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EVALUATION OF KAOLINITE-CONTAINING CLAY AS A POTENTIAL MINERAL LINER FOR LANDFILL LEACHATE CONTAINMENT

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A kaolinite-containing clay is subjected to laboratory tests to assess its potential use as a mineral liner for landfill leachate containment. Taken from the Bodelva china clay pit, the sample was shown to pass all the regulatory requirements needed to be considered suitable for use as a mineral liner. With recent research (Batchelder *et al.,* 1996), showing that smectite clay minerals are susceptible to c-axis contraction when in the presence of potassium and ammonium ions, and that more inert clay minerals such as kaolinite remain unaffected, the use of kaolinite-containing clays instead of a clay matrix dominated by higher swelling clay minerals could become increasingly popular.

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INTRODUCTION

Landfill space within Devon and Cornwall is at a premium with a present capacity of about 6 years left in Cornwall and 2 years in Devon. New environmentally acceptable landfill sites have to be found and developed as soon as possible. One of the major pollutants to emanate from a landfill site is the leachate, a liquid product of the degradation of the waste. To combat this problem landfill developers are required to place a clay/mineral liner to control the seepage of the leachate into the surrounding environment. Therefore when looking for potential new sites it is important to locate a clay source within an economically transportable distance.

Traditionally, swelling clay minerals such as those from the smectite group, have been the preferred clay mineral for use in mineral liners. Research at Camborne School of Mines shows that kaolinite-containing clays may also be used as mineral liners and that they may in fact offer distinct advantages over smectite-containing clays.

Prior to the 1980s, landfill sites were operated on a dilute and disperse principle, whereby leachate was allowed to seep into the ground where it would be attenuated by dilution and degradation. In the mid 1980s, the principle of containment was introduced with the release of Waste Management Paper 26, (DoE, 1986). This guideline stated that the liner to be used should be a 1 m thickness of clay with a hydraulic conductivity of no more than 1x10^{-09}m/s, or an equivalent man-made product such as a geomembrane. In the 1990s the Environment Protection Act (HMSO, 1990) was introduced and brought with it stricter controls on containment and how it should be regulated. This led to Waste Management 26 (DoE 1986) being broken down into 5 separate papers, with Waste Management Paper 26B (DoE 1995) concentrating on Landfill Design, Construction and Operation. Even though almost 10 years had passed between the Waste Management Paper 26 publications, the minimum criteria for hydraulic conductivity remained unchanged at a value of no more than 1x10^{-09}m/s. A more detailed breakdown of the minimum requirements of a clay liner is provided by the North West Waste Regulation Officers (NWWRO, 1996) which provides details of how a potential mineral liner should be assessed and what criteria it should achieve.

LANDFILL LEACHATE AND MINERAL LINERS

The mineral liner is not totally impermeable. It has a small but measurable permeability as indicated by a hydraulic conductivity of $\leq$ 1x10^{-09}m/s, so leachate will percolate through it at a small but measurable rate. It is important to note that all landfill liners leak. By choosing a suitable liner material and engineering it to suitable standards on site, this seepage can be kept to an acceptably low rate.

Landfill leachate is formed from water that has percolated through the emplaced waste. The infiltration of rainfall, ground and surface water into the waste mass, coupled with a biochemical and physical breakdown, produces a leachate which contains soluble components of the waste, suspended solids and microorganisms. Leachate formation can be split into two main groups, acetogenic and methanogenic. Acetogenic leachate is produced at the inception of the leachate production and is characterised by the production of volatile fatty acids, acidic pH, high Biochemical Oxygen Demand (BOD) to Chemical Oxygen Demand (COD) ratio, and high levels of ammoniacal nitrogen and oxidised nitrogen. The methanogenic stage is the methane-producing stage, where the majority of the fatty acids created in the acetogenic stage are converted to methane and carbon dioxide. Methanogenic leachates are characterised by low concentrations of fatty acids, neutral to alkaline pH, lower levels of ammoniacal nitrogen and low BOD to COD ratio. Table 1a and 1b shows typical leachate compositions for acetogenic and methanogenic stages respectively.

