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Sustainable environmental protection

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Impressum

Forschungsbericht 280:
Sustainable environmental protection

2007

Herausgeber:
Bundesanstalt für Materialforschung und -prüfung (BAM)
Unter den Eichen 87
12205 Berlin
Telefon: +49 30 8104-0
Telefax: +49 30 8112029
E-Mail: info@bam.de
Internet: www.bam.de

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Materialforschung und -prüfung (BAM)

Umschlag: Lutz Mittenzwei
Layout: BAM-Arbeitsgruppe Z.64

ISSN 0938-5533
ISBN 978-3-9811655-2-4

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Sustainable environmental protection*

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1 Introduction

Almost all (organic and inorganic) materials that support our life come from one of the biosphere's main components: the earth's crust; and they form a limited treasure-trove of mankind: natural resources. The materials must be extracted from the earth, whereby natural resources become inevitably depleted. They must be made suitable for human use, i.e. they must be processed using a variety of technologies. They are employed in virtually all branches of industry, and they produce a number of side products. At the end of their useful life, the residues or wastes must be disposed of. They come into contact with the earth's crust again and may pollute it.

With the world's population rapidly increasing, the need to support billions of people has forced technology to improve the efficacy of the methods used to extract, process, utilise and dispose of the materials indispensable for our life. The last two centuries have witnessed an incredibly sharp, exponential rise in the extent and intensity in the utilisation of these materials. The dazzling speed of this development has had, in a most unfortunate way, a serious impact on our environment, because efforts were focused on only taking and not giving back. The turnabout in our thinking came when contaminated air, water, soil and food alerted the public to the dangerous situation and made it clear that unless we quickly change our practice in exploiting natural resources, not only future generations but also we ourselves will fail to find the most essential support for life within the foreseeable future. In some cases, realising that the situation was moving out of control came too late, taking the lives or impairing the health of those most directly affected. Sadly, mankind is still not wise and disciplined enough (although we are gaining

knowledge), and a few more catastrophes will be needed to provoke badly needed decisions to restrict reckless exploitation of reserves.

Geotechnical and environmental engineering is at the forefront of this struggle for sustainable development, in which we are trying to develop reasonable damage control and to introduce methods to allow careful use of resources. Sustainability has become the conscientious and future-oriented principle of modern resource management and environmental protection, because scientists, engineers and politicians have realised that caring for the future is tantamount to providing manageable and healthy surroundings for ourselves.

The legacy of the past has been causing enormous difficulties in achieving our aims; but initiatives have been increasingly developed, which will guarantee that both our generation and those to come will do their best to preserve our main and almost exclusive source of materials, the earth. Although obtaining supplies from spaces outside our planet no longer sounds like absurd science fiction, economic considerations will make this kind of endeavour prohibitive for some time. For the foreseeable future, the primary concern for geotechnical and environmental engineers will therefore be to ensure a healthy balance between extraction, processing, manufacturing, utilising, recycling and disposal of materials and products. This will focus on achieving and maintaining a reasonable level of cleanliness for the three essential media with which we live in a close symbiosis: air, water and soil (Sarsby, Meggyes, 2001).

2 Our water resources

Life was created in water and water became our inseparable fellow traveller through history. Life is impossible without water; our body and those of animals and plants consist overwhelmingly of water. Half of our blood is water; water content is above 90 % in new-born babies and only drops below 70 % at old age. Descriptions of catastrophes show that people can survive without food over several weeks, while the chances of survival without water drop drastically after three days.

71 % of the earth's surface is covered with water: this is why the weather is mainly determined by meteorological

phenomena generated by the oceans. Changes among the three phases of water – i.e. ice, water and vapour – have not only contributed to shaping the terrain of the continents but keep influencing our everyday life even today. Major shifts in the phase balance of the earth's water reserves during glacial and interglacial periods have resulted in tremendous alterations in the extent of dry land, due to the rise or fall in sea levels.

The continents' supplies of fresh water stem from the oceans, where 97 % of the earth's water reserves are stored. The ongoing circulation of water from the oceans – through the clouds and rain, followed by water infiltration into the uppermost layers of the earth's crust, and then through the rivers back to the oceans – is maintained by the sun. This water circulation is of basic importance for all living creatures, since the main sources of drinking water are the water bodies in

* The papers published in this volume were presented at the Sustainable Environmental Protection symposium organised by the Federal Institute for Materials Research and Testing (BAM), Berlin for the 65th birthday of Tamás Meggyes on 24th February 2006.

various soil and rock layers. In Germany, for instance, about three quarters of drinking water supplies are extracted from groundwater.

Due to the inseparability of human and animal life from some form of fresh water, human settlements started near springs, rivers, lakes or, in exceptional cases, other sources of water (snow or ice). Our life and activities developed around the ancient cores of such water-side settlements over historical eras. Using the earth's mineral and vegetable resources, gradually we became able to produce objects useful in our everyday lives (tools, clothes, buildings etc.), plus those for helping to hunt and kill each other (weapons). By contributing to the manufacturing process our most valuable asset, the brain, we were eventually able to produce more sophisticated materials and objects (metals, chemicals etc.).

Technological developments, and increasing demand for better utensils, created more intensive large-scale agriculture and industry, which were not only able to generate value and wealth, but introduced a new and disadvantageous type of

activity: environmental pollution. One example is the Bitterfeld Region in Germany, where in the course of a century, open pit lignite mining and chemical industries have contaminated the groundwater over an area of 25 km² with a total volume of approx. 200 million m³. Similar examples of extensive, polluted areas can be found in Silesia, the British Midlands, the Ruhr District, Erzgebirge, Ural Mountains, Aral Sea, former uranium mining areas (e.g. Southern Hungary) etc.

Polluting the environment has not been a privilege of our generation alone: even the ancient Greeks managed to pollute land and sea with the waste products of their lead mining, metallurgy and processing, some 2000 years ago. But increasing speed in development and industrial revolution multiplied the amounts and types of emissions, and kept adding more complex types of contaminant to the established ones: chemicals, toxic heavy metals, dense non-aqueous phase liquids (DNAPLs), radioactive wastes etc. In doing so, they have generated a steadily increasing threat to our environment, and our drinking water in particular.

3 Groundwater remediation

This situation has called for remedies to prevent mankind from poisoning its own water resources. In the second half of the 20th century increasing efforts were made towards containing and cleaning contaminated groundwater. A number of techniques can be used to achieve such goals, traditionally either by treating or isolating the soil in-situ, or by removing it for washing or disposal. However, such technologies tend to be accompanied by high energy consumption and can lead to new environmental problems. More efficient and economical techniques are therefore needed to remediate contaminated soil and groundwater (*Simon et al.*, 2002).

The easiest approach appeared to be to apply well-established methods of waste-water treatment. This gave rise to the so-called 'pump-and-treat' method, in which contaminated groundwater is removed from the ground by pumping and treated in a treatment plant on the surface. It has the advantage of using proven techniques, is easy to control and the treated groundwater can be re-injected into the ground or discharged in rivers or lakes. The main disadvantages are that it disturbs the groundwater flow regime and requires steady energy and other inputs. Also, certain slowly emitted contaminants, polyaromatic hydrocarbons (PAHs) with low bio-availability and contaminants in heterogeneous sediments are not easily accessible for this technique.

In contrast, permeable reactive barriers (PRB) are a relatively new technology for groundwater remediation: a trench, arranged downstream of the contaminant source and filled with granular reactive material, enables the treatment of contaminated groundwater passing slowly through driven by natural groundwater flow. Chemical, physical and biological methods have so far been used successfully, with reduction, precipitation, adsorption and oxygen release to promote biodegradation as the most common reactions. The targeted chemicals to be removed from the groundwater by the reactive agents are either decomposed to other, less dangerous compounds or fixed to the reactive material. Their mobility, availability and toxicity may be significantly reduced in both cases (*Powell et al.*, 1998). Groundwater remedia-

tion using permeable reactive barriers is an in situ method with low energy demand and offers the prospect of a more cost-effective remediation technique, significantly enhancing natural groundwater protection.

Many reactions applied in PRBs are similar to or the same as those occurring in natural attenuation, which may be defined as a means by which the concentration of groundwater pollution is reduced to an acceptable level by natural processes. In that context, adsorption, biological degradation, cation- and anion-exchange, dilution, filtration and precipitation reactions are the dominating attenuation mechanisms. The main advantage of natural attenuation is its cost-effectiveness: in most cases costs only occur in connection with monitoring. Its disadvantages are the long time requirement to provide results, and the limited possibility of influencing the processes. Attempts to provide more favourable conditions to accelerate attenuation (enhanced natural attenuation) tend to involve some of the techniques and expense of other approaches. Research and development is under way, in Europe and world-wide, on contrasting approaches to these remediation techniques (*Holzöhner, Meggyes*, 1995).

To investigate long-term behaviour of permeable reactive barriers and enhance their efficacy, the Long-term Performance of Permeable Reactive Barriers used for the Remediation of Contaminated Groundwater (PEREBAR) integrated research project, funded by the European Union, was carried out (*Roehl et al.*, 2005). Hydroxyapatite (HAP) and elemental iron proved the most efficient materials from among the materials tested in removing uranium from aqueous solutions. The most likely uranium attenuation mechanisms are bulk precipitation when HAP is used and a combination of reductive precipitation and adsorption for elemental iron. Ageing mechanisms due to alteration of the reactive material, change in permeability, formation of precipitates, coatings etc. were investigated using accelerated column and container tests. The experiments on elemental iron showed a reduction in hydraulic permeability which may be attributed to precipitation and formation of secondary minerals (calcium, magnesium and

iron carbonates). HAP ageing was dominated by a gradual consumption of HAP due to its solubility in groundwater.

A silica coated with modified polyacryloamidoxime (PANSIL) was developed which efficiently removes uranium from aqueous solutions. Both the support matrix (sand) and the coating are durable, the sorption is most effective when the solution pH is between 4 and 8, specificity for uranium is fairly high, and there is no risk of precipitation of by-products that can block porous barriers. An electrokinetic fence may be installed upstream of the barrier to reduce the concentration of groundwater constituents that might impair barrier

function, by preventing them from being transported by the groundwater. In laboratory experiments, charged species were effectively precipitated around the electrodes, thus hindered from moving with the groundwater flow toward the reactive barrier. In an area in Southern Hungary contaminated by uranium mining, a 6.8 m long and 3.8 m deep experimental PRB was constructed with 38 t of elemental granular iron to remove uranium from the groundwater. The PRB proved very efficient: uranium concentration dropped from approx. 1,000 µg/l in early 2002 to about 100 µg/l after the start-up the PRB.

4 Hydraulic transport of solids

Extracted minerals must usually be transported to other places for processing or other place of utilisation. Since the amount to be transported is frequently in the range of millions of tonnes, there is a high risk of polluting the environment with dust, solids particles and chemicals along the transport routes. Therefore methods that do not cause pollution during transport offer great advantages from the environmental protection's point of view. Hydraulic transport is such a method where granulated solid materials e.g. coal, gravel, sand, ores etc. are mixed with water and pushed by pumps through pipelines, analogous to oil pipelining. In addition to a closed system which keeps pollution at a minimum, simple construction, high degree of automation, low labour requirement and low overall costs are further advantages of this method.

Improvements in our understanding of the fluid mechanics of solid-liquid two-phase mixtures can provide an important contribution to developing of the hydraulic transport technology. There are three flow patterns in which the solid-liquid flow systems can be categorised:

1. Flow of mono-phase, homogeneous, non-settling mixtures, also called homogeneous suspensions.
2. Flow of two-phase, heterogeneous, non-settling mixtures.
3. Complex flow of solid-liquid mixtures, where the fines and the liquid form a new homogenous phase and the coarser particles are transported in this carrier phase.

Mixtures are considered as mono-phase if solids are as finely distributed in them as to lose their character as single solid particles and the mixture behaves as a homogeneous liquid. The rheological properties of such an apparently homogeneous liquid differ substantially from those of the liquid component, i.e. the pure transporting liquid. When designing a system for the transport of mono-phase mixtures, their properties must be determined by experiments to obtain the factors that can be transferred to the pipeline to be designed.

In two-phase liquids (i.e. when the solid and liquid components can easily be separated), forces holding the solids in suspension are generated by the flow of liquid surrounding each solid particle, and also by the fluctuations caused by turbulence. The solid particles are lifted and introduced into the liquid current by the turbulent vortices, where, if they are small enough, they will be maintained in suspension. The

degree of homogeneity of this suspension increases with increasing flow rate. The interaction of the relevant forces is further complicated by the fact that the turbulence is influenced by the presence of solids.

The behaviour of mono-phase mixtures can be understood by studying the rheology of mixtures. As regards the process of flow in a pipeline, the most important criterion is the Reynolds number, which is the ratio of forces of inertia to those of viscosity within the liquid system. Another significant parameter is the criterion of dynamic similarity, expressed by the Froude number, by means of which the inertia and gravitational forces can be taken into account for every type of liquid system.

Solids immersed in liquid are prone to being lifted or sunk, depending on whether their density is lower or higher than that of the liquid (usually water). The majority of the materials to be transported hydraulically have a higher density than the transporting liquid, water being the most extensively used shipping medium.

Materials less than 40 µm in size easily form a suspension in the transporting liquid, even within the range of laminar flow. In addition, by controlling the distribution and concentration of particles larger than 40 µm, a suspension can be prepared that remains largely homogeneous even for a longer standing time.

It is characteristic of settling slurries that the solid and liquid phases can be well discerned, and the viscosity of the liquid phase does not rise when in contact with solid particles. In cases where the solid material consists partly of very fine particles forming a homogeneous suspension with water, the resulting "heavy liquid" has a viscosity and density that show a major divergence from those of the pure liquid. The suspended fines are no longer solid particles immersed in water, but have to be regarded as constituents of the newly formed liquid. The latter and the larger particles behave as two separate, liquid-solid, phases. Since the density of the new liquid exceeds that of the water, the solid phase is subjected to a greater buoyancy, which is very favourable in reducing the pressure loss due to friction.

The motion of solid particles in a flowing liquid is strongly affected by their shape, particularly if they are small in size. The mechanism of energy exchange between liquid and solids is characteristic of the system of flow in which particles move (e.g. saltation, heterogeneous suspension, etc.). The

magnitude of forces bringing about the motion of particles is, for any flow system, proportional to the velocity difference of the two phases. The concepts of the interaction between the two phases are mainly based on empirical relationships expressing the relationship of pressure losses due to friction and some other basic parameters. For this reason, the majority of relationships used for design purposes, are based on empirical formulae. Research workers, who have succeeded in deriving theoretical relationships expressing the pressure loss, were in many cases obliged to modify the constants in their “theoretical” formulae (Meggyes, 1976).

5 Tailings facilities

One of the disadvantageous features of the extraction of useful materials (ores, fuels, building materials etc.) from the earth is that usually large amounts of waste materials are produced resulting in a number of technical, financial and environmental problems. Tailings are probably the most characteristic representatives of wastes accompanying mineral extraction. Tailings are fine-grained waste products from the milling process of the mining industry which emerge after the raw materials (e.g. metals) have been extracted from the ore. These residues are mixed with water during processing, thus they emerge as slurries. The tailings are deposited in so-called tailings ponds which are the main components of tailings facilities and are surrounded by man-made embankments or dams. Failure of these facilities may result in uncontrolled spills of tailings, flow-slides and the release of poisonous chemicals (Meggyes *et al.*, 2007).

With the development of technology, in particular, when froth flotation was introduced, the mining of raw materials with low useful valuable material content became feasible. In flotation, the valuable mineral particles are attracted to tiny bubbles that rose to the surface of a liquid and then can be skimmed off. For this to be done, the ore has to be ground to very fine sizes, which also reduces the size of the tailings particles. It also enables lower-grade ore bodies to be exploited, hence increasing the volume of the tailings. The invention of gold cyanidation where gold is converted to water soluble sodium cyanoaurite had similar impact on tailings production. Disposal of the tailings therefore produced growing volumes of finer tailings particles that are discharged into tailings ponds with engineered dams. The human factor has always played an important factor in failures, thus owners, operators, designers and regulator can contribute to enhancing safety (Gipson, 2003).

Since tailings disposal is accompanied with considerable costs, mine operators in the past aimed to dispose of the waste as cheaply as possible. Most mining companies are now aware of the potential dangers posed by poorly designed, constructed or operated tailings disposal facilities. Tailings dam construction has evolved, and gradually these structures have become major edifices, applying the principles of geotechnical engineering (Sarsby, 2000). Currently, the highest tailings dam is 230 m high (Vick, 2000), while the storage capacity of an oil-sands tailings facility in Canada is 270 million m³ of sand plus 320 million m³ of sludge - probably the largest in the world (Morgenstern and Kupper, 1988). Tailings disposal or management facilities are usually continually constructed as more tailings are produced

In addition to fluid mechanical investigations, those topics extensively researched included the development of special instruments for the measurement of concentration, pressure, pressure difference, velocity and flow rate of solid-water mixtures, including industrial-scale equipment. Special devices (jet pumps, jet dredges, pipe chamber feeders) were developed and tested, transient phenomena analysed and measured. Laser-Doppler velocimetry was used for the measurement of fluid velocity and turbulence.

from the mineral extraction process, i.e. the height of the structure is gradually increased as storage capacity requirements increase with the advantage of spreading the costs over a long period of time, but meanwhile the operators may not maintain constant diligence in the management of these structures (ICOLD, 1996 and 2001).

In many cases the tailings transported by pipelines to the tailings pond are separated by hydrocyclones and the coarse fraction is used for dam construction. This is a major cost-saving feature, but it may contribute to lowering the safety of the facility. A major improvement potential lies in the quality control of the materials used in tailings dam construction. Tailings consist of cohesionless, highly erodible particles, which, when in a loose and saturated condition, are prone to liquefaction when subjected to seismic shocks (Sarsby, 2000). The fine-grained tailings are stored behind the dam, forming a soft, loose, relatively impermeable material which is usually saturated and likely to liquefy when vibrated. A liquefied mass of tailings behaves like a high-density liquid, and exerts a considerable hydrostatic pressure on the dam. The high content of fine-grained material in the tailings gives rise to long settling periods – sometimes several years or even decades. The accumulation of rainwater in humid areas provides a steady water supply which needs to be controlled even after closure of the facility. Drainage of ‘stubborn’ slurries requires complicated geotechnical methods using geosynthetics, the loading of large areas and constructing drainage wells (Biczók, 2001). Overtopping of the dam may rapidly lead to crest erosion and catastrophic failure of the dam. In addition, tailings, especially those from cyanidation, contain (toxic) chemicals, which may infiltrate into the ground or groundwater or cause extensive pollution in the case of spillages. Chemical compounds added as reagents during the ore processing (e.g. to extract valuable metals from the ores to facilitate flotation) or released from the ore as a process by-product (e.g. heavy metals) can aggravate the hazardous nature of tailings deposits. Especially cyanide is of major concern due to its high environmental mobility and toxicity (EPA, 1994; Logsdon *et al.*, 1999).

The main elements of tailings management facilities are the dam (also called embankment or containment structure) and the tailings beach and pond or lagoon. Since most construction methods employ separation of the transported slurry by hydrocyclones (except gold tailings which are usually too fine), with the coarser fraction becoming the dam material and the finer fraction being stored behind the dam, there is no sharp dividing line between the dam and the pond. The most com-

mon method of construction is the 'upstream method'. Using a starting dyke, the dam is raised step-by-step. Each step is constructed partly on the previous one and partly on settled tailings. This method has stability implications, inasmuch as the settled tailings often fail to provide a stable enough foundation if the rate of construction for the rising dam is too high. The 'downstream' method provides greater stability, but more coarse construction material is needed (EU, 2003).

The most common causes of failure are internal erosion, liquefaction and overtopping. The single most important factor governing the stability of tailings dams is whether or not they retain ponded water on the surface of the tailings deposit (Vick, 2000). In spite of a general increase in safety awareness, an increase in the number of fatalities caused by catastrophic accidents at tailings management facilities over the last decade cause concern.

