Soil-biopolymer barriers and soil-biofilm barriers

Barrières sol-biopolymères et barrières sol-biofilm

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ABSTRACT: There are many examples of biofilms and biopolymers increasing the viscosity of fluids or causing reductions in the hydraulic conductivity of porous media. In many of these cases, the phenomenon is cited as detrimental to the operation of the system in which it occurred. "Biofouling" is often reported in heat exchangers, cooling towers, filtration media, pipes and ship hulls. In these cases the biofilms may reduce conductivity, reduce flow or increase drag. There are also many widely used beneficial uses of biofilms and biopolymers. Biopolymers are used in the food and cosmetic industries as emulsifiers or thickening agents. Biopolymers have also been used in the oil well drilling industry to clog porous media to improve resource recovery.

Recent studies have sought to investigate the potential for biofilms and biopolymers to improve the hydraulic conductivity of material for use as liners in waste containment facilities. Regulatory bodies, in many countries, require part of the lining system, for such facilities, to comprise of a compacted clay layer with a hydraulic conductivity of $1x10^{-9}$ m/s or less. Suitable material can be expensive, or indeed impossible, to obtain in some areas. It is proposed that by adding relatively small quantities of biosubstances to marginal material, a low permeability can be achieved. This paper reviews some of the published research to date, as well as introducing some results of short term testing on silty sand, improved by the addition of biopolymers. Other reported properties of biofilms and biopolymers indicate further promising potential. Some of the potential geotechnical and geoenvironmental applications of these biosubstances are broached as well as some of the drawbacks to be overcome.

RÉSUMÉ: Il y a de nombreux exemples où la présence de biofilms ou de biopolymères ont causé l'augmentation de la viscosité des fluides ou la réduction de la conductivité hydraulique des milieux poreux. Dans plusieurs de ces cas, le phénomène est considéré comme nuisible à l'opération du système dans lequel elle s'est produite. "Biofouling " est souvent rapporté dans les échangeurs de chaleur, tours de refroidissement, milieux de filtration, tuyaux et coques de bateau. Les biopolymères sont utilisés dans les industries alimentaires et cosmétiques comme émulsifiants ou agents d' épaississement. Ils ont été également utilisés dans l'industrie de forage de puits de pétrole pour obstruer les milieux poreux afin d'améliorer la récupération des ressources pétrolières.

Des études récentes ont cherché à étudier la capacité des biofilms et des biopolymères à améliorer la conductivité hydraulique des sols d'étancheisation des sites d'enfouissage de déchets. Les agences de protection de l'environnement, dans beaucoup de pays, exigent souvent pour ce type de site que la couche compacte d'argile ait une conductivité hydraulique d'au moins $1x10^{-9}$ m/s. Parfois le matériau nécessaire pour la construction de ce type d'étancheisation est cher ou impossible à obtenir. Dans ce cas, l'ajout de quantités relativement petites de biosubstances peut permettre l'obtention d'un matériau de faible perméabilité. Cet article passe en revue une partie de la recherche faite jusqu'a présent et présente quelques résultats à court terme examinant un sable argileux traités à l'aide de biopolymères. D'autres propriétés rapportées des biofilms et des biopolymères indiquent un potentiel d'utilisation prometteur. Les applications potentielles de ces biosubstances dans les domaines de la géotechnique et de la géotechnique de l'environnement sont citées comme certains des inconvénients à surmonter.

1 INTRODUCTION

The performance of engineered barriers for waste containment depends upon many factors, including the type of wastes being contained, the material used, quality of construction, and long term durability under adverse environmental conditions. The most commonly used material, clay, has very good hydraulic properties, with a hydraulic conductivity of usually below 10^{-9}m/s when compacted adequately. It also is non-reactive, able to withstand the most adverse environment. Clay also has a moderate strength that is usually adequate in these containment facilities to withhold any possible settlement and deformation. In other words, it has been proven in practice that clay is probably the most viable soil to be used for waste containment.

The absence of clay in some areas poses a difficulty in constructing waste containment facilities that are economical and efficient. Alternative solutions are sought. The main problem is in finding a substitute with the same low hydraulic conductivity that can be used relatively cheaply. If a material with a higher hydraulic conductivity is used, eg, silt of k around 10^{-7} m/s, then the layer thickness needs to increase 100 *times* to achieve the same hydraulic effectiveness.