Determinand	**Minimum**	**Maximum**	**Mean**
pH value	5.12	7.8	6.73
COD	2,740	152,000	36,817
BOD$_5$	2,000	68,000	18,632
ammoniacal-N	194	3,610	922
chloride	659	4,670	1,805
fatty acids (as C)	963	22,414	8,197
iron	48.3	2,300	653.8
magnesium	25	820	384
potassium	350	3,100	1,143
calcium	270	6,240	2,241

Table 1a - Summary of composition (mg/l) of acetogenic leachates (adapted from DoE Research Report No. CWM 072/94 -1995)

Determinand	Minimum	Maximum	Mean
pH value	6.8	8.2	7.52
COD	622	8,000	2,307
BOD_5	97	1,770	374
ammoniacal-N	283	2,040	889
chloride	570	4,710	2,074
fatty acids (as C)	<5	146	18
iron	1.6	160	27.4
magnesium	40	1,580	250
potassium	100	1,580	854
calcium	23	501	151

Table 1b - Summary of composition (mg/l) of methanogenic leachate (adapted from DoE Research. Report No. CWM 072/94 -1995)

The pollution problems of leachate include the oxygen demands that would be created if it entered water courses, due to its high COD and BOD levels. The nutrients such as ammoniacal nitrogen and chloride are also a problem, as they can initiate the formation of algal blooms which in turn will starve the water course of oxygen. Another pollution problem is the creation of fatty acids within the leachate. Fatty acids entering groundwater aquifers can be a major pollution source which can prove very difficult to remove and which will remain in the aquifer for many years.

CONTROLS ON THE HYDRAULIC CONDUCTIVITY OF A MINERAL LINER

When considering a suitable local clay to use as a mineral liner for a landfill site, the ultimate criterion to be met is a hydraulic conductivity of less than $1x10^{-09}$ m/s with a thickness of 1 m, as specified in Waste Management Paper 26B, (DoE, 1995).

The hydraulic conductivity of a soil is a measure of its permeability and describes its capacity to allow the flow of a specific fluid through it under a unit hydraulic gradient. The hydraulic conductivity value of a potential mineral liner is dependent on a number of factors, which include: 1. particle size distribution, 2. void ratio and soil structure, 3. mineralogical composition of the soil and 4. nature and temperature of permeating fluid.

Particle Size Distribution

Generally speaking, the finer-grained a potential mineral liner is, the lower its hydraulic conductivity will be. However, it is also important to consider that the size distribution of a potential clay liner should not only lend itself to producing a low hydraulic conductivity, but should also provide relative ease of compaction in the field. A mineral liner containing greater than 50% clay particle size (less than 2 μm) may attain a low hydraulic conductivity in the laboratory, but compacting the same sample in the field, and hence attaining the low value of hydraulic conductivity, will prove extremely difficult. Mineral liners with high clay fractions will also prove to be more susceptible to shrinkage due to desiccation. The sand fraction of a mineral liner (0.06 mm - 2.0 mm) can also reduce the hydraulic conductivity as long as the sand size grains remain matrix supported by the clay fraction after compaction. Potential mineral liners which contain between 10-20% clay fraction and 30-40% sand fraction will provide a suitably low hydraulic conductivity mineral liner which can be engineered in the field with relative ease.

Void Ratio And Soil Structure

In the context of mineral liners, arguably the most important controls on permeability are the void ratio and structure of the soil. Both of these are controlled by particle size distribution and compaction. The way in which a soil is compacted has a considerable effect on the size and nature of the voids between the particles and hence the permeability.

The most extensive study illustrating how water content and dry unit weight influence the hydraulic conductivity of compacted clay was published by Mitchell *et al.* (1965) who demonstrated that the energy and method of compaction significantly influence the hydraulic conductivity of compacted clay. Their research also revealed that hydraulic conductivities dry of optimum moisture content (OMC) are higher than those wet of optimum. This difference in hydraulic conductivity can be explained by the change that occurs as a direct result of alterations in the void ratio and soil fabric at both microscales and macroscales.