Tailings facilities failures, e.g. leaking ponds, tailings overtopping and dam breaches, have all resulted in major hazards, such as

- floods and flood waves,
- spills of sometimes toxic sludge and water (e.g. 3 million m³ of sludge and 4 million m³ of water were released in the 'Aznaicóllar' accident in Spain in April 1998, and covered great areas in the Doñana National Park, a nature reserve area),
- spills of sludge and rubble, burying houses or settlements and their inhabitants or destroying valuable cropland (e.g. the 1996 tailings spill in Sgurigrad, Bulgaria, destroyed half of a village downstream, claiming 107 lives),
- pollution of rivers and their flood banks, e.g. cyanide and hypochlorite were spilt into the Tisza river following the

'Baia Mare' accident in Romania in January 2000 (Brauns, 2000) and spread as far as the Black Sea,

To enhance safety of tailings facilities, the Sustainable Improvement in Safety of Tailings Facilities (TAILSAFE) integrated research project, funded by the European Union, has been carried out (Roehl and Czurda, 2007). From the work performed and experiences gained, the following key conclusions have been drawn:

- Water management and monitoring, pore water pressures and seepage are the most important safety-related issues.
- High concentration tailings (thickened tailings and paste technology) are a promising technique for increasing the stability/safety of tailings bodies (Jewell *et al.*, 2002, Meggyes and Debreczeni, 2006).
- Geophysical non-destructive investigation techniques (especially geoelectrical) are suitable to supply valuable information on the internal structure of tailings dams and bodies and their surroundings, on areas of high water content and seepage, and on the spread of inorganic contaminants.
- Human actions and behaviour play a crucial role in the construction and operation of tailings facilities.
- Training and qualification of all personnel and authorities involved is of utmost importance.
- The Tailsafe Resource is an online parameters database that can contribute to increasing a user's awareness of surface tailings storage operations and related risks. It can also be used as an Internet training tool.

6 Landfills

Large amounts of wastes generated by households and industry bear a vast potential for polluting the environment. Before the passage of environmental regulations, wastes were dumped indiscriminately in open areas with no care for the environment. Many countries have developed stringent regulations on waste disposal and landfills in the last few decades. Today, landfills are engineered structures that are aimed at containing the wastes effectively. Upon stabilization of municipal solid waste, the landfill sites may be used for a beneficial purpose such as golf course, recreational park, industrial park and other such uses (Meggyes 2007).

Stief (1986) proposed the concept of a multibarrier system for landfills based on the following:

- 1 The subsoil (the geological barrier).
- 2 A basal liner and capping (the technical barrier).
- 3 The waste body itself.
- 4 The control of landfill performance.
- 5 Aftercare or post-closure plan.

The most important element for containment is the technical barrier, the standard for which is a composite liner throughout most of Germany. Landfill technology has evolved with

well-defined design and construction procedures. The salient features of landfill technology are waste separation and pre-treatment, suitable landfill concept, standard composite and alternative liner systems, design of earthen and geomembrane liners, safety analysis of liners, leachate drainage systems, and landfill gas management. Cut-off walls are a useful means of pollution control around landfill sites.

The main landfill classes are:

Municipal solid-waste landfills.

Class I. Virtually inert waste, e.g. Ni content ≤ 0.2 mg/l.

Class II. Waste with higher pollutant contents, e.g. Ni content ≤ 1 mg/l.

Hazardous solid-waste landfills, e.g. Ni content ≤ 2 mg/l.

Generally, landfills must be provided with a composite basal liner and a capping, both of which are composed of sealing, protective and drainage layers, although alternative liners with proven barrier equivalence can be used (August *et al.*, 1997; Müller, 2001). The capping system's main function is to prevent gaseous pollutant emission into the atmosphere and rainwater from percolating into the landfill so as to keep leachate generation to a minimum, while basal liners must

protect the surrounding soil and groundwater from leachate pollution. Both the sealing layers are overlain by a drainage system which in the basal liner collects leachate and transports it to a treatment plant to be cleaned of pollutants and subsequently discharged in the nearby surface water. A capping drainage layer collects rainwater which requires no processing before being released.

The most important element of these systems is the composite liner consisting of several lifts of earthen (mineral) material with a minimum thickness of 0.25 m of each lift and a certified geomembrane minimum 2.5 mm thick. The geomembrane is installed on top of the earthen layer in intimate contact with it and is protected by a protective layer against damage by gravel or aggregates in the drainage layer (Müller 2006). The permeability of the earthen layer of basal liners for all classes and in the capping system of hazardous solid waste landfills must be $k \leq 5 \times 10^{-10}$ m/s, and for Class II capping systems $k \leq 5 \times 10^{-9}$ m/s.

A composite liner must be constructed so that the geomembrane (most commonly HDPE) will lie sufficiently flat on the flawless surface of the mineral liner and that subsequent surcharges such as the protective, drainage, waste and recultivation layers produce a complete contact, known as intimate contact, between them. The geomembrane must be smoothed and ballasted after placement to prevent installation failures. Stability and the friction behaviour between the geomembrane and adjacent layers on slopes is critical and geomembranes with rough or textured surfaces are especially suited to inclined areas by virtue of their high coefficients of friction. To avoid any tensile overload of the geomembrane in basal liners, shear resistance between the geomembrane and protective layer must not exceed that beneath it, i.e. between the earthen layer and the geomembrane (Müller, 2006).

Instead of composite liners, alternative liners can be used if they can prove to be equally effective as composite barriers. Alternative liners currently in use include: mineral liners, multiple liner with a sorption layer, improved mineral liners, asphalt liners, geosynthetic clay liners, capillary barriers

Landfill gases are the result of microbial decomposition of solid waste. Gases produced include methane, carbon

dioxide and lesser amounts of other gases (e.g. hydrogen, volatile organic compounds and hydrogen sulphide). Regulations require that landfill gas should be monitored to ensure that methane concentration does not exceed 25 % of the lower explosion limit. At large landfills, the collected gases (passive or active gas collection) are used for energy recovery. Landfill gas recovery systems can reduce landfill gas odour and migration, and the danger of explosion and fire, and may be used as a source of revenue that may help to reduce the cost of closure (Sharma and Reddy, 2004).

Landfill mining is a method of treating old landfills. After eliminating emission sources, the waste body is excavated, the remaining waste compacted and useful materials are separated and recycled. As a result, landfill void space is increased, pollution potential reduced and financial gains are achieved by material utilisation (Spillmann *et al.*, 2007).

Safety analysis of landfill liners is based on the assessment of the effects of an incident (e.g. leakage), assuming a worst-case scenario. It is aimed at estimating groundwater quality in the wake of leachate escape into the subsoil. The safety of a liner (sealing) system is measured by the extent of leachate containment. A liner (sealing) system is considered safe (suitable) if it provides a sufficiently strong barrier against certain pollutants or their mixtures in leachate over a long period of time. A barrier effect is considered suitable if the liner (sealing) system yields the same leakage behaviour as a standard liner system (i.e. a composite liner) (Heibroek and Jessberger, 1995). Since landfill liners are expected to maintain their efficacy over periods of time several orders of magnitude greater than are conventional engineering structures, long-term behaviour and very slow physical processes are of special interest. Transport processes, desiccation in earthen layers and conditions which may endanger geomembranes, protective layers and drainage systems are fairly well understood. The mechanical properties of earthen materials can be improved and testing methods and apparatuses are available for the determination of critical values. Safety analysis shows that composite liners are extremely safe under the right conditions and, therefore, represent the standard by which the efficacy and equivalence of alternative liners should be measured (August *et al.*, 1997).

7 Climate change

Global warming is probably the most serious challenge threatening our environment. The globally averaged surface temperature has increased over the 20th century by about 0.6 ± 0.2 K and is projected to increase by 1.4 to 5.8 K (Houghton *et al.*, 2001) or by 1.8 to 6.4 K (IPCC, 2007) over the period 1990 to 2100. Rising temperatures brought about by climate change will cause microorganisms in the world's soils to decompose organic matter more rapidly, releasing extra carbon dioxide and accelerating climate change (Knorr *et al.*, 2005). Since the world's soils hold about 1,500 Gt of organic carbon (atmosphere: 720 Gt, vegetation: 600 Gt), relatively small changes in the flow of carbon into or out of soils could be significant on a global scale (Powlson, 2005).

The symptoms of climate change are unmistakable. It is very likely that the 1990s was the warmest decade and 1998 the warmest year in the instrumental record since 1861

(Houghton *et al.*, 2001). There has been a widespread retreat of mountain glaciers in non-polar regions during the 20th century. Arctic sea-ice thickness is likely to have decreased by 40 % during late summer. Global average sea level rose between 0.1 and 0.2 metres during the 20th century and is projected to rise by 0.09 to 0.88 metres (Houghton *et al.*, 2001) or by 0.28 to 0.59 metres (IPCC, 2007) between 1990 and 2100. Global ocean heat content has increased since the late 1950s. It is likely that precipitation has increased by 0.5 to 1 % per decade in the 20th century, the frequency of heavy precipitation events by 2 to 4 %, over most mid- and high altitudes but decreased over sub-tropical land areas of the continents on the Northern Hemisphere.

Real situation and trends may have been subdued by "global dimming" caused by particulate matter in the air, originating from Diesel engines, heating equipment, vehicle tyres, brakes

and road surface. The very fine particles form into polluted clouds which in turn collect small water droplets which act as heat reflecting mirrors with dramatic consequences on cloud pattern distribution and earth surface sunlight intensity. Drops in sunlight/solar energy attributed to this effect have now been established worldwide varying from 10 % over Antarctica and up to 50 % over industrialised countries. This means that the climate may in fact be more sensitive to the greenhouse effect than thought (*Stanhill and Cohen, 2001, Liepert, 2002, Roderick and Farquhar, 2002, BBC, 2005*). Climate change over the next 100 years will be driven by changes in greenhouse gases (GHG) and aerosols (*Cox, 2003*).

Dam failures, floods and landslides belong to the gravest consequences of extreme weather phenomena. The 1997 and 2002 floods in the Czech Republic, Germany and Poland and the 2003 heat wave in Europe were serious weather events, but they are dwarfed by the 1975 catastrophic dam failures in China. A typhoon accompanied with a metre of rainfall within three days triggered 62 dam failures in succession and 600 million cubic metres of water escaping from the reservoirs killed 85,000 people and severely affected 11 million (*Yi Si, 1998*). More than 1,800 people lost their lives in Hurricane

Katrina and in the subsequent floods in 2005, making it the deadliest U.S. hurricane since 1928 causing more than € 70 billion in damage, the costliest natural disaster in U.S. history. *Early 2007*, Kyrill was probably the most severe storm in Europe ever: wind speeds reached 200 km/h, the storm claimed about 40 lives, caused an estimated damage of up to 7 billion € and forced the German Rail (Bundesbahnen) to stop all of his trains for a night – a safety measure which never happened in post-war Germany. Tailings facilities and dams, under adverse weather conditions, also pose a serious risk to human health and the environment, the most recent European accidents being Baia Mare, Romania (triggered by an extreme weather event in 2000), *Aznalcóllar*, Spain (1998) and *Stava*, Italy (1985), the latter claiming 268 lives (*Sarsby, 2000, Anon, 2003*). Loss of human lives is often followed in the aftermath of natural disasters by a disrupted infrastructure (drinking water reservoirs, water networks and sewage systems) threatening the health of survivors.

The most important finding of the “Climate Change 2007” IPCC 4th Assessment Report is that man is at 90 % probability responsible for climate change (*IPCC, 2007*)

8 Outlook

Consequences of climate change are aggravated by the Earth's increasing population. Limited areas available lead to a competition between concurrent uses of land and soil (food production, living space, infrastructure and industrial production) due to concentration of activities in a small space (*EEA & UNEP, 2000*). Thus areas exposed to flood risk and land slides are increasingly being used for building and industrial production, which makes the consequences of floods even more devastating. There is a clear link between climate change, sustainable development, environmental quality and soil degradation.

An increased temperature in the atmosphere is tantamount to higher energy content which is likely to increase the probability of extreme weather events. This euphemism translates to torrential rains, floods, hurricanes, droughts and following incursions into the earth's crust, often causing miseries to millions of people, even civil wars (*Darfour, Sudan*). Due to higher surface temperatures an increased portion of precipitation will fall in the form of rain rather than snow, increasing both soil moisture and run-off. However, increased heating will lead to increased evaporation and this will decrease the availability of soil moisture needed both for natural vegetation and agriculture in many places. Water content is a major factor in determining slope stability. Climate change affects groundwater and therefore influences the condition and stability of existing slopes, both natural and man-made. If any of the embankments (roads, railways, spoil tips, landfills, tailings ponds, repositories for sewage and dredging sludges, water reservoirs and drainage systems, dykes) become unstable then residents living adjacent to the site and the local groundwater system will be detrimentally affected. Potential threats due to climate change through slope instability include movement of materials from slopes, which may result in sedimentation, blockages, flooding, damage to infrastructure, exposure of wastes and contaminated land to precipitation, generation of leachate, loss of vegeta-

tion and biodiversity, instability and failure of flood protection systems (*Sarsby, 2007*). Physical damage caused by landslides and slope failure may be aggravated by exposure and release of contaminants.

Climate change will influence our habitat in previously unimaginable ways, mainly due to changes in living conditions for animals and plants. Termites have spread in and near harbours under temperate climate (e.g. Hamburg, Germany) where they have been introduced by ocean-going ships from the tropics and found suitable conditions around heating pipes, cables etc. Even more intriguing are bioinvaders – plants and animals spreading by global trade and tourism and an increased temperature enabling them to expand their living space. The newly conquered territories are free from the predators of their homeland, so they can proliferate virtually without any limit: examples are *Achatina fulica* or the giant East-African snail in Brazil, the Burmese python in Florida, the Japanese knotweed in the UK, the malaria mosquito (*Anopheles*) in Europe and, unfortunately, 500,000 more. The opposite applies to certain species which lose some of the life-supporting conditions by the increased temperature. The harlequin frog, which had thrived in the Monteverde mountains of Costa Rica for perhaps a million years, became extinct: a minor warming created ideal conditions for chytrid-omycosis, a disease caused by waterborne fungus which has killed these frogs by blocking their natural ability to absorb water through their porous skin. An estimated 67 % of the 110 species of *Atelopus* (brightly coloured toads), which are endemic to the American tropics, have met the same fate, so a mass extinction is already in the making (*Pounds et al., 2006*).

Sadly, we will “need” a few more catastrophes until governments will be forced to take badly needed decisions to efficiently protect the environment and develop a slightly new strategy: adaption (*Pittcock, 2005*). Only then will the sustainable environmental protection attain the importance it deserves.

Acknowledgements

Support by the Federal Ministry for Education, Science, Research and Technology (BMBF) to the Integrated Research Programme 'Advanced Landfill Liner Systems' (1440 569A and 1440 569I) and by the European Commission to the 'Long-term Performance of Permeable Reactive Barriers

used for the Remediation of Contaminated Groundwater' (EVK1-CT-1999-00035) and 'Sustainable Improvement in Safety of Tailings Facilities' (EVG1-CT-2002-00066) is gratefully acknowledged. The issues in this paper have been the key topics covered during a 40-year research activity.

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Groundwater remediation using permeable reactive barriers

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Abstract

Permeable reactive barriers (PRBs) offer an alternative as opposed to more cost-intensive active methods for groundwater remediation, e.g. pump-and-treat. Long-term performance of PRBs, however, is crucial for the technology's success. The paper summarises the results of long-term column experiments with elemental iron (Fe^0) and hydroxyapatite (HAP) as reactive materials for the removal of uranium from groundwater. Good removal results have been achieved using elemental iron with possible reaction paths being reductive precipitation and adsorption on to corrosion products of Fe^0 .

Uranium can also be removed by dissolution of HAP and subsequent precipitation of low-solubility uranyl phosphates such as autunite $\text{Ca}(\text{UO}_2)_2(\text{PO}_4)_2$ or chernikovite $\text{H}_2(\text{UO}_2)_2(\text{PO}_4)_2$. Other possible reaction mechanisms for uranium removal with HAP are ion exchange processes and surface sorption with HAP (on two possible surface groups).

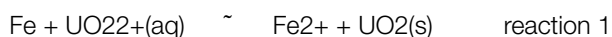
At the former uranium mining site in Pécs (Southern Hungary) a pilot PRB system was installed. Results from operation are reported.

1 Introduction

Uranium contamination in groundwater resulting from uranium mining and processing occurs world-wide at numerous locations. In view of its toxicity rather than radioactivity, a limiting concentration of 0.015 to 0.030 mg/l is currently being considered (Giddings and Fawell 2004).

Approximately one third of the global uranium production prior to 2000 was mined in Europe. Table 1 displays the production of uranium of the world's largest producers (OECD 2001). The data for the Russian Federation and several other states are based on IAEA secretariat estimation.

Uranium occurs mainly in the oxidation states +4 and +6. Hexavalent uranium, mainly in the form of the uranyl ion UO_2^{2+} is more mobile than U(IV) compounds. Good removal results have been obtained by the reduction of U(VI) to U(IV) by elemental iron:



The reduction of UO_2^{2+} is thermodynamically favourable as illustrated in Figure 1. The solubility of uraninite UO_2 is in the range of 10^{-8} mol/l for pH values between 4 and 14. Below

pH 4 uraninite is soluble. The behaviour of uraninite, the main constituent of the uranium mineral pitchblende, has been intensively studied because of its application as nuclear fuel (Bickel *et al.* 1996). The dissolution of spent fuel in deionized water under non-oxidising conditions was determined to be in the range of 10^{-9} to 10^{-5} mol/l. Under oxidising conditions UO_2 can be transformed into the uranyl ion and solubility is enhanced, especially in the presence of complexing agents. It is therefore important that Eh is below the U(VI)/U(IV) curve in the U(IV) stability field. In the presence of CO_2 at 0.01 bar, which is typical for groundwater conditions, it is more difficult to maintain uranium in the +4 oxidation state as the boundary line shifts towards lower Eh at high pH (see Figure 1).

An excellent review on the complex uranium solution equilibria is given by Guillaumont (Guillaumont *et al.* 2003).

Lowering the uranium concentration without changing the oxidation state is possible by the precipitation of sparingly soluble uranyl phosphates. With the addition of phosphate, hydroxyapatite (HAP) or bone char (HAP with a small amount

Table 1
Global producers of uranium (Uranium production total to 2000 in tons)

Country	Uranium production	Country	Uranium production
USA	353,796	Niger	81,853
Canada	340,523	France	73,664
Germany (former GDR)	213,380	Namibia	72,127
South-Africa	153,337	Gabon	27,872
Russian Federation*	114,223	Hungary	21,030
Czech Republic	107,080	Romania*	17,729
Uzbekistan	95,758	Bulgaria	16,720
Australia	91,157	Ukraine (since 1992)	10,000
Kazakhstan	88,372	China (since 1990)	7,435

* IAEA secretariat estimation

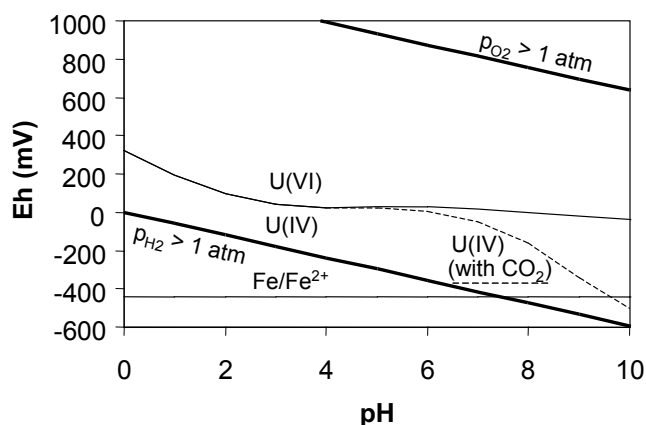
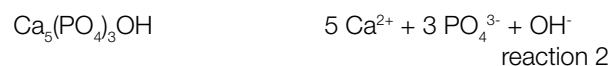
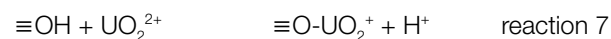


Figure 1
pH-Eh diagram showing the boundary line between U(IV) and U(VI) without and in the presence of 0.01 bar CO₂

of carbon) to the water (UO₂)₃(PO₄)₂ (log K_{sp} = -49.09 (Brown *et al.* 1981)), autunite Ca(UO₂)₂(PO₄)₂ (log K_{sp} = -47.28, (Brown *et al.* 1981)) or chernikovite H₂(UO₂)₂(PO₄)₂ (log K_{sp} = -45.48, (Grenthe *et al.* 1992)) may form. The mechanism of the interaction of uranium with HAP is not yet completely understood. Jeanjean *et al.* proposed a dissolution-precipitation mechanism (Jeanjean *et al.* 1995). With either autunite or chernikovite as the precipitation product the reaction may occur via the sequence displayed below.



Ion exchange processes (see reaction 6) or surface sorption using HAP (two possible surface groups, reaction 7 and reaction 8) are described by the following equations (Wu *et al.* 1991, Leyva *et al.* 2001).



In the work of Fuller *et al.* (Fuller *et al.* 2002) autunite and chernikovite have been identified as solid phases after adding uranyl ions to a saturated solution of hydroxyapatite. Evidence for adsorption of U(VI) to hydroxyapatite surfaces as an inner-sphere complex was found for certain concentration ratios.

