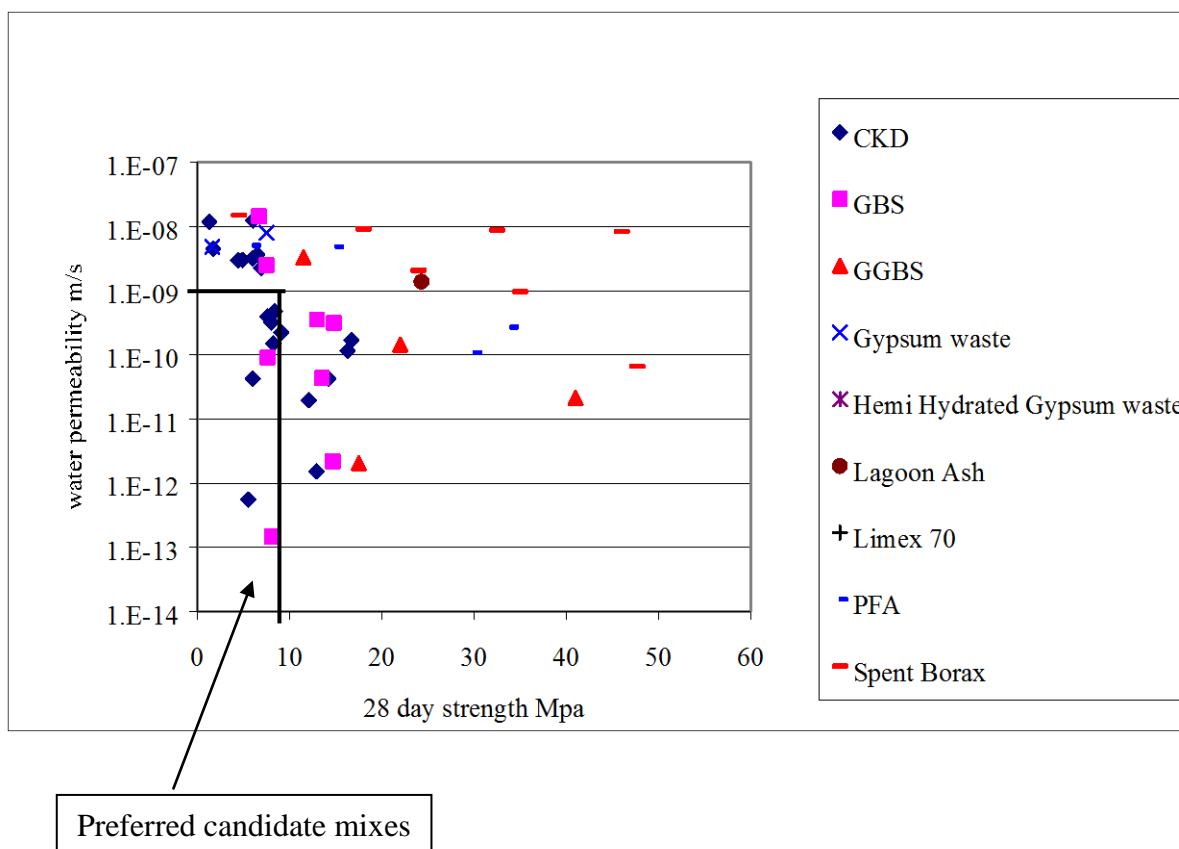
### 8.3 Dismantling objectives:

At the end of the work the cells are dismantled. This gives the opportunity to carry out the following:

1. Determine the extent of leachate penetration through the barriers.
2. Investigate the effect of leachate exposure on the materials in the different layers.
3. Investigate interactions between the layers in the presence of leachate.



## 9. RESULTS AND DISCUSSION



**Fig. [8]. Results from the laboratory testing. The materials listed in the legend are the primary cementitious components.**

Fig. [8] is the graph of results from the laboratory work from which the candidate mixes were selected. The selected mixes were intended to have a permeability below  $10^{-9}$  m/s and a strength below 10 MPa. It may be seen that relatively few mixes lie in the required range for both strength and permeability.

The site trial has already served to show that the mixes could be made and placed in large quantities. Some particular problems were identified, such as crystallization of the sodium sulphate solution (a waste product used in place of the mix water) on a cold day and difficulties with mixing large ferrosilicate aggregate. Analysis of samples from the vacuum lines is in progress.

## 10 CONCLUSIONS

1. The concept of performance must be defined relative to the intended purpose of a concrete mix.
2. For waste containment the key performance indicator for concrete is the capacity factor.
3. For landfill liners it is possible to make concretes which perform well but have negative cost.

## 11 ACKNOWLEDGEMENT

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