This paper considers the feasibility of creating new barrier materials by using soils modified by commercially available biopolymers. It also reviews some of the published research to date, on biofilms and bioplymers. This investigation stems from the fact that the plugging effect of bacteria in porous materials was found to reduce the permeability of oil reservoir rock (Hart et al. 1960). This was originally noted as a reduction in recovery after injection of water in to oil recovery wells. The same mechanism was later utilised to plug high porosity materials to facilitate the recovery of oil from lower permeability layers (Lappin-Scott et al. 1988).

2 BIOPOLYMER VS BIOFILM

"Biopolymer" is a term used to describe the viscous suspension that can be obtained from a variety of biological sources. Biopolymers primarily consist of polysaccharides and water. Polysaccharides comprise of repeating sugars, which create a large compound with many functional groups that interact to produce a viscous suspension. Some of the most widely used biopolymers are Guar Gum (derived from the ground endosperm of a legume), Sodium alginate (derived from the cell walls of Brown Algae) and Xanthan Gum (a biopolymer produced by *Xanthamonas campestris*). Biopolymers derived from different sources have different rheological properties. Some biopolymers, such as Sodium Alginate, set when mixed with divalent cations, such as calcium or magnesium. The cations create a bond between the functional groups of the polysaccharides, which increases the shear strength of the suspension.

"Biofilm" describes a matrix of bacterial cells, living in sessile mode, and the extra cellular polysaccharides (EPS) that they produce. Not all bacteria produce EPS and, of those that do, the properties of the EPS vary with the strain of bacteria involved. The EPS, often referred to as "slime", provides physical and chemical protection for the live bacterial cells contained within. It has been shown that bacterial cells living in a biofilm can withstand more severe chemical and physical conditions than the same strain of bacteria in a suspension. The EPS has very low hydraulic conductivity and limits transport within it to diffusion. Due to this constraint, biofilms often contain conduits thought to provide nutrients to the cells deep within the film and to transport wastes out of the matrix. Biofilms can bond strongly to almost any surface. The bacterial cells attach to a surface by proteaceous appendages, called fimbriae, then the polysaccharides engulf the cells and bond irreversibly to the surface. In the event of death of the bacterial cells, the EPS remains largely unchanged (Shaw et al. 1985). This characteristic makes the removal of biofouling caused by biofilms particularly difficult, as the application of anti-bacterial solutions does not remove the EPS, which is the source of the problem.

3 MECHANISMS OF HYDRAULIC CONDUCTIVITY REDUCTION

Several mechanisms of reduction of hydraulic conductivity in porous media due to biological action have been reported by various authors. Suspended bacterial cells can cause particulate clogging of pores (Hart et al, 1960, Lappin-Scott & Costerton, 1990). Biofilms can develop in the porous media reducing the volume of pores and altering the pore geometry and roughness (Li et al. 1993, Dennis & Turner 1998). Biopolymers, which are in effect the EPS without the living cells, can be applied to reduce the void ratio of a material (Martin et al. 1996). These mechanisms are described in more details in the following.

3.1 *Particulate clogging*

Several researchers have investigated the phenomena of pore clogging by bacterial cells. The studies indicated that full size suspended cells caused skin plugging at the surface of porous media. This resulted in a low permeability layer at the inflow end, but had very little effect on the rest of the sample (Hart et al. 1960). In an attempt to improve the cell distribution throughout the sample, Lappin-Scott & Casterton (1990) introduced ultramicrobacteria into a sample, followed by nutrient dosing to return the cells to full size. The ultramicrobacteria, as the name suggests, are smaller than full size cells and are produced by starvation of bacteria. The smaller cells were able to penetrate deeper into the sample prior to resuscitation, thus producing a reduction in hydraulic conductivity along the entire length of the sample. Some of the research noted non-Darcian behaviour by clogged samples. This was attributed to the displacement and/or shearing of bacteria aggregates. The particulate clogging of pores has been shown capable of reducing hydraulic conductivity by 95 %. Most studies were conducted on sand or sandstone cores. The observed improvement of hydraulic conductivity may have been quite different for finer grained base materials.