Arey *et al.* postulated the formation of secondary phosphate phases but could not identify any controlling phase such as autunite (Arey *et al.* 1999).

The present paper summarises the results of column experiments performed using elemental iron and hydroxyapatite for uranium removal from groundwater.

2 Description of experiments

Experiments were performed with columns of 50 cm height and 5.9 cm inner diameter with a layer of filter sand at either end to achieve uniform flow conditions within the reactive material, consisting of either iron or HAP admixed with sand. Elemental iron was provided by Gotthard Maier, Rheinfelden, Germany (granulated grey cast iron, 0.3 to 1.3 mm), HAP by CFB Chemische Fabrik Budenheim, Germany (food grade quality, fine white powder, 99 % < 0.42 mm). Sand was normal filter sand (0.8-2 mm). The artificial groundwater (AGW) used for the experiments is characterized by high uranium, bicarbonate, and sulphate concentrations (Ca 80 mg/l,

Mg 41 mg/l, Na 60 mg/l, K 14 mg/l, SO₄²⁻ 300 mg/l, HCO₃⁻ 250 mg/l, Cl⁻ 50 mg/l). The experimental parameters are listed in Table 2.

In experiment (1) elemental iron was used in concentrations between 30 and 70 % in sand. The concentration of HAP was between 10 and 20 %. In experiment (2) iron was placed in two layers with different concentrations (5 and 15 %) in the column to avoid problems with clogging. HAP concentration in experiment (2) was 10 %. A more detailed description of the experimental set-up can be found elsewhere (Biermann *et al.* 2005).

Table 2
Experimental details for column experiments

	(1)	(2)
Concentration of uranium	0.49 mg/l	9.6 mg/l
Length of reactive zone in test column	0.4 m	0.2-0.3 m
Residence time	7-12 h	3-4 h

3 Results

In both column experiments Fe⁰ and HAP exhibited a uranium retention of more than 99 %. Effluent uranium concentrations were below the limit of detection (13 to 87 µg/l and 2 to 11 µg/l in experiment (1) and (2) respectively) of the spectrophotometric method employed for routine uranium analysis. Measurements with ICP-MS yielded maximum

effluent uranium concentrations of 1.3 µg/l corresponding to a uranium retention of at least 99.7 % for all columns. In experiment (1) no breakthrough was observed during 19 to 30 months of column operation, which corresponds to the treatment of 0.35 l to more than 8.7 l of AGW (1), with lower U concentration, per g of reactive material.

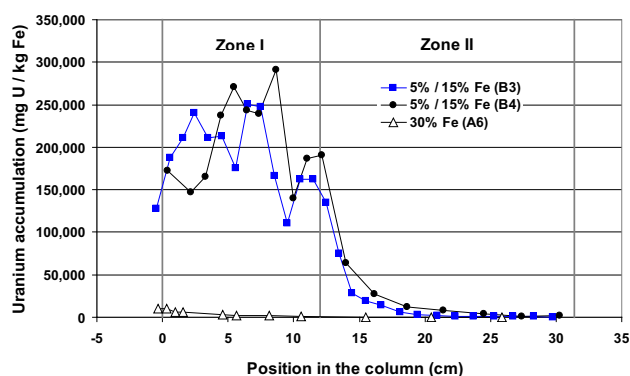


Figure 2
Uranium distribution in iron-bearing columns

After dismantling the columns the uranium concentration as function of column height was determined with chemical analysis. Figure 2 displays the uranium distribution in the columns with iron as reactive material. In experiment (1) the highest uranium content was found at the lowest point of the reactive layer, i.e. where the first contact of reactive material and pollutant occurred. In experiment (2) the reactive material was placed in two layers starting with a low concentration of 5 % of iron in sand. Most of the uranium was already removed

in this first layer. Note that the dimension of the y-axis is mg U/kg Fe, resulting in low values for experiment (1) where the iron concentration was high and the uranium concentration of the AGW lower than in experiment (2).

Reacted iron material was further analysed using an ESEM/EDAX system (environmental scanning electron microscopy with energy dispersive x-ray analysis). Figure 3 shows a micrograph of an iron particle and element maps of the same particle for oxygen, calcium, and uranium highlighting the existence of a thin uranium layer in the immediate vicinity of the iron surface.

In experiment (2), a breakthrough occurred in the HAP-column with effluent uranium concentrations exceeding the proposed limit of 15 µg/l in drinking water (Giddings and Fawell 2004) after the treatment of 3.2 l of AGW (2), with high uranium concentration, per g HAP (see Figure 4). Up to this point, however, more than 30 000 mg U/kg HAP had been retained in the column. After the average uranium load had reached 50 000 mg U/kg HAP the column was operated with AGW with no uranium to test for reversibility of the pollutant removal. Desorption was indeed observed, however, uranium concentrations in the effluent decreased with time. It is assumed that most of the uranium removed will be stable against remobilisation.

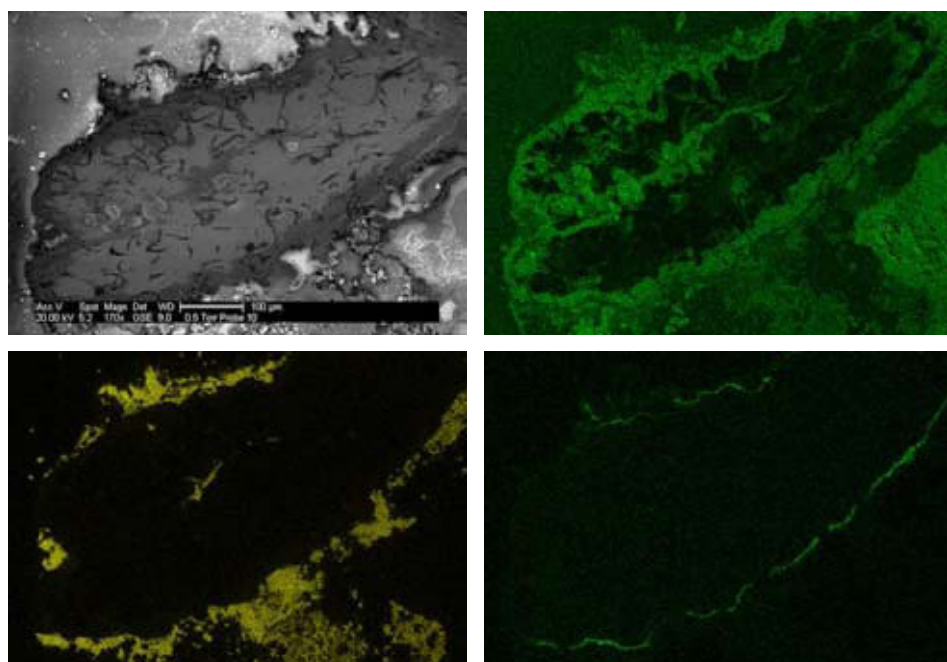


Figure 3
Micrograph (top left) and element maps of an iron particle with uranium bearing oxidation layer observed by an ESEM/EDAX system (top right oxygen, bottom left calcium, bottom right uranium).

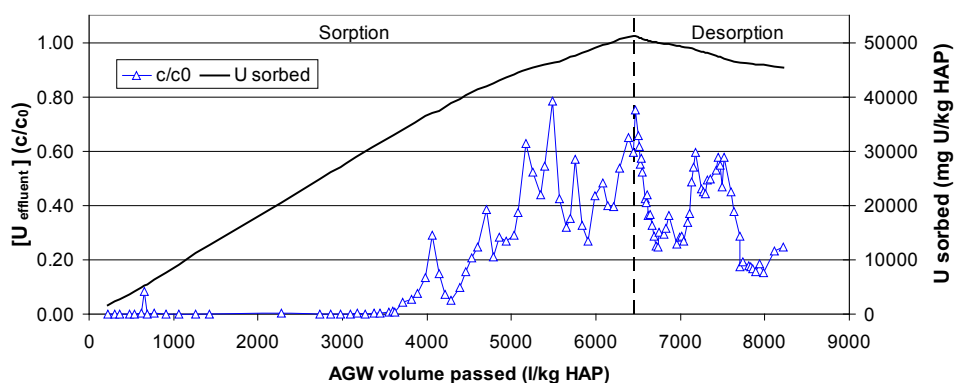


Figure 4
Breakthrough curve and subsequent desorption of uranium in experiment (2). At the vertical dashed line AGW (2) was substituted by AGW without uranium. c/c_0 is the ratio of effluent uranium concentration to influent uranium concentration during the sorption phase. The solid line indicates the cumulative sorption of uranium onto the column.

4 Field experiment

In Hungary, uranium was mined and processed in the southern part of the country, in the Mecsek mountains, near the city of Pécs. The mined out rock amounted to 46 million tons from which different wastes resulted. Mining activity was terminated at the end of 1997. Rehabilitation of the former industrial sites is in progress. Among different remediation tasks one of the most important issues is the restoration of the quality of groundwater contaminated by seepage from tailings, waste rock piles and heap leaching residues. A pilot-scale permeable reactive barrier was built at the site near Pécs during the course of the EU project PEREBAR (Csövári *et al.* 2005), www.perebar.bam.de). The PRB test facility has been in operation since August 2002.

The PRB consists of two different zones, zone I with a lower content of coarse elemental iron (12 % by volume or 0.39 t/m³, grain size 1-3 mm) and zone II with a higher content of fine elemental iron (41 % by volume or 1.28 t/m³, grain size 0.2-3 mm). On both sides (upstream and downstream) sand layers were inserted to distribute water inflow and outflow. The PRB is sealed with clay and geosynthetic clay liners at the bottom and with a geomembrane (high density polyethylene) at both sides and on the top. The design is displayed in Figure 5. The total mass of elemental iron installed as reactive material was 38 t, from which 5 t was coarser material.

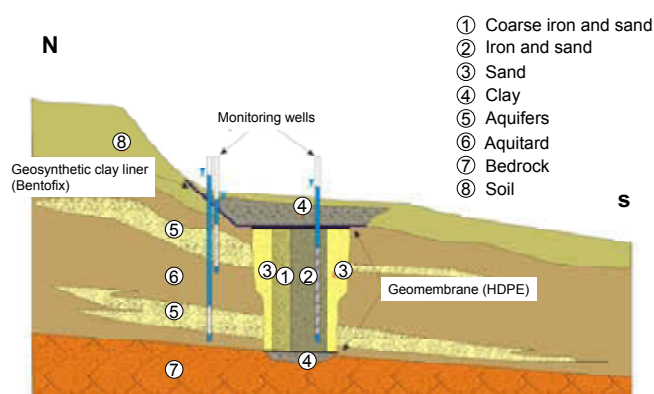


Figure 5
Sketch of the design of the experimental PRB near Pécs.

Good removal results for uranium (99 %) were obtained during the first 2.5 years of operation. Table 3 lists average values for different parameters for the operation period. It can be seen that the pH increases in the PRB by almost 2 pH units and that calcium is almost completely removed. The impact of precipitates on the long-term performance of the installation will be investigated in further research.

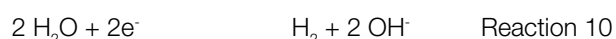
Table 3
Monitoring data of the PRB as of March 22nd 2005 after 31 months of operation.

Well	Location	pH	EC μS/cm	U(VI) μg/l	TDS mg/l	Ca mg/l	SO ₄ mg/l	HCO ₃ mg/l	Fe mg/l
PRB-1	Inflow	6.9	1400	940	1010	150	320	525	0.002
PRB-10	Zone I	7.3	1330	37	937	125	300	275	5.5
PRB-11	Zone II	8.7	865	10	550	10	185	299	0.03
PRB-14	Outflow	8.7	780	2	540	14	170	268	0.03
PRB-3 ^a	Downstream	8.5	420	56	240	5	95	116	0.017

^a Water is probably diluted by lateral water flow

5 Discussion

The experiments show that both elemental iron and HAP are able to retain uranium from groundwater. In the case of elemental iron the redox chemistry of the water is changed (see Figure 1) and the removal of pollutants is accompanied with an increase in pH due to iron corrosion (see Reaction 9 to Reaction 11).



As the solubility of calcite decreases with increasing pH, CaCO₃ precipitation occurs within a reactive treatment zone containing elemental iron. Alkaline earth elements are almost completely removed from the groundwater (see Table 3 for the results from the pilot PRB, TDS is decreased from 1010 to 240 mg/l). Almost 800 g of precipitation products

(i.e. alkaline earth carbonates) are formed for every litre that passes through the PRB system. This accumulation of solid material could lead to a loss of permeability and finally to a failure of the system. Figure 6 shows precipitates associated with reactive material from test columns and test boxes after operation with artificial groundwater with high alkalinity containing uranium.

Long-term column experiments with iron as reactive material showed that the pH increased initially by 2 pH units from pH 7 in the feed water to pH 9 in the column effluent. But after about 2 to 3 months the effluent pH decreased and stabilised around pH 8 after 7 to 8 months of operation (see Figure 7). This change in pH could indicate a change in the uranium retention mechanism from reductive precipitation (reaction 1, supported by the detection of uranium in the seam of iron particles, see Figure 3) to sorptive removal on iron oxyhydrate surfaces. More likely the lower pH values are

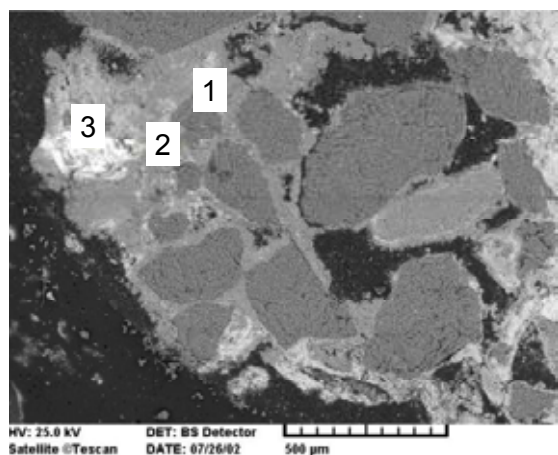


Figure 6

Image from secondary electron microscopy (SEM) from sand:iron mixture. 1 is a sand particle, 2 is mainly precipitated CaCO_3 , 3 is an iron particle. The surface is covered with $\text{Fe}(\text{OH})_2$ and FeCO_3 precipitates (Böhm et al. 2005).

indicative of the development of preferential flow paths as a result of precipitate build-up in the pore space and shorter residence times in the column (Kamolpornwiji et al. 2003). In contrast to this the pH of the effluent from columns with HAP as reactive material remained nearly unchanged.

The results with HAP are in agreement with the results of a technical demonstration at Fry Canyon in Utah using HAP (Naftz et al. 1999). In contrast to the reaction of lead with HAP where the sparingly soluble mineral hydroxypyromorphite $\text{Pb}_{10}(\text{PO}_4)_6(\text{OH})_2$ is formed after dissolution of HAP (Ma et al. 1993, Chen and Wright 1997) uranium removal might occur via sorption on HAP surfaces. In the non-mined Coles Hill uranium deposit phosphate mineral precipitation has been verified (Jerden and Sinha 2003). However, it was stated that the formation of autunite group minerals requires a longer time interval. Such a transformation was described by Sowder et al. for schoepite ($\text{UO}_2(\text{OH})_2$) (Sowder et al. 1996).

Whether sorption of uranium occurs via a sequence as displayed in reaction 7 or reaction 8 strongly depends on the speciation of uranium as a function of pH. The uranyl ion UO_2^{2+} is stable at low pH values only. If phosphate and carbonate are present, speciation is dominated by carbonate complexes ($\text{UO}_2\text{CO}_3(\text{aq})$, $\text{UO}_2(\text{CO}_3)_2^{2-}$ and $\text{UO}_2(\text{CO}_3)_3^{4-}$) at a pH of 8 and above. Between pH 4 and 7 soluble phosphato complexes ($\text{UO}_2\text{H}_2\text{PO}_4^+$, $\text{UO}_2\text{HPO}_4(\text{aq})$, UO_2PO_4^- , etc.) are formed (Giammar 2001). Therefore ternary complexes adding anions such as carbonate or phosphate are also conceivable.

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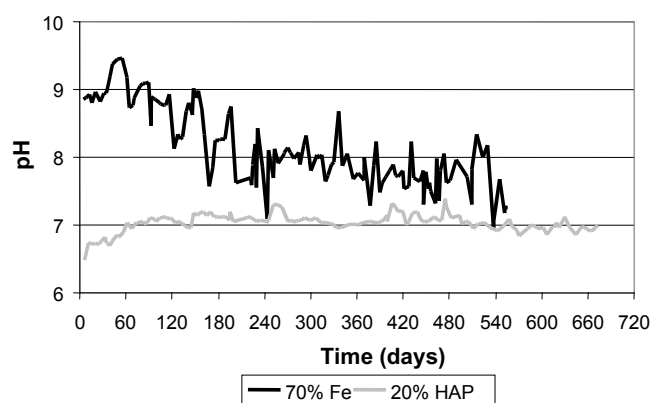


Figure 7

pH of the effluent from long-time column experiments.

Structures have been suggested for such ternary complexes for uranium on hematite surfaces and for uranium on HAP (Bargar et al. 2002).

Long-term stability of sorption complexes or mineral phases is essential for PRB systems using HAP for uranium removal. Evidence exists that under common groundwater conditions uranium remains immobile (Giammar 2001). Proper functioning of a PRB system depends on the adequate design. Lessons learned from the demonstration PRB at Fry Canyon show that site characterisation and groundwater flow patterns are the most important parameters. If the contaminant plume is not completely intercepted by the reactive medium removal efficiency is poor.

The cost of HAP as a reactive material is comparable to that of granular iron and is around 350 US\$ per ton (EnviroMetal Technologies Inc., Conca et al. 2000). Lower prices for HAP are feasible if waste materials, e.g. fishbone or phosphatic clay (Singh et al. 2001) containing HAP are used. A more detailed discussion on the economic aspects of PRB systems in comparison with active groundwater remediation technologies can be found elsewhere (Simon et al. 2002, Simon et al. 2005).

Acknowledgement

The research has been conducted within the European Project 'Perebar' (Project No. EVK1-CT-1999-00035, see www.perebar.bam.de).

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Hydromechanisation for Environmental Protection: from Dilute Hydraulic Transport to Paste Technology

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Abstract

The paper reviews the development of environmental protection research at Miskolc University, Hungary from the 1960s. Highlight of this activity has been hydromechanisation, i.e. handling and transporting solids mixed with water. Special cases of application justify implementation of this technology: economical benefits in long-distance transport, protecting the environment from pollution, dredging subaqueous deposits, lack of access for more bulky equipment, hygiene and safety reasons. Hydraulic transport started with the use of dilute mixtures to ensure high operational safety and was developed by implementing high-concentration mixtures to enhance economic advantages and topped by introducing thickened tailings and paste technology.

Extensive research has been carried out at the Faculty of Earth Science and Engineering of University of Miskolc aiming at improving various aspects of environmental protection. The beginnings go back to the 1960s and 1970s when major research projects were initiated by industrial demands and problems. The University of Miskolc was then called Technical University of Heavy Industry, and the faculty's name was Faculty of Mining Engineering. Therefore, mineral exploitation, mineral processing and energy production formed the backbone of research. Under uniquely favourably conditions it was possible to implement the results of theoretical considerations and laboratory measurements in laboratory-scale experimental rigs. In addition, full-scale experiments were also possible in a nearly real-scale experimental rig constructed on the premises of the University. The results gained from these experiments were implemented directly by industry.

Most research topics have their ascending period peak and descending period according to current demands, solutions of problems and practical implementation of the results. The 1970s and 1980s were the prime time of hydromechanisation and hydraulic transport. The term hydromechanisation comprises winning or excavating solid or granular materials, their wetting or mixing with water if required, their transport by pipes or pipelines, separation and processing after transport and then storage or disposal. As a major technological unit, hydraulic transport is often dealt with separately.

Both the ease of its structure and the possibility of automation encouraged the development of hydromechanisation and

hydraulic transport. These features provided major economic benefits which gave the main impetus to promoting these technologies. Included was, however, one more important aspect which was much ahead of its time, and this was its environmentally friendly character. Hydromechanisation helped protect the environment even before environmental protection became a major issue for society and industry.

Environmental protection

Environmental benefits of hydromechanisation and hydraulic transport are obvious due to a closed system. The environmental impact is minimum throughout the hydraulic transport system since there are no points or leaks where contamination can escape the system during processing and transport.