At the microscale, increases of water content and compactive effort result in re-orientation of the clay particles and hence subsequent reduction in the size of the inter-particle pores. Figure 1 shows a typical compaction curve. Dry of OMC we see a flocculated pattern of clay minerals which naturally leads to an increase in inter-particle pore size and an increase in the hydraulic conductivity, at OMC the soil has a mixture of a flocculated and dispersed structure and above OMC we have a dispersed pattern of the clay minerals leading to a reduction in size of inter-particle pores and hence a lower hydraulic conductivity.

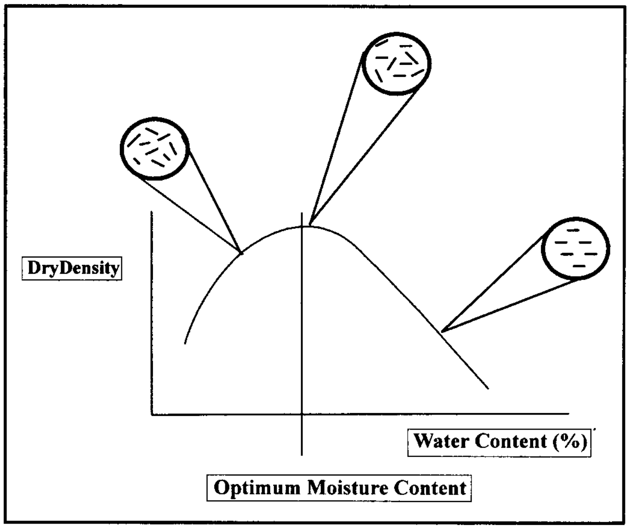

Figure 1 - Typical compaction curve showing orientation of clay particles (adapted from Lambe 1958)

On a macroscale, increasing the water content results in an increased ability to break down the clay clods or aggregates within the soil, hence eliminating inter-aggregate pores. In the field, the majority of the flow of water in a compacted clay occurs in relatively large pore spaces between clods of clay. Soft wet clods are easier to remould than hard dry clods, therefore compacting wet of the OMC results in smaller inter-clod voids and lower hydraulic conductivities.

Mineralogy

An additional important factor to consider is the mineralogical composition of the soil. Having smectite as the dominant clay mineral in the soil instead of kaolinite would improve the soil's containment properties, because it is finer-grained and because of its greater tendency to adsorb water and swell, therefore lowering the permeability. The other benefit that smectite would bring to the soil would be increased attenuation capabilities compared to that of a clay mineral such as kaolinite. However, the reverse of this is that smectite is also more susceptible to being broken down by certain species contained within the landfill leachate than kaolinite, this is explained in more detail below.

Nature and Temperature of the Permeating Fluid

When studying the hydraulic conductivity of a soil, the nature of the fluid that is to permeate through the sample in-situ should always be considered. This is especially true in the case of landfill leachate where two factors come into consideration. Firstly the constituents of the leachate may have an effect on the clay minerals in the soil. A plethora of research has been carried out on the effect that landfill leachate has on clay mineralogy. Research has shown that in its normal dilute state, landfill leachate does not significantly alter the hydraulic conductivity of compacted clays (Sai and Anderson, 1991; Daniel and Liljestrand, 1984). However, the levels of sodium, potassium, and ammonium ions in Municipal Solid Waste (MSW) leachate can be sufficiently high as to effectively exchange some of the calcium and magnesium found in clay minerals as the leachate permeates through. The adsorption of sodium ions can lead to the decrease of the hydraulic conductivity due to double layer expansion (Griffen *et al.*, 1976). The role of potassium and ammonium cations however, is quite different. These two species tend to fix onto swelling clays such as vermiculites and smectites, contracting the c-axis and hence causing a reduction in the mineral or solids volume, (Batchelder *et al.*, 1996). This in turn can lead to the further opening of the voids or fractures between them, thus causing an increase in the hydraulic conductivity of the compacted mineral liner.