3.2 *Biofilms Biopolymers*

Biofilms develop when bacteria attach to a surface and the cells excrete EPS. Over time the EPS matrix expands, encompassing more cells which in turn increases the rate of expansion. The EPS is a viscous material that is highly hydrophilic. Studies by Li et al. (1993) and Dennis & Turner (1998) demonstrated a significant reduction in hydraulic conductivity over time, given a suitable nutrient source. This reflects the reduction in voids as the biofilm develops. Dennis and Turner were able to achieve a reduction in hydraulic conductivity of up to three orders of magnitude in a silty sand material. The effectiveness of biofilms relies on suitable conditions for bacterial and EPS growth. Turbulent flow, low nutrient levels or extreme chemical or physical conditions may hinder the formation of a biofilm. However, one of the benefits of biofilms is the ability to lay dormant until more favourable conditions occur, to withstand extreme conditions once a biofilm has formed and to spread through a medium without external intervention. Not only can the EPS protect the cells from undesirable environmental fluctuations, it can also modify the environment. For instance, anaerobic bacteria are able to live in biofilms under aerobic conditions. Certain biofilms can also produce mineral deposits (for instance, plaque is a biofilm, tartar is the associated mineral deposit). These mineral deposits further reduce the void diameter and are very difficult to dislodge.

Biopolymers reduce hydraulic conductivity by filling voids with a viscous material, similar to the mechanism employed by biofilms. However, unlike biofilms, biopolymers do not grow. The biopolymer material, which usually comes in powder form, needs to be distributed through the medium and hydrated. Due to the interaction of the polysaccharides the suspension becomes viscous. The time required for peak viscosity varies for different biopolymers. Chan (1998) achieved a reduction in hydraulic conductivity of three orders of magnitude in a clayey sand material. Some biopolymers can be physically stabilised by the addition of divalent cations. This stabilisation would further increase the viscosity of the biopolymer, which would be expected to lead to a further reduction in hydraulic conductivity. Results of studies involving this procedure have not been located.

4 SHORT TERM INDICATIONS

While the long-term behaviour of low permeability barriers is obviously of interest, observing short-term behaviour is a good indication of the potential of a technology for further investigation. Table 1 summarises the reduction in hydraulic conductivity obtained by various researchers including the present investigation. Materials used and mechanisms, considered to be responsible for the reduction, are also included.

5 METHODOLOGIES

Dennis & Turner (1998) investigated the reduction of hydraulic conductivity in silty sand with the addition of *Beijerinckia indica*, which subsequently forms a biofilm. The durability of the reduced permeability material to several permeants was also investigated. The samples were prepared by mixing the soil with a bacterial and nutrient solution and compacting at optimum moisture content.

Martin et al. (1996) investigated the reduction in hydraulic conductivity and the increase in strength of clayey silt with the addition of commercially available biopolymers. The samples were prepared for permeability testing by mixing 1% and 2% solutions of Xanthan Gum and 1% solution of sodium alginate with the soil to achieve 20% moisture content (wet of optimum).

Samples were cured for at least 5 days prior to testing. Samples were also prepared for triaxial testing at a moisture content of 13.7% (OMC of untreated material) with biopolymer concen-

Table 1: Summary of selected results indicating reduction in hydraulic conductivity of porous media

Matrix	Hydraulic Conductivity (m/s)		Material added	Mechanism	Reference
	Initial	Reduced			
Berea sandstone	1.3×10^{-6}	5×10^{-8}	Bacillus subtilis (dead)	Particulate clogging	Hart (1960)
Sandstone cores	3×10^{-6}	9×10^{-7}	Klebsiella pneumoniae UMB	Particulate clogging	Lappin-Scott et al. (1988)
Sandstone cores	3×10^{-6}	4×10^{-8}	Klebsiella pneumoniae UMB (resuscitated)	Particulate clogging and Biofilm	Lappin-Scott et al. (1988)
Clayey silt	5×10^{-8}	8×10^{-10}	Xanthan Gum (1%) at 30% moisture content	Biopolymer	Martin (1996)
Clayey silt	10^{-6}	10^{-8}	Xanthan Gum (1%) at 13.7% moisture content	Biopolymer	Martin (1996)
Clayey silt	10^{-8}	5×10^{-10}	Xanthan Gum (1%) at 20% moisture content	Biopolymer	Martin (1996)
Clayey sand	10^{-6}	3×10^{-10}	Guar Gum (2%)	Biopolymer	Present investigation
Clayey sand	10^{-6}	2×10^{-10}	Sodium alginate (1%)	Biopolymer	Present investigation
Clayey sand	10^{-6}	1×10^{-10}	IDPAC (1%)	Biopolymer	Present investigation
Clayey sand	10^{-6}	3×10^{-11}	Xanthan Gum (1%)	Biopolymer	Present investigation
Silty sand	10^{-7} to 10^{-8}	10^{-10}	*Beijerinckia indica*	Biofilm	Dennis & Turner (1998)

trations of 0.3%, 0.5% and 1% for Xanthan Gum and 0.5% and 1% for sodium alginate.