There are three major fields of application where hydraulic transport bears the potential of providing substantial benefits:

- in all cases when an industrial process is carried out using wet technology, i.e. solids are available in form of a slurry,
- long-distant transport when low transport costs outweigh the expenses of slurry production before and dewatering after transport plus potential impacts on the environment,
- in cases when the transport should be carried out in an almost "invisible" fashion (e.g. construction and operation in built-up cities).

Unlike pneumatic transport, no dust is generated at the site of disposal, not to mention pollution caused by belt conveyors.

A disadvantageous feature of hydraulic transport is that treatment of process water after transport has environmental implications.

Simple construction

Simple pipeline structure is a major advantage in areas where infrastructure or alternative transport methods such as road

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or railway are not available, or the existing infrastructure is overloaded. This is the case in remote areas (e.g. large forests), open pit mines or slag or waste heaps at mines or power stations whose life time is usually only a few years. Increasing the transport capacity of a mine shaft also bears a high application potential.

Automation

Engineers are always looking for methods of automation to reduce the amount of manpower in operation or production. This also applies to underground mining where production without human presence might revolutionise the process and reduce hazards. Advanced industrialised countries, first of the USA, Germany, the former Soviet Union and Japan, made major efforts to improve underground mining technologies, even experimental mines applying hydraulic technologies were built and operated.

Most of these aspects were implemented in the research and experiments of the University of Miskolc in the 1970s and 1980s. Three key topics should be highlighted here:

- theoretical and laboratory investigation of two-phase flow of solid-liquid mixtures,
- transport of bottom and fly ash from coal-burning power stations,
- vertical mine shaft transport and dredging of sand and gravel.

Theoretical and laboratory investigation of two-phase flow of solid-liquid mixtures

Characteristic types of motion of solid particles in a multi-phase heterogeneous mixture are illustrated in *Figure 1* (Wasp et al., 1977). In low-concentration transport, which was used to ensure high operational safety, floating-swimming transport is considered the most suitable flow regime from the fluid mechanics' point of view. Curve a in *Figure 1* indicates

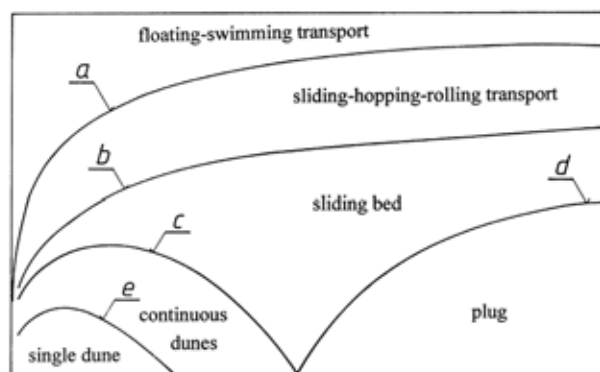


Figure 1
Regimes of multi-phase heterogeneous mixture flow (Wasp et al., 1977):
flow velocity vs. concentration

the minimum flow velocity required for this way of transport as a function of concentration. Coarse-grained materials require very high velocities for floating-swimming transport. If transport occurs at low velocities, the solid particles travel along the pipeline by sliding, hopping or rolling. The lower velocity limit of this transport regime is indicated by ensuing deposition (curve b). If the velocity is further reduced, sliding-bed transport occurs with another boundary curve (c and d). Velocities below this limit result in a stationary bed with dunes being formed (under curve c) or the pipeline clogged (curve d), depending on the concentration.

The process of transporting sand and gravel particles 1 to 6 mm in size in a 50 mm dia pipe is depicted in *Figures 2 a to d* (Meggyes, 1976). The concentration gradually increases from *Figure 2a* toward *Figure 2d*. Three flow regimes exist simultaneously: floating-swimming transport, sliding, hopping, rolling and sliding bed. With increasing concentration, a shift takes place from floating-swimming transport toward sliding bed.

To investigate the main parameters of hydraulic transport, several experimental rigs were constructed, one of them is shown in *Figure 3* (Debreczeni and Debreczeni 2001).

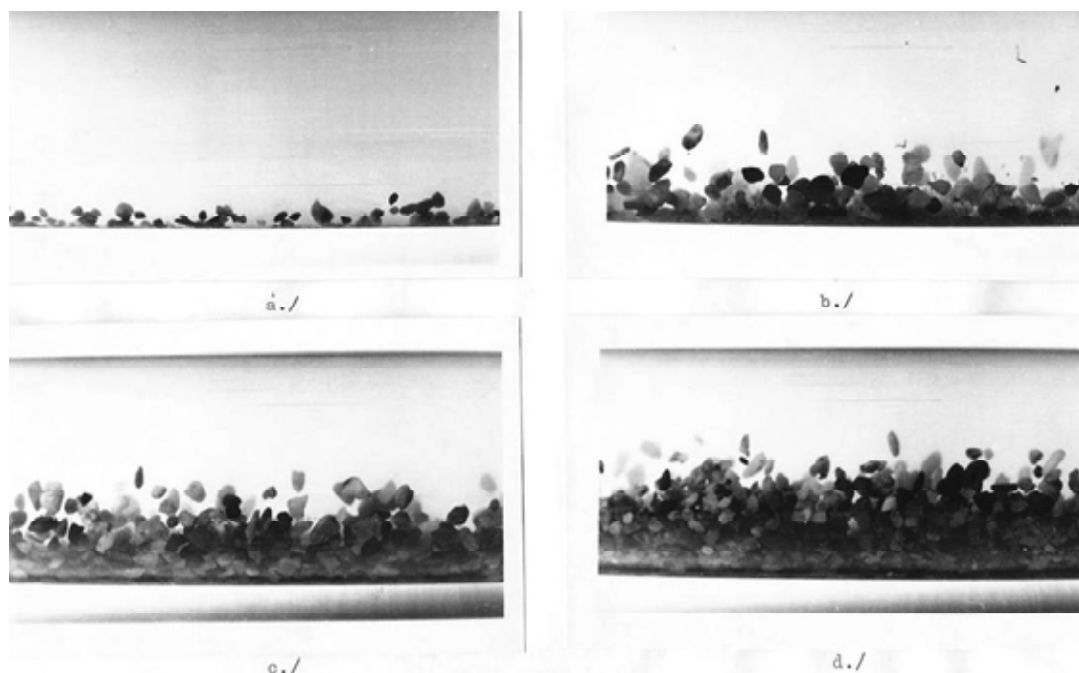


Figure 2
Flow regimes of 1 – 6 mm
sand in a 50 mm diameter
pipe

Usually, local and transport velocity, flow rate, concentration, pressure, frictional pressure loss, power requirement of the pump, solids density, particle size characteristics, viscosity, depositing velocity, temperature and, in some cases, layer thickness were measured (Debreczeni and Meggyes 1977; Debreczeni and Meggyes 1983). In long-term experiments cooling was often necessary to compensate for the temperature increase caused by friction. One of the most complex tasks was to measure the local concentration of two-phase mixtures. A method developed was based on weighing a section of the pipe connected flexibly by rubber hoses to the pipeline (Figure 4). One end of the section was hinged using bearings and the other end supported on a cantilever equipped with strain gauges (Figure 5). This measurement principle was also implemented in a large-scale industrial measurement device in a 350 mm fly-ash transporting pipeline of the lignite-burning Gagarin power station in Hungary.

Venturi meters were developed and used to measure flow rate, and diaphragm-type manometers equipped with strain gauges to measure pressure and pressure differential. A major industrial measurement task to analyse the operation of an 8 km long, 600 mm diameter fly-ash-transport system was carried out at the Tusimice Power Station, Czechoslovakia in 1980.

The final objective of the analysis of two-phase flow is to determine the frictional pressure loss along the pipeline, since this is the single most important parameter the power requirement of the entire system depends on. Kriegel and

Brauer (1966) developed a calculation method based on elementary physical phenomena and including a large number of parameters:

$$\Delta p_0 = \frac{\rho v^2}{2} \frac{L}{D} \left\{ \lambda + 0.282 c (s-1) \left(\frac{v_s'}{gv} \right)^{1/3} \right. \\ \left. \frac{(gD)^{4/3}}{v^{8/3}} \left[1 + 2.7 \left(\frac{c}{c_{\max}} \right)^4 \right] \right\}$$

where:

c	delivered volumetric concentration
c_{\max}	0.5 (50 %)
D	pipe diameter (internal)
g	acceleration due to gravity
L	pipe length
Δp	pressure differential
s	relative specific gravity and density (related to water)
v	mean velocity of flow
v_s'	terminal velocity of hindered settling
λ	pipe friction factor
ν	kinematic viscosity
ρ	density

Figure 3
Experimental rig for slurry transport investigation (Debreczeni, Debreczeni 2001)

- [1 - membrane-type separating device,
2 - pressure difference meter,
3 - layer thickness meter,
4 - measurement section (80 mm and 150 mm diameter),
5 - pump,
6 - ultrasound velocity meter,
7 - discharge velocity meter,
8 - thermometer,
9 - slurry container,
10 - cooling pipe,
11 - cooling water,
12 - safety pipeline]

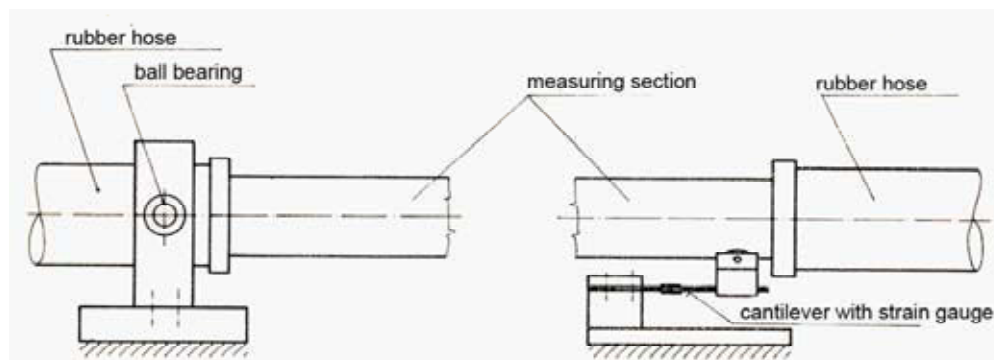
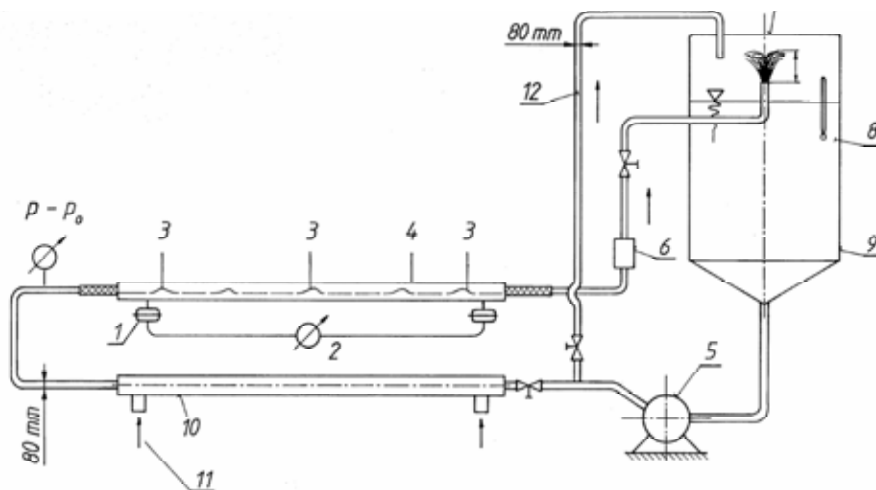


Figure 4
Concentration meter using the weighing principle

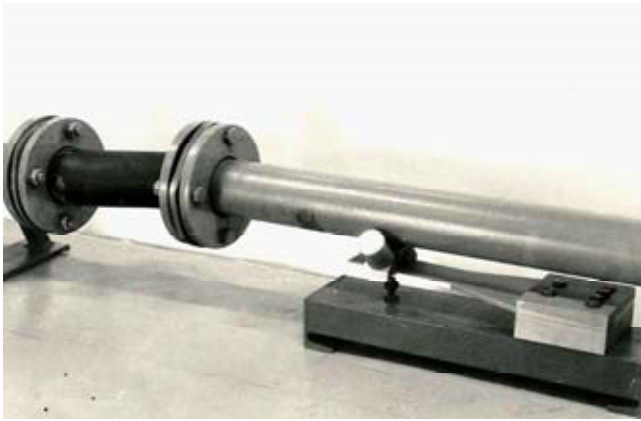


Figure 5
Concentration meter: cantilever and rubber hose of concentration meter

Based on numerous laboratory- and large-scale experiments on various transported materials, the method was further developed to comply with measurement results. An example of pressure loss calculation for 1.6 m t/a 0-10 mm coarse coal is shown in Figure 6 (Meggyes, 1976).

Hydraulic slag and fly ash transport

Currently, bottom and fly ash from coal-burning power stations is often transported hydraulically to the disposal site. In the 1970s, these residual materials were usually transported using pneumatic transport or belt conveyors. The University of Miskolc investigated extensively hydraulic fly-ash transport simultaneously to the introduction of this technology in all coal-burning power stations in Hungary. Electricity was mainly produced in coal-burning power stations in the 1970s. The nuclear power station at Paks, south of Budapest, was already being built, but the first block only went on line in 1982. This was the time when environmental consciousness emerged in the public and hydraulic transport of bottom and fly ash made a major contribution to protecting the environment by preventing pollution.

The question arises why was it necessary to carry out extensive experiments on this topic? Is it not possible to estimate

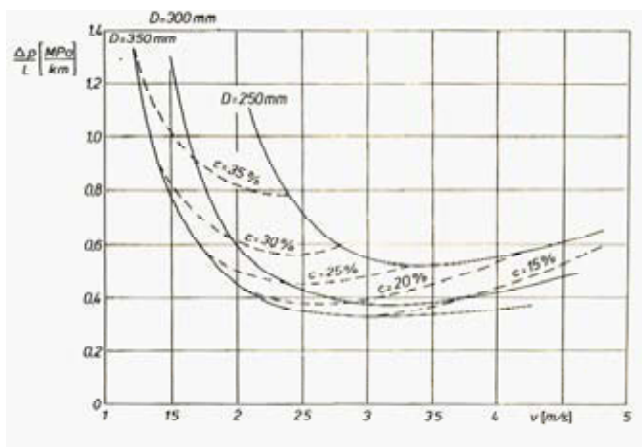


Figure 6
Pressure loss of 1.6 m t/a 0-10 mm coarse coal

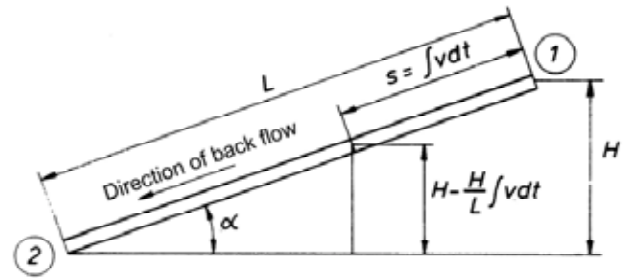


Figure 7
Fluidmechanical model of back flow in an inclined fly-ash-transporting pipeline

the results if test results on samples taken from one or two power stations are available? The problem is that the properties of ash produced in different power stations are completely different. Every fly ash/water mixture has its own, special rheological characteristics due to the mineral composition of the coal and the grain size distribution of the fly ash. The binding process of the water/fly ash mixture is a complicated chemical process similar to that of Portland-cements. There are fly ash types with no binding capability which fail to set even after long periods while certain fly ash types yield the strength of concrete on the site of disposal. It was found that reliable results could only be achieved by experimental investigation in each case.

A special problem arose in those fly-ash-transporting pipelines whose destination was at a higher elevation than its starting point. If the flow is interrupted due to power failure or breakdown, the ensuing back-flow (reflux) may cause water hammer by the sudden closure of the no-return valve behind the pump. Figure 7 illustrates the fluidmechanical model of the problem.

The problem translates to an integro-differential equation:

$$H - \frac{H}{L} \int v dt = \frac{L}{g} \frac{dv}{dt} + \lambda \frac{L}{d} \frac{v^2}{2g}$$

with

- H height difference between start and destination,
- L length of the pipeline,
- v velocity,
- g acceleration due to gravity
- t time,
- d internal pipe diameter.

The equation can be solved by introducing a new variable:

$$\frac{dv}{dt} = u(v)$$

The solution is obtained as

$$v = \frac{2d}{\lambda} \left(\frac{gH}{L} - u - \ln \frac{K + \frac{gH}{L}}{K + u} \right)^{1/2},$$

$$K = \frac{gdH}{\lambda L^2}$$

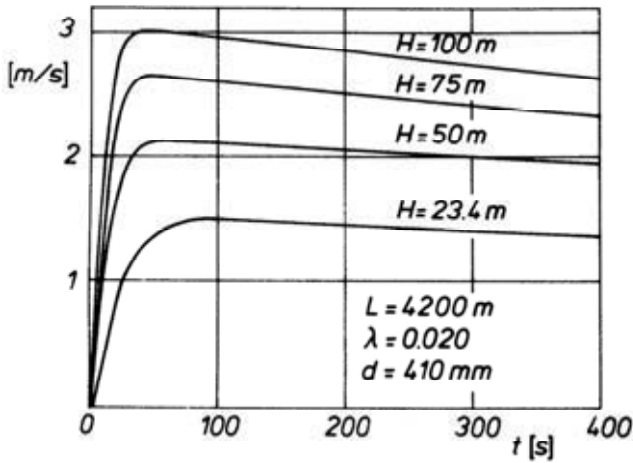


Figure 8
Back-flow velocity as a function of time at various inclinations in a fly-ash-transporting pipeline

and is shown in Figure 8. Velocity v is an indirect function of time t via u .

To avoid water hammer, it has to be made sure that the instance of the closure of the no-return valve does not coincide with the peak velocity (Meggyes, 1981).

Hydraulic shaft transport for the mining industry

Extensive investigations on vertical transport and jet devices provided excellent results both in terms of research and practical application. A potential application for vertical hydraulic transport using jet devices is the increase of shaft transport capacity. Due to small space requirement a vertical pipe can easily be constructed in an existing shaft, thus the expensive construction of a new shaft can be avoided. Savings on this item are so overwhelming that they compensate far more for the low efficiency of hydraulic transport equipment using a jet device.

In the various types of jet pumps, energy is transferred to the transported fluid by means of jets that enter the mixing chamber through a centric nozzle with a circular cross-section, a ring-shaped side channel or holes in the side wall of the pump (Debreczeni, Meggyes, 1999).

When solid-liquid or solid-gas mixtures containing large particles are to be transported, the most suitable type of jet pump is that with side nozzles. This type of jet pump has nozzles in the side wall of the mixing chamber designed and manufactured in accordance with fluid mechanics principles (Figure 9).

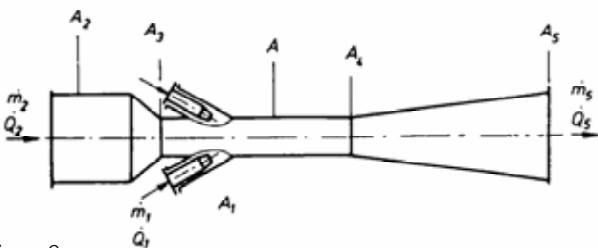


Figure 9
Side-nozzle jet pump

Using the denotations shown in Figure 9 we can introduce the dimensionless mass and volumetric flow rates

$$\psi_m = \frac{\dot{m}_2}{\dot{m}_1} \quad \text{and} \quad \psi = \frac{\dot{Q}_2}{\dot{Q}_1}$$

and the dimensionless head

$$\varepsilon = \frac{p_5 - p_2}{p_1 - p_2} + \frac{c_5^2 - c_2^2}{p_1 - p_2} \frac{\rho_{25}}{2} = \varepsilon_p + \varepsilon_c.$$

In this expression ρ_{25} denotes the characteristic density of the flowing medium between cross-sections 2 and 5. If the jet pump axis is horizontal, the numerator of ε_p gives the increase in pressure energy and the numerator of ε_c does the increase in kinetic energy of the transported fluid or mixture. The velocities in cross-sections 2 and 5 are generally almost equal thus $\varepsilon_c \approx 0$ and $\varepsilon = \varepsilon_p$.