The other consideration to take into account is the density and viscosity of the fluid flowing through the soil. The hydraulic conductivity is dependant on these distinct properties of the fluid. The following equation shows the relationship of hydraulic conductivity to the density and viscosity of the fluid flowing through it along with the value of intrinsic permeability. The intrinsic permeability is a true measure of the soil alone and is a constant property of the soil. The term in parentheses reflects the fluid properties. This equation helps us to study how increasing temperature affects the value of hydraulic conductivity.

$$K= \left(\frac{\text{Þ}g}{\mu}\right) k.$$

Where: K = Hydraulic conductivity (m/s), (.1c; Density of permeating fluid (kg/m^3), (μ= Dynamic viscosity of permeating fluid (kg/ms), g = Gravitational acceleration (m/s^2), k = Intrinsic permeability (m^2).

Dilute landfill leachate has density and viscosity properties which are very similar to water, however when leachate is produced in a landfill site the temperature can be upwards of 50°C, at these temperatures the viscosity of the leachate decreases resulting in an increase in the observed hydraulic conductivity. Calculations using Equation 1 show that if a clay has a hydraulic conductivity of $1x10^{-09}$m/s at 15°C, the same clay at 50°C could undergo a twofold increase in its hydraulic conductivity due to the changes in the physical properties of the permeant. These calculations perhaps provide a possible explanation for small differences sometimes found between field and laboratory hydraulic conductivity values.

METHODOLOGY

Standard Requirements and Tests

Recent guidance (NWWRO, 1996) published to assist licensees in the preparation of liner design and to assist regulation officers at the Environment Agency, sets out minimum requirements for the properties of a soil to be considered as a landfill liner. They use the minimum requirements set out by Daniel (1993) which based on American Society of Testing Materials (ASTM) definitions and not the British Standards. The minimum requirements include: 1. Percentage fines (particles less than 0.075mm) > 2030%, 2. Plasticity Index > 7%, 3. Percentage gravel (particles greater than 4.76mm) < 30%, 4. Maximum particle size 25-30mm.

The NWWRO (1996) document suggests a standard suite of tests which should include: 1. Natural moisture content (B51377: Part 2 1990: 3.2), 2. Particle size distribution (BS1377: Part 2: 1990: 9), 3. Atterberg limits (BS1377: Part 2: 1990:4.3, 5.3 and 5.4)), 4. Soil density (B51377: Part2: 1990: 8), 5. Compaction Tests (BS1377: Part 4: 1990: 3.3), 6. Hydraulic conductivity (BS1377: Part 6 : 1990: 6).

Sample Material

If it is to be used as a landfill liner, the kaolinite-containing clay has no need to be processed, it can and should be used in its raw state. The kaolinite-containing clay used in this study was a sample kindly donated by Redland Minerals from the Bodelva china clay pit, now in the ownership of Goonvean and Rostowrack China Clay Co. Ltd. A 25 kg bag of china clay was taken directly from the face of the Bodelva china clay pit. This 25 kg of china clay was separated into two samples via the cone separation method. The two samples were then tested for the properties specified above and were compared with the standard minimum requirements for a potential mineral liner.

Hydraulic Conductivity Tests

Hydraulic conductivity tests were carried out on samples compacted at both 2.5 and 4.5 kg levels. The moisture content of the soil was controlled at between 3 and 5% above optimum moisture content (OMC) during compaction for both tests. Prior to hydraulic conductivity testing the sample was compacted into a cell and allowed to soak under water for 48 hours to achieve fully saturated conditions.

The hydraulic conductivity tests were performed on the falling head apparatus developed at the Cambome School of Mines. The apparatus has a selection of tubes ranging from 3.1 mm up to 20 mm diameter. The higher the permeability the wider the diameter of tube used, therefore for a clay of $1x10^{-09}$ m/s, the smallest diameter tube is used leading to faster response and more accurate readings. A pressure transducer is set up to relay the head of the water in the tube being used to a data logger. The data logger was set to take a reading every half hour. When the head, h, had fallen over 0.5 m from the initial head, h_0, (which can take between 1-2 days) the test is stopped and the data is downloaded onto a computer and placed on a spreadsheet, from which the hydraulic conductivity of the sample is calculated. When plotting the results, the test is accepted if the graph of log. (h/h_0) is linear and has a correlation coefficient of 97% or more.