The present work investigated the reduction of hydraulic conductivity and the change in shear strength of clayey sand with the addition of commercially available biopolymers. The materials were prepared by mixing 0.5% and 1% solutions of Guar Gum, Sodium alginate, Xanthan Gum and IDPAC with the soil at a moisture content 2% wet of optimum.

5.1 *Reduction in hydraulic conductivity*

Figure 1 shows the reduction of hydraulic conductivity obtained by Martin et al. (1996) and the present investigation. The results indicate a valuable reduction in hydraulic conductivity of the materials being tested. The results also indicate the relative effectiveness of the types of biopolymers used. In both studies Xanthan Gum was more effective than Sodium Alginate. The present investigation showed Guar Gum, to be the least efficient biopolymer trialed, with 2% being required to reach the target hydraulic conductivity of 10^{-9} m/s.

5.2 *Time dependency*

As mentioned previously, the effectiveness of biofilms and biopolymers can be time dependent. It was found that the hydraulic conductivity of a 0.5% Sodium Alginate mix reduced from 2×10^{-9} m/s at 7 days curing time, to 2×10^{-10} m/s at 70 days. The results presented by Martin showed a gradual reduction in hydraulic conductivity, from 10^{-9} to 10^{-10} m/s, over a 6 months period.

5.3 *Increase in material strength*

Due to the viscosity of the biopolymer and biofilm and the adhesion to soil surfaces, it is reasonable to expect that there may be an increase in shear strength of soil due to the addition of biopolymers or biofilms.

Martin et al. (1996) conducted a series of undrained triaxial tests to assess the change in material strength with the addition of biopolymers or the growth of biofilm. The results were reported as maximum deviatoric stress for a strain of 20%. If full saturation is assumed then the stress can be considered to be the cohesion of the sample. A summary of the results is presented in Table 2.

Chan also investigated the change in shear strength of clayey sand mixed with biopolymers. The results indicated a slight increase in cohesion with curing time. There was no apparent change in friction angle.

Table 2: Change in material strength (Martin et al. 1996)

Material	Deviatoric stress (kPa)
Untreated clayey silt	120
0.3% Xanthan Gum	160
0.5% Xanthan Gum	170
1% Xanthan Gum	180
0.5% Sodium Alginate	140
1% Sodium Alginate	150

6 DISCUSSION

The body of research into the use of biofilms and biopolymer to modify soil properties, has demonstrated that a significant decrease in hydraulic conductivity of porous materials is achievable. A reduction of at least two orders of magnitude has been reported by independent researchers using as little as 0.5% Xanthan Gum mixed with a fine grained soil matrix. Up to three orders of magnitude reduction in hydraulic conductivity has also been achieved by the addition of bacteria, and subsequent growth of biofilm, in silty sand.

The reduction of hydraulic conductivity has been shown to be dependant on the initial porosity of the material, the mechanism of clogging, the concentration and variety of biosubstance applied, the moulded moisture content and the curing time. Where the major mechanism of reduction is particulate clogging of pores and biofilm production, a lower initial hydraulic conductivity resulted in a greater reduction. For both biofilm and biopolymer induced reductions, there appeared to be a minimum achievable hydraulic conductivity for similar conditions. This may be related to the bulk hydraulic conductivity of the polysaccharide suspension. Therefore, any further reduction of hydraulic conductivity may require the modification of the suspension properties or a reduction in the base material porosity. Where soils were mixed with a biopolymer solution and then compacted, the moulded moisture content had an effect on the final hydraulic conductivity achieved. The optimum moisture content for minimum hydraulic conductivity appears to be several percent wet of compaction optimum. The optimum moisture content for minimum hydraulic conductivity appears to be several percent wet of compaction optimum. This may be related to providing sufficient water to achieve a suspension of low enough viscosity to mix evenly through the sample and to fill an optimum void volume. The curing time also had an effect on the reduction of hydraulic conductivity. A longer curing time generally achieved a lower conductivity. Based on these positive indications, the application of biofilms and biopolymers to im-