The final versions of the dimensionless heads can be written:

$$\varepsilon_p = \frac{B_1 - B_2 \psi - B_3 \psi^2}{B_4 - B_5 \psi^2};$$

$$\varepsilon_c = \frac{B_1^* - B_2^* \psi - B_3^* \psi^2}{B_4 - B_5 \psi^2}$$

where

$$B_1 = 2 \frac{A}{A_1} \cos \alpha - \left(2 + \zeta \frac{\rho_{34}}{\rho_1} \right) + \eta_D \frac{\rho_{45}}{\rho_1} \left[1 - \left(\frac{A}{A_5} \right)^2 \right];$$

$$B_2 = 2 \left(1 + \frac{\rho_2}{\rho_1} + \zeta \frac{\rho_{34}}{\rho_1} \right) - 2 \eta_D \frac{\rho_{45}}{\rho_1} \left[1 - \left(\frac{A}{A_5} \right)^2 \right];$$

$$B_3 = \zeta \frac{\rho_{34}}{\rho_1} + \frac{1}{\varphi_2^2} \frac{\rho_{23}}{\rho_1} \left[1 - \left(\frac{A}{A_2} \right)^2 \right] - \eta_D \frac{\rho_{45}}{\rho_1} \left[1 - \left(\frac{A}{A_2} \right)^2 \right];$$

$$B_4 = \frac{1}{\varphi_1^2} \frac{\rho_{23}}{\rho_1} \left(\frac{A}{A_1} \right)^2;$$

$$B_5 = \frac{1}{\varphi_2^2} \frac{\rho_{23}}{\rho_1} \left[1 - \left(\frac{A}{A_1} \right)^2 \right];$$

$$B_1^* = \left(\frac{A}{A_5} \right)^2;$$

$$B_2^* = -2 \left(\frac{A}{A_5} \right)^2;$$

$$B_3^* = \left(\frac{A}{A_2} \right)^2 - \left(\frac{A}{A_5} \right)^2.$$

ρ_{23} , ρ_{34} and ρ_{45} in the above expressions denote the density of the fluid or mixture flowing in the confuser, mixing chamber and diffuser which are characteristic in the calculation of pressure losses. If the motive and transported fluids are the same, all three densities equal ρ_1 . If an air-solid mixture has to be transported with the aid of water, then a two or three phase mixture flows through the cross-sections in question. In this case the fluid mechanical parameters and density ratios have to be determined experimentally, especially for the diffuser, where a relatively large increase in pressure takes place.

A major implementation of the research results was the design and construction of a booster jet device for shaft sinking commissioned by Ruhrkohle AG, Germany. The task was to lift rock fragments cut by a large-diameter rotary shaft drilling equipment to the platform of the high-capacity shaft hoist equipment 25 m above the shaft bottom. It had to be proved that the jet device is capable of transporting 50-mm sized particles to this height. The equipment was constructed in the power plant of the University because it was the only building that was able to accommodate a 25-m high experimental rig. Experts of Ruhrkohle inspected the jet device in operation and were satisfied with the performance of this industrial-scale test equipment.

Sand and gravel dredging

The third important area was sand and gravel dredging in lakes. Mechanical excavators have a limited depth capacity and they leave considerable amounts of untouched resources behind. This is a loss in terms of resource exploitation and an environmental problem because other deposits have to be worked to compensate for non-excavated gravel resources, which spoils further areas of land. Jet devices are capable of working in greater depths thus reducing the loss of gravel deposits. High-performance hydraulic dredges using jet-cutting heads and jet pumps were developed, designed and constructed. The University of Miskolc developed several floating dredges which were operated in North-Western Hungary (Figure 10).

These dredges applied an overall wet technology for

- gravel excavation by jet cutting,
- vertical transport by jet pumps,



Figure 10
Floating dredge using jet technology for gravel excavation

- horizontal transport of the extracted material to the shore.

Smaller units were built to remove sludge from channels and lakes.

Hydraulic transport has not only advantageous features, there are also some disadvantages. The two most important of them are:

- transport water may contaminate the environment around the pipeline discharge, and
- the high energy consumption also has environmental implications.

High-concentration transport and paste technology

At the early stage of using hydraulic transport, feasibility was the main objective of technical development. Low-concentration hydraulic transport was considered the safest because of a low risk of clogging. However, environmental considerations require as small amounts of water as possible to reduce the extent of contamination to groundwater and surface water. Therefore research moved toward high-concentration hydraulic transport and, simultaneously, the major coal-burning power stations also started implementing high-concentration slag and fly ash transport. In addition to saving the environment, high-concentration hydraulic transport may have further benefits.

Certain fly ash/slag suspensions exhibit binding properties. The binding mechanisms are influenced both by mineralogical composition, viz. CaO , MgO , SiO_2 , Fe_2O_3 and Al_2O_3 contents and the fraction of fines (under 0.02-0.03 mm), i.e. particle size distribution. In the shaded area in Figure 11 (University of Miskolc, Dept. of Mining & Geotechnical Eng, 1999) good self-hardening capability can be expected due to the particle size distributions. The smaller the fraction smaller than 0.02-0.03 mm, i.e. the greater the distance between the particle size distribution of a particular fly ash and the shaded area, the smaller the expected self-hardening capability (Debreczeni and Debreczeni 2001).

Improvements of high-concentration transport resulted in the emergence of thickened tailings and paste technology. The Sustainable Improvement in Safety of Tailings Facilities

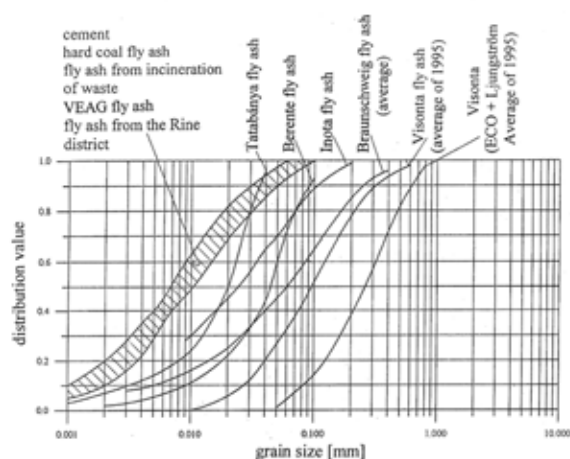


Figure 11
Self-hardening capability as a function of grain size distribution

(TAILSAFE) research project was carried out in 2002 to 2005 with the participation of scientists of BAM and the University of Miskolc in addition to other partners. This was a 5th Framework Programme Research Project supported by the European Union within EESD-2000-7.1 RTD Activities of a Generic Nature, Fight against major natural and technological hazards. The task of the University of Miskolc was to investigate how paste technology can contribute to increasing the safety of tailings facilities.

The environmental advantages of paste technology are twofold: the moisture content of the slurry is minimum, as shown by the typical underflow from an paste thickener (Figure 12).

Consequently no water leaves the tailings at the site of disposal and the risk of contaminating the water resources is minimum (Figure 13). Whether or not a material is suitable for paste production depends on the particle size distribution and the surface properties of the solid material and several other parameters.

The University of Miskolc carried out investigations on tailings/water suspensions from the Gyöngyösoroszi tailings



Figure 12
Typical underflow from an ultra-high density "paste" thickener (Bedell et al., 2002)



Figure 13
Paste disposal from an artificial tower. The gently sloping surface helps rain water run-off. (Newman, 2003)

pond within the TAILS SAFE project (Böhm et al. 2006). The material had a maximum particle size of 100 μm . The fraction smaller than 20 μm was about 75 % and below 2 μm 22 %. The slime content was about 52%. The plasticity index corresponded to the value of a lean clay (15-20), but it was close to silt values (10-15). Plasticity properties and particle size distribution data showed very similar results. The particle density was 2701 kg/m^3 . The flow properties of the tailings suspension showed a Newtonian character under 20 % concentration and Bingham-plastic properties at higher concentration. Pressure loss measurements for a near-paste state at 40 % are shown in Figure 14.

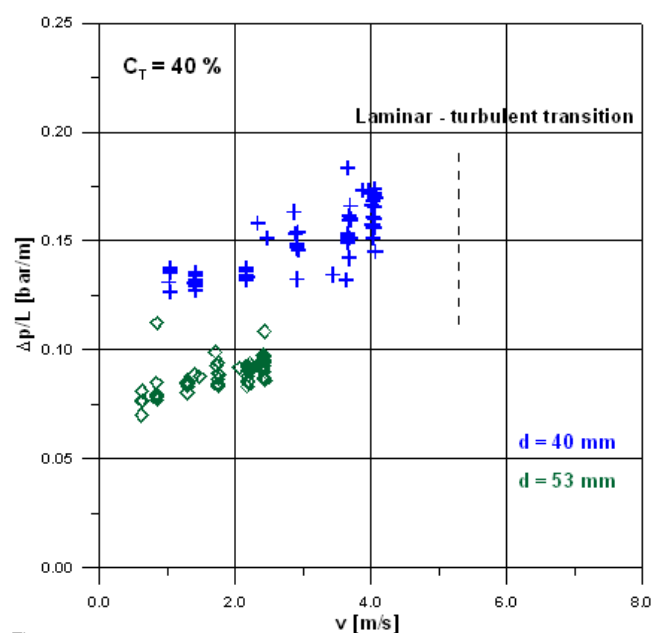


Figure 14
Measured pressure loss curve at 40 % transport volumetric concentration of a Gyöngyösoroszi tailings/water suspension (Böhm et al., 2006)

Conclusions

Hydraulic transport and hydromechanisation have provided significant technological improvements and economical benefits during the last few decades. Starting with dilute transport to ensure high operational safety, it was further developed toward the high-concentration method, which finally resulted in paste technology. Development in this field

provides a good example of the increasing role of environmental considerations in addition to technical and economical aspects. It indicates that thoughtful design helps save the delicate balance between technology and nature.

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Underground disposal of mining tailings by hydraulic conveyance A proven, sustainable alternative for the potash industry

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1 Background

The European Union is a major producer, consumer and exporter of potash (potassium chloride), more than 90 % of which is used in fertiliser production. Other uses include specialist chemical manufacture, glass making and pharmaceuticals. Potash production currently employs around 10,000 people in five EU countries (France, Germany, Italy, Spain and UK).

Potash production involves the separation of the saleable product from largely inert waste materials. These waste materials typically comprise a coarse soluble salt component (mostly sodium chloride with some other salts) and fine-grained insoluble clay. The clay fraction of the waste may contain associated trace metals. Traditionally, these wastes have been disposed of either by discharge into rivers and

coastal waters or by storage in large surface disposal facilities. In recent years, the industry has come under increasing pressure to develop alternative options for waste disposal to reduce the environmental impacts associated with traditional disposal options.

Cleveland Potash Limited (CPL) currently supplies some 55 % of the UK's potash consumption from one of Europe's largest potash mines with a depth of 1100 m located at Boulby in the county of Cleveland. Boulby is situated on the north-east coast of England within the North Yorkshire Moors National Park, an area noted for its scenic beauty and within which leisure activities and tourism play key roles in the local economy.

3 Mt ore are mined annually in the Boulby mine and 2 Mt of process waste are produced each year. 1.8 Mt is a soluble salt residue with 200,000 t of insoluble clay residue (filter cake) containing trace heavy metals (cadmium and mercury), approximately 50 kg/year.

Given the sensitivity of the location, surface waste disposal at Boulby (such as in conventional slimes dams, landfill, disposal to local quarries or old ironstone workings) is not a viable option and the mine currently disposes of both the coarse-grained soluble waste and the fine-grained clay waste directly into the North Sea. This disposal route has come under increasing scrutiny in recent years for two reasons:

1. The potential environmental impacts arising from the discharge of fine-grained clay material onto the seabed.
2. The presence of trace metals (principally cadmium and mercury) associated with the fine-grained clays. Although these trace metals are tightly bound within the clay lattice (and therefore not mobile within the marine environment), the UK government is committed to reducing discharges of cadmium and mercury to the North Sea in accordance with international commitments (the OSPAR Commission).

The future sustainability of the Boulby Mine is dependent on the identification of an alternative, cost-effective, waste disposal option that will significantly reduce existing environmental impacts. The original marine discharge limit set by the UK Environment Agency was 140,000 tpa. Due to increased

production, a temporary consent of 200,000 tpa of insoluble clays was granted provided that CPL implemented alternative disposal options. Accordingly, CPL undertook innovative research and development work to investigate the potential for disposal of the waste materials in the underground voids that exist in the many worked-out areas of the mine.

The practice of returning some or all of the waste produced underground ("backfilling") has been adopted at many mining operations. However, at Boulby this presented a major challenge as a combination of the physical characteristics of potash wastes (high water content, small particle size, low strength and high consolidation times) and the particular nature of the underground environment (subject to considerable post-mining deformation, very low humidity, high temperature and the long transport distances involved) had constrained the application at Boulby of the backfill technology that been adopted elsewhere in the international mining industry.

Research and development work commissioned by CPL in 1996 and 1997, however, showed that these constraints could be overcome. Accordingly, an application was made to the LIFE Environment Programme in December 1997 for financial support in order to design, build and operate a waste handling facility to demonstrate the feasibility of underground disposal of potash wastes. The project proposal was based upon three principal phases of activity:

1. A programme of laboratory analysis and testwork, to develop a specification for the mixture of the two waste products with cementitious additives to form a single waste product with defined and consistent physical properties, suitable for underground disposal.
2. The design, construction and operation of a small-scale pilot plant to develop final design criteria and operating procedures for a full-scale demonstration plant.
3. The design, construction and operation of the full-scale demonstration plant, comprising a waste mixing plant at surface and pipework for the transport of the combined waste material up to 11 km underground for final disposal in the worked-out areas of the mine.

2 The backfill project (1998 – 2003)

Laboratory analysis and testwork demonstrated that the addition of large quantities of cementitious binders, although technically feasible in certain cases, was essentially uneconomic and unnecessary.

A waste mixture without any additives that fulfilled all the technical criteria required for waste backfill, and upon which the design of the pilot plant could be based was defined. This conclusion had the implication of simplifying the conceptual design requirements for the waste mixing plant and enhancing the economic viability of the backfill option. A mixture comprising a 3.3:1 ratio of coarse soluble salt to insoluble clay with a controlled quantity of cementitious binding agent, mixed with sea water achieved the minimum necessary density of 1600 kg/m³ to meet system requirements and restrict the bleed water volume to less than 6 %.

The backfill material did not have the geotechnical properties necessary for use as roof support, and it would not be used, therefore, to permit the mining of adjacent pillars of potash.

The critical technical issues that were identified as requiring priority attention in subsequent tasks were all related to the need to ensure adequate quality controls during waste mixing to produce a consistent waste mixture of the required specification for disposal underground. *Table 1* lists the backfill system requirements.

Pilot Plant design, construction and testwork proceeded as originally envisaged based on the outcome of the laboratory testwork. The pilot plant confirmed the feasibility of transporting the waste mixture by gravity (i.e. without the need for pumps) to disposal points underground. The transport of

Table 1
Backfill system requirements (see text)

Backfill materials	Filter cake
Carrying fluid	Sea water
Backfill tonnage	200,000 t/yr
Backfill density	Maximum practicable (1,400 to 1,600 kg/m ³)
Operational days	330 days/yr
Placement	3 times per day
Maximum placement distance	11,040 m
Maximum gravity head available	1,100 m
Flexibility	Divert to sea
Density of carrier fluid	1026 kg/m ³
Sea water viscosity	1.221×10^{-3} Pa s

a waste mixture comprising solely fine-grained clay waste, mixed with seawater to an appropriate consistency, proved to be suitable for transport and placement underground.

The pilot testwork established that addition of coarse soluble salt waste or rheology modifying additives indicated by the laboratory testwork did not improve the mixture characteristics (in fact, the addition of coarse salt caused some problems with blockages in the pipeline). The optimum waste mixture was one with a density of 1.56 kg/l comprising 60.48 % fine-grained clay waste and 39.52 % sea water. The ability to place waste with a higher than anticipated proportion of fine-grained clay was considered to be a significant benefit. This would enable greater use of underground voids for disposal of the insoluble waste associated with the potential environmental impacts, without the need to incorporate benign salt waste in the mixture. It was also found that the system could be successfully restarted after stoppage without a risk of blockage or settling within the pipework.

It was clear that the priority for the design and operation of the full-scale demonstration plant would focus on the parts of the system on the surface, particularly those relating to quality controlling the consistency of the waste mixture, as this would ensure that the physical transport and disposal of material would not be unduly problematic.

The Demonstration Plant Design aimed to maximise the backfill density to minimise the volume of supernatant water introduced to the mine. Suitable underground panels for backfill placement were identified and a schedule for disposal operations was developed.

The key component of the backfill system is the mixing tanks. Maintenance of the correct viscosity of the waste mixture is fundamental to the successful operation of the system; the required viscosity itself varies according to the distance the

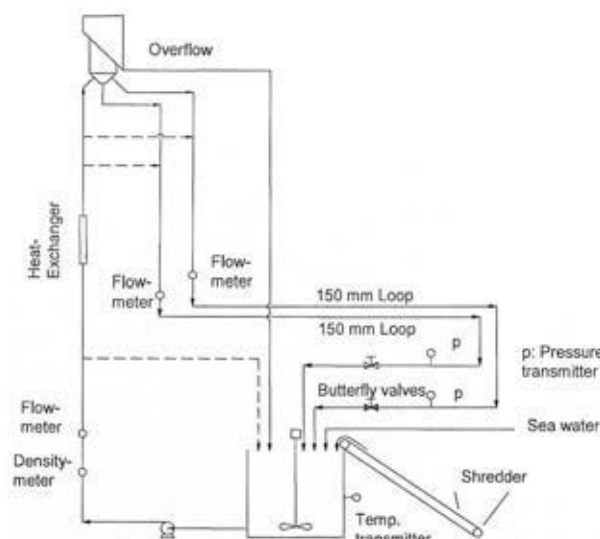


Figure 1
Sketch and photo of the pilot plant.

material has to be transported for disposal – the closer the disposal site is to the shaft the higher the viscosity. A quality control loop in each backfill batch tank ensured the slurry rheology varied so that backfill could be delivered by the gravity head to the placement panels underground at various distances from the shaft column, whilst minimising the risk of pipe blockage.

The potential impact on the underground air quality and the risks to roof support pillars were found not to be significant. Figure 1 displays the pilot plant.

The complete demonstration plant was fully operational and started on 19th May 2003. A 9-month Demonstration Test Programme confirmed the original assumptions underlying the project as well as the detailed design criteria developed during the project and the plant design and operating parameters were shown to be robust.

3 Environmental Benefits and Applications

3.1 Compliance

Two waste products are produced in the production of a saleable product at Boulby mine: a coarse soluble salt (mostly sodium chloride) and fine insoluble clay. Prior to the commissioning of the backfill plant, both waste materials were discharged into the North Sea under consent from the UK Environment Agency. Consent limits on the quantity of insoluble

clay discharged to the North Sea have varied since the mine was first commissioned. However, in 1999 a consent was agreed under which discharges would be progressively reduced from a peak of 200,000 tonnes per annum to 130,000 tonnes per annum by November 2004.

The reduced figure is considered by the Environment Agency to be below the 140,000 tonnes per annum capacity of the discharge area to assimilate clay material – in years when the tonnage has increased above this value the impacted area has been shown to increase, whilst at lower annual tonnage discharged natural dispersal mechanisms associated with winter storms mitigate the impact.

Two key environmental criteria were cited to justify reducing the quantity of insoluble clay discharged to the North Sea :

1. The presence of elevated concentrations of trace metals in the clay. Although these trace metals, which include cadmium and mercury, are naturally occurring and are confined within the clay lattice – and hence are not chemically reactive or bioavailable – regulatory pressure to reduce discharges of these elements has remained.
2. The impact of the clay discharges on the benthic flora and fauna that inhabit the sand onto which the discharge is made. Although this flora and fauna is naturally poor (due to the shifting nature of the sand and high turbidity) – which is why the location was first identified as being suitable for a discharge – regulatory pressure to reduce discharges of insoluble clay has remained.

The moves to reduce the discharge to sea coincided with a predicted increase in the requirement to dispose of insoluble clay waste at the mine arising from a predicted increase in the proportion of insoluble clay within the mined ore. This has necessitated a review of waste disposal options at Boulby. The ability to place waste material underground offers an opportunity to address environmental concerns whilst supporting the viability of potash production at Boulby for many years to come.

3.2 Waste Disposal at Boulby Mine

The backfill system will provide the capability to dispose of 155,000 m³ per annum of slurry containing the insoluble waste material underground. At current rates of production, this represents 35 % of the total quantity of insoluble waste produced.

A review of the available void space underground suitable for waste disposal has identified the potential for the disposal of 1,550,000 m³ of slurry. At current rates of production, this represents 10 years of operation. CPL anticipates that additional void space beyond this capacity could be identified to extend the operational life of the backfill system beyond 10 years.

In future, CPL proposes to manage the disposal of insoluble clay waste using a combination of the existing sea discharge at much reduced quantities together with underground backfill.