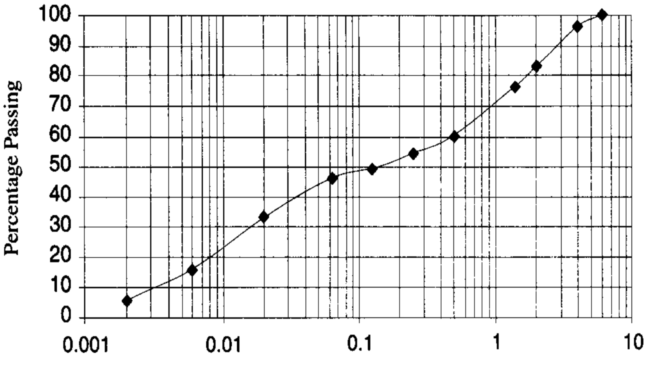

Figure 2 - Particle size distribution curve for china clay

RESULTS

Sample Description

The raw china clay was a pink colour (suggesting presence of iron oxides) and consisted of quartz grains within a clay matrix. The mineralogy of the china clay was analysed via x-ray diffraction and showed that the samples contained the following minerals: kaolinite, quartz, feldspar, mica, and rutile. The only clay mineral found in this sample was kaolinite, no additional clay minerals which might be expected, such as smectite or illite, were found.

Particle Size Distribution

The particle size distribution of the samples was determined as per BS1377: Part 2: 9: 1990. Figure 2 shows the particle size distribution curve for the sample. The results show that 45% of the sample was less than 75 microns, which is well within the regulatory requirement, added to this 4% of the sample was greater than 4.76 mm, again well within the regulatory limit.

Property	Value
Natural Moisture Content	14%
PSD <0.075mm	45%
>4.76mm	8%
Liquid Limit	24.75%
Plastic Limit	16.73%
Plasticity Index	8.02%
Soil Density	2.63 kg/m^3
Maximum Dry Density	
2.5kg	1.936
4.5kg	2.073
Optimum Moisture Content	
2.5kg	12.01%
4.5kg	8.10%
Hydraulic Conductivity	
2.5kg	$1.31x10^{-09}$m/s
4.5kg	$7.6x10^{-10}$m/s

Table 2 - Evaluation results for the china clay sample.

Atterberg Limits

The Atterberg limits showed that the sample had a liquid limit of 24.75% and a plastic limit of 16.73%, giving the sample a plasticity index of 8%, which satisfies the regulatory requirement and indicates that there are enough clay minerals in the sample to provide a cohesive and low hydraulic conductivity liner.

Compaction Characteristics

It is important to know the maximum dry density and the optimum moisture content of the sample and to obtain a compaction curve as it assists the laying and compaction of the liner in-situ. Samples were compacted at both 2.5 and 4.5 kg levels and the results are shown in Table 2.

Hydraulic Conductivity

The hydraulic conductivity of the sample was very close to the regulatory requirement. At the 2.5 kg level, the hydraulic conductivity was $1.3x10^{-09}$m/s, however at the 4.5 kg level the hydraulic conductivity satisfies the criteria as it is $7.6x10^{-10}$m/s. Therefore this clay would have to be compacted to an equivalent of the 4.5 kg test in the field. With the modern technology available today however, this is not a problem.

CONCLUSIONS

Kaolinite-containing clay which would usually be considered poor quality for its orthodox use has the potential to be used as a mineral liner for containing landfill leachate. The kaolinitecontaining clay used was an unprocessed iron oxidised china clay from Bodelva Pit, Cornwall. The tests carried out were as recommended by the Environment Agency and all criteria and minimum requirements were satisfied. The plasticity index of the material is only just above the regulatory limit. However, the most important parameter has been satisfied with the sample attaining a hydraulic conductivity value of $7.6x10^{-10}$m/s, albeit at the higher level of the 4.5 kg compaction test.

A china clay matrix sample taken from one china clay pit was used to provide the results in this study. It should be remembered that china clay occurs as extremely variable deposits and that these results do not necessarily prove that all deposits will be suitable as mineral liners. The results have shown however, that kaolinite-containing clays, whether in the form of china clay or ball clay, have the potential to form a suitable mineral liner to assist in the containment of landfill leachate

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