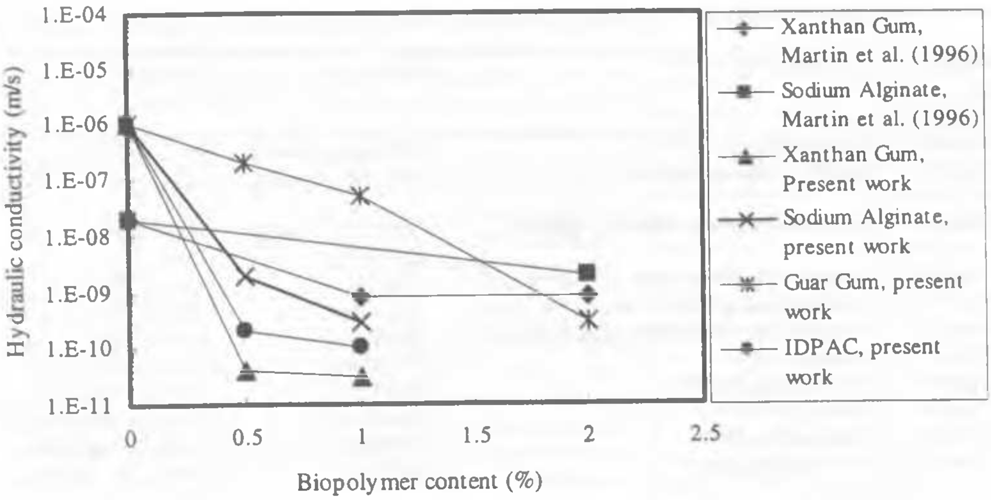

Figure 1: Hydraulic conductivity of soil biopolymer mixes

prove marginal materials for waste containment facility liners appears to be worthy of further assessment.

7 POTENTIAL FOR FURTHER INVESTIGATION AND CONCLUSION

In addition to the beneficial reductions in hydraulic conductivity already achieved, there are other aspects of biofilms and biopolymers, which make their use attractive. Some of the properties that are considered to be worthy of further investigation are briefly discussed below.

7.1 *Ion Exchange, Gelation and Mineral Deposits*

Due to the action of the functional groups on the polysaccharides, which are the basis of both biofilms and biopolymers, some substances exhibit ion exchange. This property could be exploited, for instance, in the beneficial adsorption of heavy metals.

Some polysaccharides, such as sodium alginate, set when mixed with divalent cations, such as calcium. This property may be used to further reduce the bulk hydraulic conductivity of the biopolymer. It may also enhance the strength increase effect of biopolymers on soil.

Some biofilms produce mineral deposits, which over a long period of time may increase the strength of the treated material, by bridging between particles. Mineral deposits may also decrease the hydraulic conductivity by reducing the porosity of the matrix.

7.2 *Colonisation, Biodegradation, Gas Production*

One of the benefits of biofilms over biopolymers as a method of improving soil properties, is the ability to migrate through a porous material without external intervention. It may be possible to induce a biofilm to evenly improve the properties of a layer by injecting bacterium into only part of the layer. Over time, the process of colonisation could result in the bacteria cells, and their associated biofilm, being evenly distributed through the layer. However, this characteristic also presents a potential disadvantage. If a biofilm was used in a lining system, which comprised of a drainage layer and/or leak detection layer, precautions would need to be taken to ensure clogging of these layer did not occur.

The introduction of bacterium, which produces biofilm and also breaks down contaminants of concern, may be able to create a low permeability reactive barrier. This technology could have applications in waste containment or contaminated site remediation.

Several researchers have noted gas production during testing. Generally it has been cited as a potential source of errors in hydraulic conductivity testing, as gas present in the sample reduces the apparent permeability. The gas production is generally related to sulfate reducing bacterium. The control of gas production through discriminating selection of bacteria or possibly the exploitation of gas production may be areas of interest.

Gas production has also been noted in soil-biopolymer mixes, where no bacteria have been artificially added. This may indicate that the polysaccharides in the biopolymers promote the growth of the indigenous soil bacteria. This effect may increase the biodegredation potential of soil-biopolymer mixes.

In conclusion, results of the different research programs discussed in this paper demonstrated the potential for using biopolymers or biofilms to create waste containment barriers. However, a large amount of additional research work is needed in order to address some remaining questions before any practical application can be considered.

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