This combination of waste disposal options provides for the optimum operational life of the mine and retains flexibility to continue disposal operations during periods of system maintenance, for example, whilst meeting CPL's commitments to reduce waste discharges to sea.

Insoluble salt wastes, comprising principally sodium chloride, will continue to be discharged to sea with no perceived environmental impact.

3.3 Environmental Monitoring

The site of the existing sea discharge has been subject to intense monitoring since the 1970s and the data collected are reviewed by a committee of stakeholders comprising CPL, the Environment Agency, the National Park Authority, English Nature and fisheries interests. The discharge area is recognised as one of the most intensively studied of any in Europe.

This monitoring will continue and as the quantity of insoluble clay waste discharged is reduced any improvements will be identified and reported through the monitoring committee. Initial indications based upon the findings of previous studies are that there will be a fairly rapid response to reductions in solid burdens with an increase in numbers and diversity of peripheral benthic communities.

In addition, although the mercury and cadmium content in the current discharge has no discernible environmental impact (because the metals remain bound within the clay lattice), reducing the quantity of these metals discharged to the North Sea is consistent with the UK Government's stated objectives and is in accordance with commitments made to the OSPAR Commission.

3.4 Transferability

All potash mines in Europe are based upon similar oceanic deposits and production generates wastes of similar types to that at Boulby. Traditional methods for the disposal of the insoluble fraction of this waste material have included:

1. Surface disposal into tailings dams
2. Discharge into marine or riverine environments

The disposal option selected at individual mines has been based upon site specific criteria. For example, French potash mines in Alsace discharged insoluble residues (and dissolved salts) to the River Rhine until the mines closed during the 1990s. In contrast, potash mines in Sicily favoured disposal into surface tailings impoundments.

Both these options can be associated with significant environmental impact. The French potash mines were required to progressively reduce discharges into the Rhine whilst concerns over groundwater contamination forced the closure of operations in Sicily in 1991.

Although the option of backfilling waste materials into underground voids has been considered previously, the plant now operating at Boulby was the first of its kind to demonstrate the technical and economic viability of backfill as a potash mine waste disposal option. The extent to which this technology might be adopted at other operations in Europe (and worldwide) is unclear. The selected option for waste disposal will continue to be decided on site-specific criteria. Backfill technology may not always be the preferred option. However, the European industry now has three proven options to consider.

4 Summary and Conclusions

Cleveland Potash Ltd instigated a research and development project, with the support from the EU LIFE-Environment Programme, to determine the feasibility of using potash tailings as backfill. Extensive initial test work established the rheological characteristics of a range of waste slurry combinations and assessed their potential for quick solidification by binder addition. It was found that whilst there were binders available that could solidify the waste slurries, the quantities of binders required would have been unfeasible on purely economic grounds.

Pilot test work successfully demonstrated that the insoluble tailings slurry could be hydraulically conveyed, using the head available owing to the depth of the workings below surface, via a shaft column and horizontal pipe haulage to a designated disposal area underground. The results of the pilot

plant test work were used to establish the backfill rheology and formed the basis for the design of the full-scale plant.

The full-scale plant was designed to maximise the backfill density and hence minimise the volume of supernatant water introduced into the mine. As the distance to each backfill placement panel varies and only the gravity head provided by the shaft column is available to deliver the slurry, the slurry rheology has to be varied to ensure that slurry at the maximum practicable density is delivered to each panel. To achieve this a quality control loop was incorporated in each backfill batch tank to ensure that a consistent slurry viscosity was achieved.

The operation of the quality control loop has proved to be very successful in controlling the properties of the batched slurry.

Acknowledgements

This paper is based largely on work and reports completed by the staff of Cleveland Potash Limited, the Mineral Industry Research Organisation (MIRO), Nottingham University, *Pater-son* and *Cooke* and others during a four year demonstration project funded by the EU Life Environment Programme (LIFE ENV/UK/000603: Reduction of Waste Discharges by Under-ground Disposal of Process Residue), in particular those

by *Matt Wilkins*, *Mark Dodds-Smith* and *Abbie Richards*. This paper contributes to the dissemination of information, which was primarily the responsibility of MIRO during and subsequent to the project. It was presented at the valedictory workshop held to mark the retirement of Dr *Tamás Meggyes*, at BAM, Berlin on 24 February 2006.

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Increasing the Safety of Tailings Facilities

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Tailings Management Facilities

Tailings are fine-grained wastes of the mining industry and can be defined as ore from which as much as feasible of the desired minerals have been removed (BAT, 2003). They usually contain residual minerals, process water and often also process chemicals. Due to the mixing with water during the mineral processing, e.g. milling and flotation, they are output as slurries with sometimes high water contents. Deposits of these residues in ponds or lagoons, usually confined by dams, can present a serious threat, especially when there is improper handling and management. Failure of mine tailings facilities may result in uncontrolled spills of tailings, dangerous flow-slides, and/or the release of poisonous chemicals, leading to major technological disasters. In Europe, recent accidents such as the Aznalcóllar (Spain) and the Baia Mare (Romania) disasters raised public awareness for the risks related to such facilities:

- Los Frailes Mine near Aznalcóllar, Andalusia, Spain: 15,000,000 m³ of tailings from the processing of sulfidic Zn, Pb, and Cu ores were deposited in a 27-m high tailings impoundment. On 25th April 1998, breaching of the dam occurred due to ground failure, releasing 1,500,000 m³ of tailings and 5,500,000 m³ of acidic water to the Agrio and Guadiamar Rivers. The flow of tailings reached 40 km downstream, contaminating the Donana National Park. Although discussed controversially, foundation failure is widely assumed as main reason for the dam break, due to unsuitable subsoil containing swelling clays (see also discussion of the accident on <http://www.wise-uranium.org/mdaff.html>).
- Aurul Gold Mine, Baia Mare, Romania: 800,000 m³ of cyanide-containing tailings were deposited in a tailings impoundment with a dam height of a couple of meters. On 30th January 2000, overtopping of the dam occurred due to heavy rains, leading to the erosion of the dam crest and the release of 100,000 m³ of cyanide-contaminated liquid (containing up to 150 mg/l free cyanide) to the Lapus River. Subsequently the cyanides travelled downstream severely contaminating the Tisza and Danube Rivers systems. As main reason for the disaster poor management of the freeboard and the ponded water are cited. Further

information is given in the UNEP (2000) and EU (2000) reports.

Tailings dams are usually constructed using part of the material coming from the milling process (Vick, 1990). This is achieved by using hydrocyclones at the end of the pipelines transporting the slurry from the mill. The coarser particles (sandy fraction) are used for dam construction, while the fines are deposited in the pond. A high percentage of fines in the tailings results in long settling periods, sometimes several years or even decades. In handling large amounts of inhomogeneous wet slurries, water management is a key safety factor. Deficient water management has proven to be one of the main causes of accidents and hazards emanating from tailings facilities.

The most popular and widely used construction method for tailings dams is the 'upstream' method (Figure 1 and Figure 2), providing only a thin outer surface of coarse mate-

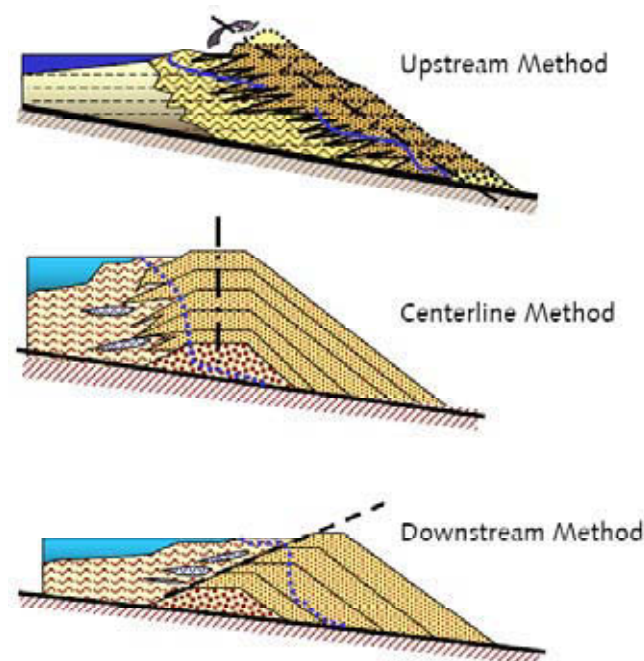


Figure 1
Methods of tailings dams construction (taken from Witt & Schönhardt, 2004).



Figure 2
Example for tailings dam construction using the upstream method (photo: Roehl).

rial as a dam (Vick, 1990). Although the method has obvious advantages in terms of storage capacity and land use, it needs to be carefully managed to maintain its internal and external stability. Critical factor governing the dam stability is the water management, with beach width and location of the phreatic surface being the most critical parameters. The 'centreline' and 'downstream' methods (Figure 1) yield better stability but require larger land areas and larger amounts of solids for the construction of the dam. One significant feature of 'downstream' dams is that they allow a full water cover on the tailings pond's surface enabling the storage of tailings with very high water contents.

To prevent or reduce the percolation of contaminated seepage to the subsoil and/or the dispersion of dust from the tailings surface to the surrounding landscape, the application of base and capping liners is possible, similar to systems used for landfills. Recommendations for landfill basal liner and capping systems, as given by the German landfill regulation, are shown in Figure 3. In the case of existing tailings facilities, considerations will be mainly related to the implementation of capping systems. A wide range of options are available ranging from simple soil covers to more advanced solutions, e.g. based on geosynthetics. For new tailings facilities, basal sealings using composite systems consisting of compacted clay liners and geomembranes (HDPE) are a viable option and have already been implemented at a number of sites.

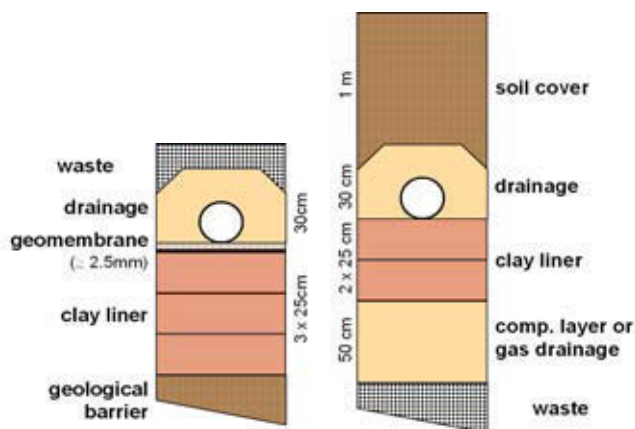


Figure 3
German landfill regulation for basal liner (left) and capping (right) systems (after TA Siedlungsabfall, 1993).

Water management is considered the most critical parameter of tailings facility safety. Inflow/outflow water quantities need to be balanced to avoid excess water. Inflow is mainly generated by process water which is used to hydraulically transport the tailings to their disposal place, precipitation, and surface run-off. Outflow includes evaporation from the tailings surface, removal of ponded water by decantation, often combined with reclamation and reuse of the decanted water in the ore processing scheme, and seepage of pore water through the dam or into the subsoil. Leachate from tailings bodies is often contaminated with chemical compounds originating from the mined ores (e.g., heavy metals, radionuclides) or from the ore milling and treatment process (e.g., flotation additives, flocculants etc.).

The TAILS SAFE Project

TAILSAFE (Sustainable Improvement in Safety of Tailings Facilities), a European research and technological development project, was funded under the European Commission's 5th Framework RTD Programme, and was carried out from December 2002 to November 2005 (Roehl *et al.*, 2003). The general objective of TAILS SAFE was to develop and apply methods of parameter evaluation and measurement for the assessment and improvement of the safety of tailings facilities, paying particular attention to the stability of tailings dams and slurries, the special risks inherent when such materials include toxic or hazardous wastes, and authorization and management procedures for tailings facilities. Aspects of tailings facilities design, water management and slurry transport, non-destructive and minimally intrusive testing methods, monitoring and the application of sensors, intervention options and remediation strategies have been considered in TAILS SAFE. A risk-reduction framework – the "Tailsafe Resource" – has been established to contribute to the avoidance of catastrophic accidents and hazards from tailings facilities.

Tailings from the mining and primary processing of metals, minerals and coal are in principle included within the scope of TAILS SAFE. Due to the limitations of any such research project, not all mineral tailings can be addressed in the same detail, but a representative range is intended. Industrial minerals, such as aggregates and clays, are one important group of raw materials whose tailings have not been covered so fully by this project. However, much in the project may be relevant to such tailings too, and the Tailsafe Resource, a key output, was designed so that its use can be readily extended to a wider range of tailings. Although TAILS SAFE primarily addressed European needs and priorities, and was based strongly on European case studies, the project has built on world-wide experience and scientific results, and much of its output has world-wide applicability. Further information on the project is available at its website, <http://www.tailSAFE.com/>.

The TAILS SAFE team consisted of ten partners from six European countries: Department of Applied Geology, Karlsruhe University, Germany (coordinator); Federal Institute for Materials Research and Testing, Department of Environmental Compatibility of Materials (BAM), Berlin, Germany; Department of Foundation Engineering, Bauhaus University of Weimar, Germany; Department of Mining and Mineral Engineering

and School of Civil Engineering, University of Leeds, United Kingdom; Minerals Industry Research Organisation (MIRO), Leeds, United Kingdom; Finnish Environment Institute, Helsinki, Finland; Department of Mining and Metallurgical Engineering, National Technical University of Athens, Greece; Department of Mining and Geotechnical Engineering and Department of Process Engineering, University of Miskolc, Hungary; Mecsek-Öko Zrt., Pécs, Hungary; and Research and Design Institute for Rare and Radioactive Metals, Bucharest, Romania.

Results of the TAILS SAFE Project

The approach chosen by the project team to contribute to the safety of tailings management facilities was to focus on certain innovative, relevant and tractable parameter evaluation and measurement work, but within a systematic framework which shows how that focus relates practically to wider and generally better understood parameters. This dual approach on focus and framework is clearly reflected in the achievements of the project, which feature a selection of reports and publications on specific aspects of tailings management and a comprehensive web-based parameter framework realised as an online risk reduction tool.

An initial review and definition phase included the review of mineral processing and slurry handling technologies, tailings facilities design and authorisation, management, monitoring and inspection practices (Meggyes *et al.*, in prep.), and the review of reliability factors for tailings facilities, as expressed by impact, risk, instability and failure mechanism parameters. Based on the initial review work, options for risk reduction at dangerous or impaired tailings facilities were considered. The applicability of intervention actions was assessed, including actions related to the tailings ponds contents, such as evacuation, dewatering and chemical stabilisation, and actions related to the strengthening of tailings dams.

Acknowledging the fact that the reduction of risks associated with each phase of a tailings facility's life – construction, operation, closure – should be taken into account in the planning phase, the design and authorisation procedures for proposed tailings facilities were studied. Issues exam-

ined included preliminary site investigation, characteristics of tailings materials, tailings facility design, and the role of external experts.

The safety of tailings facilities can be considerably increased using high concentrate transport and deposition of the particulate solid material. Paste technology produces and handles much denser slurries in the transport into tailings facilities than conventional technology. Advantage of this method is that smaller dams are sufficient. The load on the embankment is also smaller which means less risk to failure. It is also important that there is no excess water that has to be treated and the material cannot be liquefied. In case of a failure it spreads only within a limited area causing less damage than conventional tailings material. The optimisation of tailings water management was addressed by experimental research into further developing paste technology, which allows the transport and disposal of dewatered tailings. Experiments on hydraulic paste transport and paste thickening were conducted in specially-developed pilot-scale test systems.

Closure issues were covered in a survey addressing cover systems, stabilisation measures and control technologies, supplemented by a case study on reclamation measures for uranium tailing ponds. To address the case of substandard or impaired tailings facilities, the options and procedures for actually implementing risk reduction measures were studied and evaluated. Looking beyond these more technological matters, the field of tailings-related legislation and regulation was addressed, including all aspects of legislation on country, EU and global levels, and tailings facilities authorisation, management, monitoring, and inspection.

Approaches and methods to assess the state of tailings bodies and dams, with special attention to their internal and external stability and contaminant inventory, were studied in laboratory and field experiments. This work focused on the development of investigation and monitoring techniques and their practical testing under real-life conditions at case study sites. To cover a wide spectrum of different types of tailings facilities, six case study sites in Finland, Germany, Hungary, Romania and UK were selected. The characterisation of the case study sites included the analysis of mineralogical and geotechnical parameters of tailings material. In a study on the

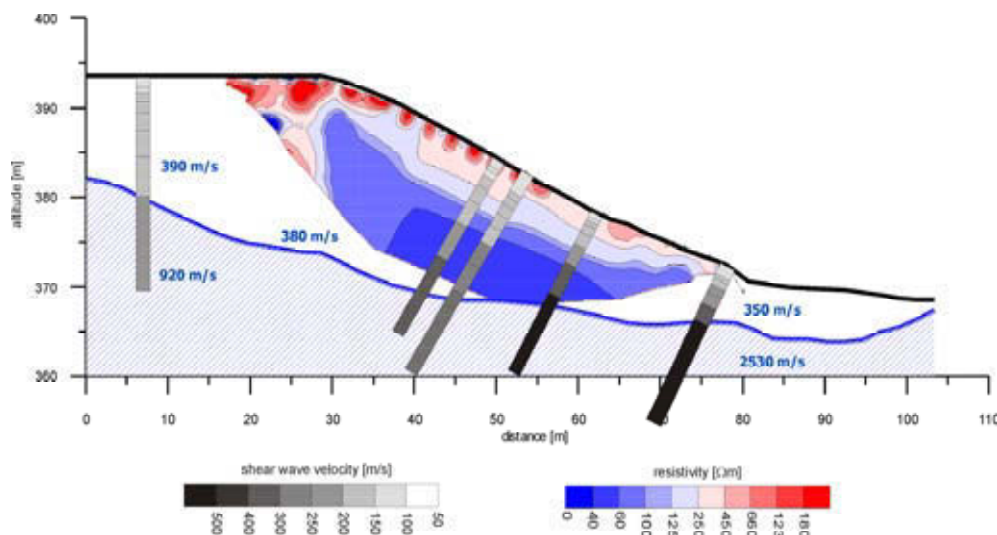


Figure 4
Example for geoelectrical and seismic investigations at a tailings facility. The measured resistivity clearly shows a dry dam surface, but also indicates potential seepage at the dam toe (TAILSAFE Final Report, 2006).

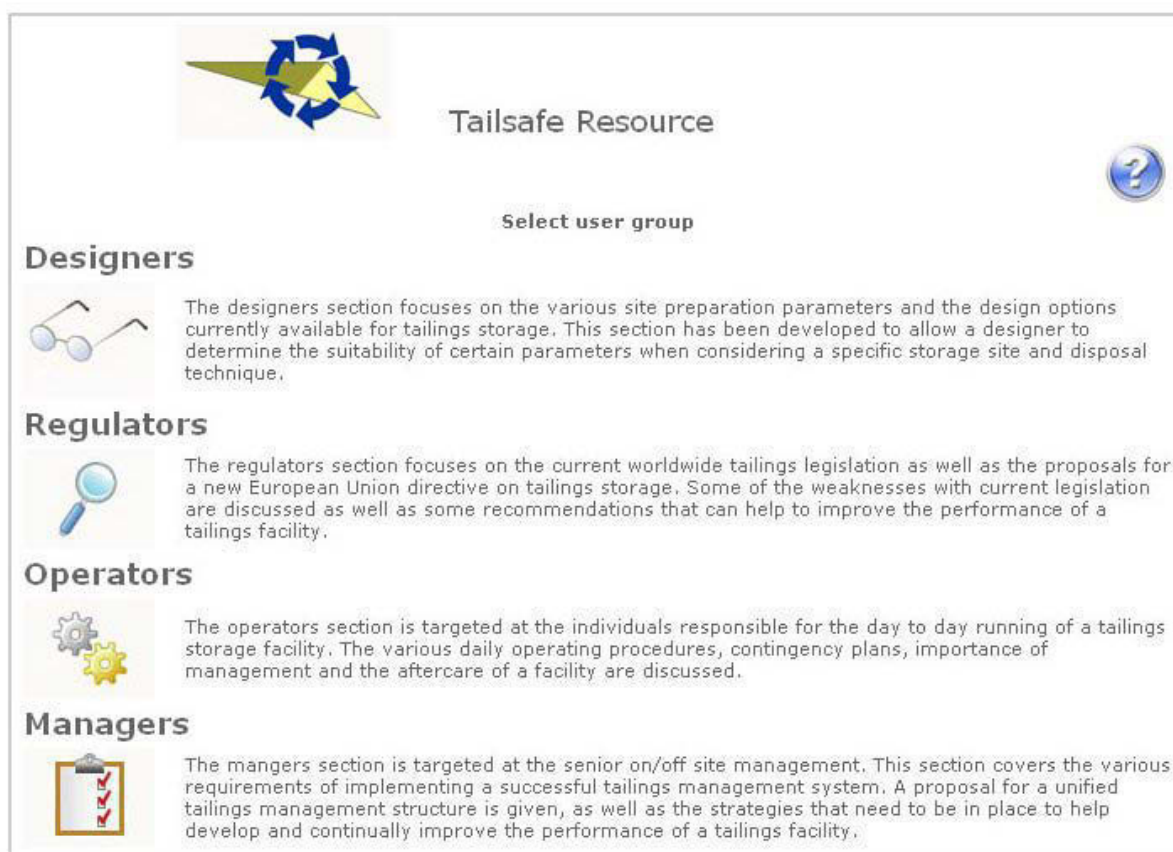


Figure 5
Starter-screen of the Tailsafe Resource highlighting the four major disciplines, their logos and a brief overview of their descriptions (<http://www.tailsafe.net/>).

flow-deformation behaviour of tailings material the geomechanical behaviour of tailings bodies was also investigated.

The applicability of non-destructive methods for the investigation of tailings dams and bodies was studied and evaluated in laboratory and field tests. The methods tested included geophysical techniques such as geoelectrical methods, ground penetration radar and seismics (Figure 4). Other techniques such as the neutron probe were also tested. Of these methods, geoelectrical techniques and the neutron probe proved to provide useful information on the internal structure of tailings dams and bodies. A number of field test campaigns were conducted on the case study sites. A major objective of all the work conducted on the case study sites was to develop tools suitable for the detection and description of the safety status of tailings facilities. This effort was supported by the development of models to simulate the structure of tailings impoundments. Existing and improved methods dealing with uncertain and random data for the calculation of the structural risk of tailings impoundments were studied, and software applications enabling the creation of stochastic soil profiles on the basis of geo-statistical assumptions were developed.

Finally, a web-based risk reduction framework, the "Tailsafe Resource", has been formulated, assembled, and made available at <http://www.tailsafe.net/>. The Tailsafe Resource provides an online database of common and specific parameters that can increase a user's awareness of surface tailings storage operations and safety. The Resource is structured so as to lead different types of users (Figure 5) logically through evidence and intervention options relating to their roles and

problems. It contains a diagnostic tool linked to from the various sections within the Resource. Industrial case study examples are included, and the site is fully illustrated by numerous photographs and diagrams. The Tailsafe Resource can be considered one of the major outcomes of the project, with a high potential for further development and practical application by a wide range of stakeholders.

The project's achievements are available as reports, books and software applications. Most of the deliverables will be available through the project website at <http://www.tailsafe.com/>, where further information on the project and its outcomes is being provided.

Conclusions and Recommendations

A significant number of studies and investigations have been performed within the framework of the TAILSFAE project, culminating in the development of a systematic risk reduction framework (the TAILSFAE Resource). From the technical work and experiences gained within the framework of the project, scientific and technical conclusions were drawn and recommendations for a successful and safe management of tailings were formulated. Some selected key conclusions are listed below:

- Water management and monitoring, pore water pressures and seepage are the most important safety-related issues.

- High concentration tailings (thickened tailings) are a promising technique for increasing the stability/safety of tailings bodies.
- Geophysical non-destructive investigation techniques (especially geoelectrical) are suitable to supply valuable information on the internal structure of tailings dams and bodies and their surroundings, on areas of high water content and seepage, and on the spread of inorganic contaminations.
- Human actions and behaviour play a crucial role in the construction and operation of tailings facilities.
- Training and qualification of all personnel and authorities involved is of utmost importance.
- The Tailsafe Resource is an online parameters database that can increase a user's awareness of surface tailings storage operations and related risks. It can also be used as an Internet training tool.

Some selected key recommendations for a successful and safe management of tailings facilities are as follows:

- The planning and designing of new tailings facilities should include a detailed site investigation by qualified geoscientists to prove suitability of the potential disposal site.
- The dryer the better: the dewatering of tailings tend to increase their stability. Paste and thickened tailings technologies should be developed further to enable their cost-effective use in the mining industry, leading to dryer and therefore more stable tailings impoundments.
- In the framework of the implementation of the new EU mining waste directive the new national regulations should be adapted to local conditions.
- Training risk reduction - capacity building and education to raise the qualification level of all involved (operators, regulators).
- The Tailsafe Resource should be developed further to a professional, fully functional, multilevel and interdisciplinary risk reduction tool.
- Discuss the failures: one can learn from these far more than from successes.
- Only interdisciplinary joint work of mining engineers and geoscientists guarantees success and safety of tailings management facilities.

A more comprehensive list of conclusions and recommendations can be found in the final report of the TAILS SAFE project which is available for free download at <http://www.tailsafe.com/>.

Acknowledgements

The TAILS SAFE project was supported by the European Commission within the Thematic Programme "Energy, Environment and Sustainable Development" of the 5th Framework Programme for Research and Technological Development (project name: Sustainable Improvement in Safety of Tailings Facilities – TAILS SAFE; contract no: EVG1-CT-2002-00066).

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Effect of voltage gradient on integrated electrochemical remediation of contaminant mixtures

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Abstract

This paper evaluates the effect of voltage gradient on the efficiency of integrated electrochemical remediation (IECR) of low permeability soils contaminated with both heavy metals and polycyclic aromatic hydrocarbons (PAHs). The IECR remediation process aims to oxidize organic contaminants within the soil by electroosmotic delivery of Fenton's reagent consisting of hydrogen peroxide (H_2O_2), utilizing native soil iron catalyst as well as simultaneously removing heavy metals by electroosmotic advection and electromigration. A series of bench-scale experiments was performed on kaolin (a low permeability soil) spiked with nickel (a representative heavy metal) and phenanthrene (a representative PAH) each at a concentration of 500 mg per kg of dry soil using two voltage gradients, 1 and 2 VDC/cm. The H_2O_2 solution in two different concentrations at 5 % and 10 % was introduced at the anode, and each test was conducted for a total duration of four weeks. The results showed that the application of 2 VDC/cm did not induce significant electroosmotic flow as compared to the application of 1 VDC/cm. Phenanthrene removal from the soil was insignificant in all tests; however, 28 % and 34 % of phenanthrene was oxidized within the soil in the 5 % and 10 % H_2O_2 tests, respectively, using 1 VDC/cm. The phenanthrene oxidation increased to about 32 % and 42 % under similar H_2O_2 concentration conditions using 2 VDC/cm. Nickel migrated towards the cathode and then precipitated close to the cathode due to high pH conditions in all the tests. Nickel migration was slightly higher in case of 2 VDC/cm than in case of 1 VDC/cm due to comparatively greater acidic pH front migration towards the cathode under 2 VDC/cm. Overall, the results showed that an increase in the voltage gradient from 1 VDC/cm to 2 VDC/cm does not improve overall remedial performance significantly; therefore, strategies to increase H_2O_2 delivery and/or to prevent precipitation of metals near the cathode are warranted.

Keywords

Chemical oxidation, electrokinetic remediation, contaminant mixtures, soil, clays, heavy metals, organic contaminants

Introduction

The presence of mixed contaminants in soils poses a great challenge for their remediation. The mixed contaminants include combinations of organic compounds, heavy metals, and radionuclides. Very few conventional techniques are found to address the remediation of such mixed contaminant

sites (Sharma and Reddy, 2004). Moreover, these techniques are ineffective or expensive to remediate mixed contaminated sites that contain low permeability and heterogeneous soils. The synergistic effects of one type of contaminant on the remediation of another type of contaminant may complicate the treatment of such mixed contaminated sites.

Electrokinetic remediation has shown to have a great potential to remediate organic contaminants and heavy metals when they exist individually as well as to deliver the reactive agents in substantial quantities in different subsurface formations (Acar *et al.*, 1995; Reddy and Saichek, 2004; Reddy *et al.*, 2003a,b; Reddy and Chinthamreddy, 2003). Due to its effectiveness in low permeability and heterogeneous soils, where other conventional remedial methods have been proven to be ineffective or expensive, electrokinetic remediation has been receiving increased attention for in-situ remediation of contaminated sites. Recently, electrokinetic remediation has been investigated to remove both heavy metals and organic compounds from low permeability soils (Maturi, 2004). To enhance solubilization of contaminants, flushing solutions such as surfactants, cosolvents, chelates, and organic acids, were used, and the contaminant-laden solutions collected at the electrodes required further treatment. Overall, the process can be effective, but the costs associated with it may be high. Fenton-like chemical oxidation that uses hydrogen peroxide (H_2O_2) and iron catalyst is a simple process of oxidizing organic contaminants within the soil into relatively benign products such as carbon dioxide, water and oxygen. Therefore, aiming to oxidize organic contaminants within the soil and simultaneously aiming to remove heavy metals has potential to be an effective and less expensive remedial strategy.

Previously, the combined electrokinetics and Fenton-like oxidation of polycyclic aromatic hydrocarbons (PAHs) in low permeability soils has been shown successful (Chandhuri, 2003). Recently, the combined electrokinetics and Fenton-like oxidation was investigated to remediate clayey soils contaminated with both PAHs and heavy metals (Karri, 2005). In these studies, batch experiments and bench-scale electrokinetic experiments with different concentrations of H_2O_2 were conducted to determine the remedial efficiency of H_2O_2 on PAHs alone and when PAHs co-exist with heavy metals. Kaolin soil was used as model clay soil, and phenanthrene and nickel were used as representative PAH and heavy metal, respectively. These research studies have concluded that as the concentration of H_2O_2 increased, the oxidation of phenanthrene within the soil also increased. However, high residual phenanthrene concentrations were still observed throughout the soil after electrokinetic testing implying that the complete oxidation of phenanthrene was not achieved. There was a gradual increasing trend of nickel concentrations from the anode to the cathode in all the tests, which

implies that nickel migrated towards the cathode in all the tests irrespective of the concentration of H_2O_2 used. However, the removal of nickel from the soil was low, which could be due to precipitation as nickel hydroxides due to high pH near the cathode. In these tests, electroosmotic flow could not be sustained for a greater duration of time. The cumulative flow was less than two pore volumes in all the tests. Due to low electroosmotic flow, sufficient amount of H_2O_2 may not have been delivered into the soil for complete phenanthrene oxidation. Therefore, to achieve sustained electroosmotic flow during the electrokinetic testing process, implementation of strategies to enhance electroosmotic flow were suggested.

According to the Helmholtz-Smoluchowski theory, electroosmotic flow is directly proportional to applied electric potential (Reddy and Saichek, 2004). When the voltage gradient increases, the electric current can increase due to increase in ionic strength in the electrode solutions and more ions entering into the soil from the respective electrodes. The increased current can result in increased or sustained electroosmotic flow during the electrokinetic process. In this study, a series of bench-scale experiments was conducted to investigate the effects of voltage gradient on electroosmotic flow and delivery of H_2O_2 , oxidation of phenanthrene within the soil, and removal of nickel from the soil. In addition, this study investigated if the naturally available iron in the soil would act as a catalyst in the oxidation reaction.

Experimental methodology

Table 1 summarizes the variables involved in the experimental program. The experimental program allowed to systematically investigating the effect of voltage gradient on the Fenton-like oxidation of phenanthrene within the soil and the removal of nickel from the soil. Replicate experiments were conducted to ensure the accuracy and repeatability of the test results.

Materials

For all the experiments, commercially available laboratory grade kaolin soil, obtained from VWR Scientific Products and

Table 2
Properties of Kaolin

Property	Test Method	Value
Mineralogy		Kaolinite: 100% Muscovite: trace Illite: trace
Specific Gravity	ASTM D 854	2.6
Grain Size Distribution	ASTM D 422	% gravel = 0 % sand = 4 % silt = 18 % clay = 78
Hydraulic Conductivity (at porosity = 46%)	ASTM D 2434	1.0×10^{-8} cm/sec
pH	ASTM D 2974	4.9
Organic Content	ASTM D 2974	Near 0
USCS Classification	ASTM D 2488	CL

manufactured by EM Science, was used. Kaolin was selected as a model clayey soil due to its low hydraulic conductivity. It consists of mostly fine particles (up to 96 %) and near zero organic matter. The composition and properties of kaolin are presented in Table 2.

Phenanthrene was selected as a representative PAH. It is a neutral organic compound with three aromatic rings. It is hydrophobic in nature with very low aqueous solubility of 1.1 mg/L at 25 °C. Nickel was selected as a representative heavy metal due to its occurrence at majority of contaminated sites. For the testing purposes, nickel chloride salt ($NiCl_2 \cdot 6H_2O$) was used as source chemical for nickel. Both contaminants were selected at 500 mg/kg of dry soil each, which represent the typical concentrations at the contaminated sites.

In order to investigate the effect of Fenton-like chemical oxidation, H_2O_2 was used as a flushing solution at anode in all the tests. Native iron in the kaolin soil was utilized as catalyst in Fenton's reagent. Diluted stock solutions in 5 % and 10 % concentrations were prepared from 30 % H_2O_2 solution using deionized water.

Contaminated Soil Preparation

For each experiment, about 300 g of dry soil was taken in a glass beaker, and the predetermined amount of phenanthrene required to achieve the target concentration was dissolved in hexane in a separate glass beaker. The phenanthrene-hexane mixture was then mixed with the soil. The solution was allowed to mix within the soil homogeneously by constant stirring with stainless steel spoons. Additional amounts of hexane were added to allow thorough mixing of the soil. The mixing was carried out in a fume hood and care was taken to reduce the volatilization of hexane with phenanthrene. Once the mixing was completed, the soil was kept in the same fume hood for about three days to dry. After the soil was completely dry, it was transferred into a plastic pan and

Table 1
Experimental Program

Test*	Voltage Gradient (VDC/cm)	Time Duration (Weeks)	Flushing Solution
1	1.0	4	5 % H_2O_2
1R	1.0	4	5 % H_2O_2
2	1.0	4	10 % H_2O_2
2R	1.0	4	10 % H_2O_2
3	2.0	4	5 % H_2O_2
3R	2.0	4	5 % H_2O_2
4	2.0	4	10 % H_2O_2
4R	2.0	4	10 % H_2O_2

* Tests 1R, 2R, 3R and 4R are the replicate tests

mixed with nickel chloride solution to yield nickel concentration of 500 mg/kg. For this, nickel chloride salt was mixed in known volume of water that results in soil moisture content of 35 % by weight and the resulting solution was mixed with the soil. After mixing continuously for homogeneity, a representative sample from the soil was taken for analyzing moisture content, pH, and the actual initial concentration of phenanthrene and nickel in the soil.

Electrokinetic Experiments

Figure 1 shows a schematic of the bench-scale electrokinetic test set-up used for this study. A detailed description of the test set-up is given in Reddy *et al.* (2001) and Reddy and Saichek (2004). The spiked soil was uniformly compacted in the electrokinetic cell to achieve final dry density of 1.24 to 1.37 g/cm³. Once the soil was packed in the cell, filter papers were attached to either ends of the soil and then the cell was closed by electrode assemblies that consisted of porous stone and slotted graphite electrode on either ends. The anode reservoir was connected to the anode assembly, and a sample collection bottle was placed at the cathode outlet. Initially, the anode reservoir and assembly were filled with hydrogen peroxide solution and the cathode assembly was filled with deionized water. A constant voltage gradient of 1 or 2 VDC/cm was applied across the soil sample. Electric current and the electroosmotic flow were measured at regular time intervals. Periodically, outflow samples were collected at the cathode.

At the end of the each test, aqueous solutions from anode and cathode reservoirs and the electrode assemblies were collected separately and the volumes were measured. The soil specimen was extruded from the electrokinetic cell and sliced into five equal sections. The section close to cathode was then sliced into two equal subsections to better define the pH and distribution of contaminants near the cathode. Each soil section was stored in a glass jar and a representative sample was taken for final moisture content determination. About 10 grams of soil was taken from each sample

for the determination of pH, redox potential and electrical conductivity of the soil at the end of the testing period. The moisture content and pH of the samples were determined by using the standard methods ASTM D2216 and ASTM D4972, respectively.

Chemical Analysis

Phenanthrene Analysis

To determine the phenanthrene distribution in the soil at the end of the testing, Soxhlet extractions were first performed on each soil section according to the USEPA test method 3540C (USEPA, 1986; Reddy and Saichek, 2004; Maturi, 2004). For this extraction, about 10 grams of dry soil was taken in a Whatman cellulose extraction thimble and mixed with 10 grams of anhydrous sodium sulfate. About 95 mL of acetylene and 95 mL of methylene chloride were taken in a round-bottomed flask. The thimbles were placed in the Soxhlet extraction tube and these tubes were connected to the round-bottomed flasks with solvents. The Soxhlet extraction process was operated for at least 24 hours. At the end of the extraction, the final volume of extractant was measured. The same extraction procedure was performed on the initial spiked soil to determine initial phenanthrene concentration in the soil prior to treatment.

For analyzing the amount of phenanthrene in the Soxhlet extractants, a gas chromatograph (GC) of Hewlett Packard Model HP6890 Series GC system was used. The GC was equipped with a flame ionization detector (FID) and a column HP-5 (cross linked 5 % PH ME Siloxane) with dimensions 30 m x 0.32 mm x 0.25 µm film thickness. The injection of samples was carried out manually. One µL sample was injected for each run. Helium was used for make-up flow at a constant pressure of 25 mL/min. Initial oven set point temperature was 100 °C and ramped at 15 °C per minute for 10 minutes up to 250 °C with a total run time of each sample equal to 11 minutes. The GC was calibrated by using 2-fluorobiphenyl as an external standard. Standard phenanthrene

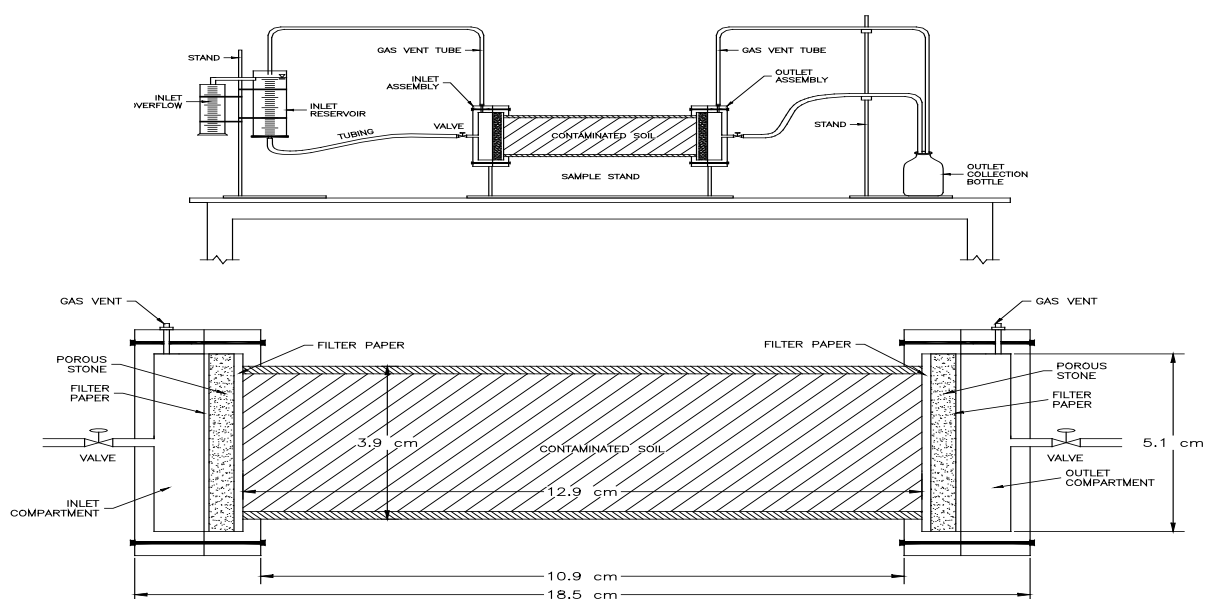


Figure 1
Bench-scale electrokinetic testing set-up

solutions were prepared in different concentrations ranging from 1 to 40 mg/L. Based on the surrogate concentrations, the extraction efficiencies were calculated.

The liquid samples collected at the electrodes were analyzed for phenanthrene concentration after performing liquid-liquid extraction (Maturi, 2004). For this, one mL of the liquid sample was taken in a test tube and it was diluted with water to 1:10 ratio. Then 200 μ L of 2-fluorobiphenyl was added to the test tube. Later, 2 mL of methylene chloride was added to the mixture and the test tube was then hand-shaken for about 5 minutes. The aqueous and methylene chloride phases were allowed to separate and then about 1-2 mL of separated organic phase of the solution was taken into an auto-sampler vial using a syringe. The extractants were later analyzed using gas chromatography. The extractants were analyzed for phenanthrene using the GC as previously described.

Nickel and Iron Analysis

Acid digestion of the initial spiked soil and all soil sections was performed according to the USEPA 3050 procedure (USEPA, 1986). Then the concentrations of nickel and iron were measured using atomic absorption spectrophotometer (USEPA, 1986). Iron was analyzed to assess availability of natural iron in the soil to function as catalyst in Fenton-like oxidation of phenanthrene. For acid digestion, about 2 g of soil sample was taken in a conical flask. Next 10 mL of 1:1 nitric acid (HNO_3) was added to it, and the soil and acid solution were mixed thoroughly. Then the soil-acid solution was covered with a watch glass, heated to about 95 $^\circ\text{C}$, and then refluxed for about 15 minutes. The sample was then allowed to cool and 5 mL of concentrated HNO_3 was added to it. The solution was then refluxed for 30 minutes. After cooling, the last step was repeated one more time. Covering the conical flask with a ribbed glass, the sample was allowed to reflux until it evaporated to about 5 mL. After cooling the sample, about 2 mL of deionized water and 3 mL of 30 % hydrogen peroxide (H_2O_2) were added. It was heated again to observe the effervescence. As the effervescence subsides, the sample was allowed to cool and another 1 mL of H_2O_2 was added again and the mixture was heated. This addition of H_2O_2 was continued for another 8 times, so that the total H_2O_2 added was less than 10 mL. Once the sample was allowed to cool, 5 mL of concentrated hydrochloric acid (HCl) was added with 10 mL of deionized water. The mixture was then heated for 15 minutes and finally cooled. Later, it was centrifuged at about 4000 rpm for about 30 minutes so that both the solid and liquid phases get separated. The supernatant was diluted to 100 mL, which then was used to determine the concentrations of nickel and iron using atomic absorption spectrophotometer (AAS). Liquid samples collected at anode and cathodes were also analyzed for nickel and iron using AAS. A calibration curve was prepared based on a range of standard concentrations. Samples with high concentrations were diluted whenever concentrations exceeded the highest standard concentration used in the calibration.

Quality Control/Quality Assurance

In order to ensure accuracy and reproducibility of the results, all the experiments were conducted in duplicates. Uniform testing conditions were maintained in all the tests by properly

sterilizing the electrokinetic cells, electrode assemblies and electrode reservoirs. During the analysis of iron and nickel using the AAS and phenanthrene using the GC, care was taken to ensure the samples were checked for accuracy by regularly checking the instruments for standard concentrations. Also, the mass balance analyses were performed for nickel and phenanthrene in the baseline tests using deionized water (Karri, 2005). The mass balance for nickel was confirmed in all the experiments.

Results and Discussion

The measured initial properties of the soil are summarized in Table 3. Also, the initial properties of the flushing solutions, along with deionized water are presented in Table 4. The results of the electrokinetic experiments were analyzed to assess the electric current and electroosmotic flow during the electric potential application as well as the soil pH, redox potential, electrical conductivity, and contaminant removal and/or oxidation in the soil at the end of testing.

Table 3
Initial Properties of Contaminated Soil

Parameter	Value
Dry Bulk Density (g/cm^3)	1.24-1.37
pH	5.80
Electrical Conductivity ($\mu\text{S}/\text{cm}$)	1840
Redox Potential (mV)	246.9
Moisture Content (%)	34.62
Nickel Concentration (mg/kg)	482
Phenanthrene Concentration (mg/kg)	447
Iron Concentration (mg/kg)	805

Table 4
Initial Properties of Flushing Solutions

Flushing Solution	pH	Electrical Conductivity ($\mu\text{S}/\text{cm}$)	Redox Potential (mV)
Deionized water	7.87	6.80	122.2
5% H_2O_2	6.32	17.42	218.2
10% H_2O_2	6.48	20.56	209.5

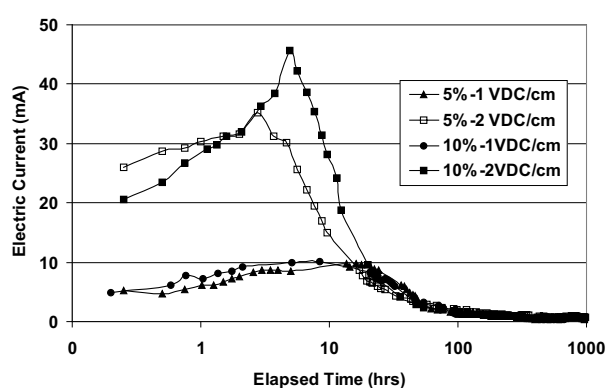


Figure 2
Electric current during testing

Figure 2 shows the variation of electric current at different intervals of time throughout the testing period. Electric current increased rapidly and then decreased rapidly during the initial stage of testing within first 24 hours. With 1 VDC/cm, the current increased to 10 mA, then dropped rapidly to about 3 mA and finally stabilized at 0.3 mA. Whereas with 2 VDC/cm, the current increased to 44 mA, then dropped rapidly to about 2 mA and finally stabilized at 0.3 mA. The high initial currents in case of 2 VDC/cm tests can be attributed to the higher initial mobility of ions in the soil. These initial high currents are due to the presence of salt precipitates that go into the solution (Mitchell, 1993). In addition, due to electrolysis reactions, H^+ ions are generated at the anode and OH^- ions are generated at the cathode. These ions being constantly generated possess high mobility into the soil. Due to the inflow of acidic solution, pH of the soil will be lowered, which may lead to additional mineral dissolution and a higher current. The ions move constantly into soil due to electromigration and electroosmosis processes. Over time, these ions are depleted as they move into the electrode chambers causing the current values to drop as time progresses (Eykholt, 1992; Pamucku, 1994; Grundl and Michalski, 1996). Although the initial current values are higher for 2 VDC/cm tests than in 1 VDC/cm tests, after a period of about 48 hours, all the tests showed similar values of electric current. As soon as ionic movement progressed into the soil in case of 2 VDC/cm tests, the depletion of ions also occurred at a similar rate. Therefore after a certain time, the ionic strength of pore solution in the tests with 2 VDC/cm became equivalent to that in the tests with 1 VDC/cm.

Figure 3 shows the variation of electroosmotic flow during the testing process. Flow increased rapidly in case of 2 VDC/cm during the initial 24 hours. As much as 30 mL was collected in the first 24 hours of testing and later on as the current value decreased, the flow rate also dropped simultaneously. The flow rate remained almost constant later during the remainder of the testing period. During the initial stages, flow increased at a uniform rate in case of 1 VDC/cm but not as high as that observed in the test with 2 VDC/cm. The flow rate continued to increase during the entire test duration in the tests with 1 VDC/cm. The cumulative electroosmotic flow in case of 1 VDC/cm was about 107 mL whereas in case of 2 VDC/cm it was about 91 mL. This shows that the flow in 1 VDC/cm was comparable to that in 2 VDC/cm test. In the 1VDC/cm tests, approximately 1.4-1.6 pore volumes of electroosmotic flow was obtained near the cathode in the 5 % H_2O_2 tests,

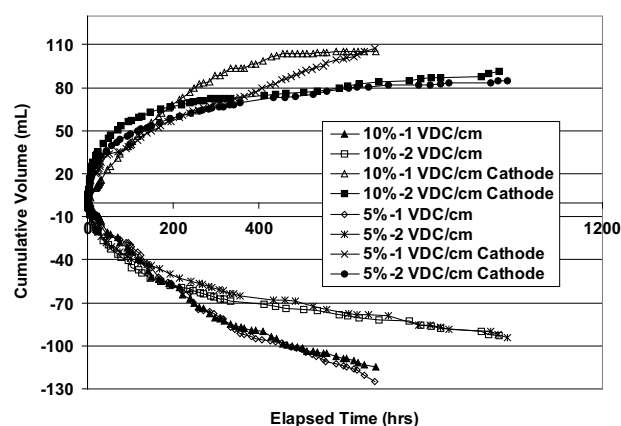


Figure 3
Electroosmotic flow during testing

and about 1.3-1.5 pore volumes were obtained in 10 % H_2O_2 tests. In the 2 VDC/cm tests, approximately 1.2 pore volumes were obtained in 5 % H_2O_2 tests, and about 1.3 pore volumes were obtained in 10% H_2O_2 tests. The high flow rate during the beginning stages was due to high initial currents. The flow rate decreases as ionic movement subsides, which is shown from the lower current values at a later stage of testing. In 2 VDC/cm tests, the flow rate decreased gradually due to the depletion of migrating ions. Also due to the precipitation of nickel ions at the cathode, the movement of ions could be reduced.

The difference in electroosmotic flow conditions between 1 VDC/cm and 2 VDC/cm tests was not significant. Therefore, it appears that increasing the voltage gradient may not necessarily increase the electroosmotic flow. The reason is that the higher voltage gradient may increase the electrolysis reaction at anode and consequently increase the H^+ ions generation. Thereby the soil pH near anode would be reduced which causes the charge on the soil mineral surface to be more positive, which may counteract the electroosmotic flow and/or the number of ions in the solution (Reddy and Saichek, 2004). Therefore, the low pH conditions can reduce the electroosmotic flow with time. Thus, there is no appreciable increase in cumulative flow towards the end of the testing in case of 2 VDC/cm tests. Another significantly adverse impact on electroosmotic flow could be the precipitation of nickel ions as nickel hydroxide near cathode. Consequently, ionic strength decreases which was shown from the lower current values at a later stage of testing (Figure 2).

As shown in Figure 4, the soil pH increased from anode to cathode in all the tests. In the first five sections from anode, the soil pH was reduced to values in the range of 2-4 in both 1 and 2 VDC/cm tests. In the sixth section that is near the cathode, high pH values about 10.5 were observed in 1 VDC/cm test. In case of 2 VDC/cm test, pH values increased to about 7.5. Comparatively low pH values were observed in case of 2 VDC/cm test than the 1 VDC/cm test. Due to electrolysis reactions, the acidic solution generated by H^+ ions at the anode migrates into the soil towards cathode thereby reducing the pH of soil. The alkaline solution generated by OH^- ions at the cathode migrates towards the anode increasing the soil pH near the cathode. The extent of transport of H^+ ions under 2 VDC/cm was greater than that observed under 1 VDC/cm resulting in lower pH through most of the soil. The observed non-uniform pH changes significantly affect the surface charge of soil particles (hence the electroosmotic flow) and solubilization and adsorption/precipitation of heavy metals.

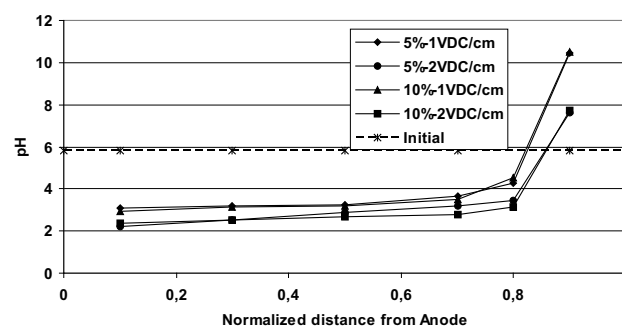


Figure 4
Soil pH distribution after testing

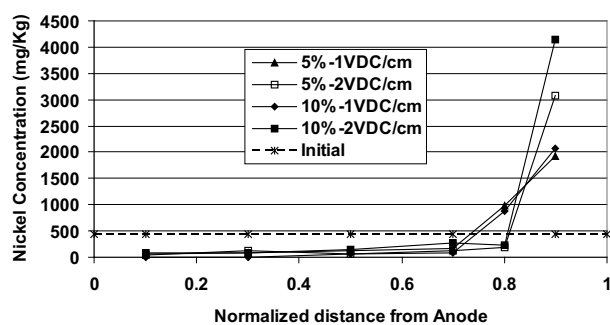


Figure 5
Nickel distribution in the soil after testing

Figure 5 shows the distribution of nickel in various sections of soil from anode to cathode at the end of testing. Nickel concentration remained very low and almost similar in all the tests throughout the first four sections from anode. It increased in the fifth and the sixth sections in case of 1 VDC/cm test. Whereas in case of 2 VDC/cm test, nickel concentration in the fifth section decreased almost to same as in the first four sections and in the sixth section, however, it increased to a very high value. In general, the concentration of nickel at cathode was very high in all the tests due to the high pH conditions existing near cathode. Nickel migrates towards the cathode in its cationic state. Under acidic conditions, solubilization of nickel as well as its hydroxides is favorable. Therefore, in the first four to five sections from anode, the concentration of nickel was very minimal. This implies that nickel was dissolved in the pore solution and moved towards the cathode due to electromigration and electroosmosis. The low pH conditions as in case of 2 VDC/cm tests caused high solubilization of nickel. As a result the mobility of nickel in most of the sections towards cathode was higher in the tests with 2 VDC/cm. However, in case of 1 VDC/cm tests, the pH was comparatively high, implying lesser solubilization and mobility of nickel towards cathode. Electromigration was the dominant transport process as compared to electroosmotic advection for the nickel in all tests. The electric potential was increased with the premise that it would increase the ionic mobility in the soil, thereby inducing higher electroosmotic flow. However, the creation of very low pH conditions in the soil near the anode caused the soil mineral surface to become positive,

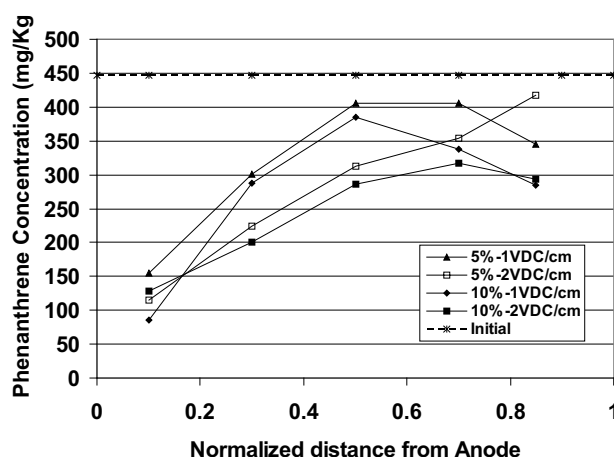


Figure 6
Phenanthrene distribution in the soil after testing

implying a positive zeta potential that decreases electroosmotic flow towards the cathode. In addition, the rapid flow conditions during the initial stages of testing with 2 VDC/cm could not be sustained due to faster nickel migration and precipitation near the cathode.

The distribution of residual phenanthrene in the soil after testing is shown in Figure 6. Phenanthrene concentrations decreased throughout the soil as compared to the initial concentration in all the tests. However, phenanthrene removal into the electrode reservoirs was insignificant in all the tests. Phenanthrene concentrations were higher from anode to cathode in all the tests. The concentration of phenanthrene remained lower throughout the first five sections in case of tests with 2 VDC/cm than those in tests with 1 VDC/cm. However, in the sixth section, the concentration of phenanthrene was comparatively higher in case of 2 VDC/cm tests (ranging from 340 mg/kg to 411 mg/kg) than that in case of 1 VDC/cm tests (ranging from 323 mg/kg to 360 mg/kg).

Based on the concentrations in effluent as well as the initial and final concentrations of phenanthrene within the soil, the percent removal, percent oxidation, and combined remedial efficiencies were calculated and these results are summarized in Table 5 and Figure 7. These results show that the percent removal of phenanthrene is negligible and overall remedial

Table 5
Phenanthrene Removal, Oxidation and Overall Remedial Efficiency

Test	Number of Pore Volumes	Initial Mass of Phenanthrene in Soil (mg)	Final Mass in Soil (mg)	Mass in Effluent (mg)	Mass Removed %	Mass Oxidized %	Remedial Efficiency %
1	1.6	89.8	64.6	0.6	0.7	28.0	28.7
1R	1.4	90.6	66.3	0.6	0.7	26.6	27.6
2	1.5	89.8	62.5	0.5	0.5	30.4	30.9
2R	1.3	88.9	56.2	0.5	0.6	36.9	37.4
3	1.2	93.1	63.5	0	0	31.8	31.8
3R	1.2	88.2	59.8	0	0	32.3	32.3
4	1.3	84.1	49.4	0	0	41.2	41.2
4R	1.2	87.3	49.4	0	0	43.5	43.5

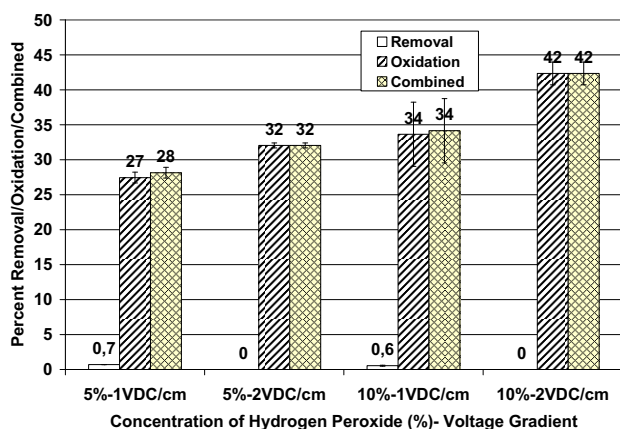
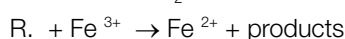
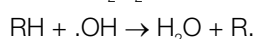
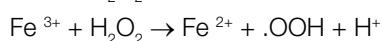
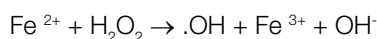


Figure 7
Phenanthrene removal, oxidation and overall remedial efficiencies

efficiency of phenanthrene is mainly attributed to oxidation. The remedial efficiency increased with voltage gradient. With 1 VDC/cm, about 28 % remedial efficiency was obtained in 5 % H₂O₂ test and about 34 % remedial efficiency was obtained in 10 % H₂O₂ test. With 2 VDC/cm, about 32 % remedial efficiency was obtained in 5 % H₂O₂ test and about 42 % remedial efficiency was obtained in 10 % H₂O₂ test. The oxidation of phenanthrene can be attributed to the following Fenton-like oxidation reactions (Walling, 1975):



In the above reactions, R represents phenanthrene or other organic contaminant. In this study, native soil iron was used as catalyst in above reactions. H₂O₂ upon reaction with ferrous ion produces hydroxyl radicals which oxidize phenanthrene. Therefore, in all the soil sections of all the tests, the concentration of phenanthrene reduced significantly. The increasing concentrations of phenanthrene from anode to cathode reflect the extent of H₂O₂ transport into the soil. The H₂O₂ solution was injected from the anode, and due to electroosmosis, more contaminated soil near anode was exposed to H₂O₂. As the contaminant comes in contact with H₂O₂, Fenton-like oxidation occurs and therefore more phenanthrene oxidizes near anode. The transport of H₂O₂ further into the soil depends on the extent of electroosmotic flow. However, as the current values decreased with time, the electroosmotic flow also decreased. Therefore, the interaction of H₂O₂ and contaminated soil decreased towards the cathode. Hence, the residual phenanthrene concentrations increased from anode to cathode in all the tests.

Figure 8(a) shows that the total iron (Fe) concentration in soil increased gradually from anode to cathode in all the tests with 1 VDC/cm. The iron concentration was higher in all the sections from anode to cathode, with the concentrations ranging from 544 to 1200 mg/kg of dry soil. Only a very low amount in the range of about 0.1 to 0.3 mg of iron was removed from the soil in all the tests. The iron fraction available for Fenton-like oxidation reactions was determined by leaching experiments. These experiments consisted of mixing 2 g of soil with 10 mL of deionized water, shaking the mixture, and finally separating and analyzing supernatant for Fe concentrations. The distribution of leachable iron (Fe) in different soil sections

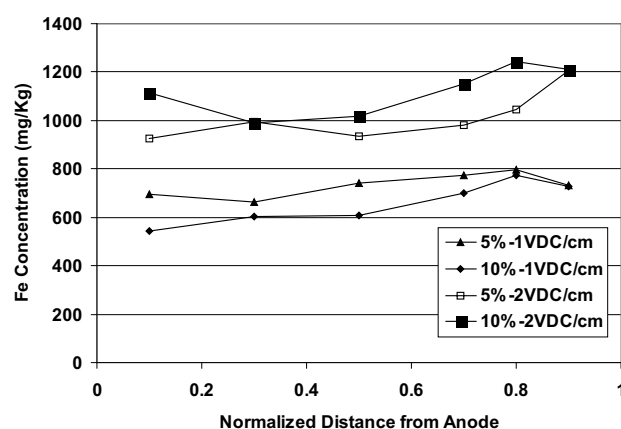
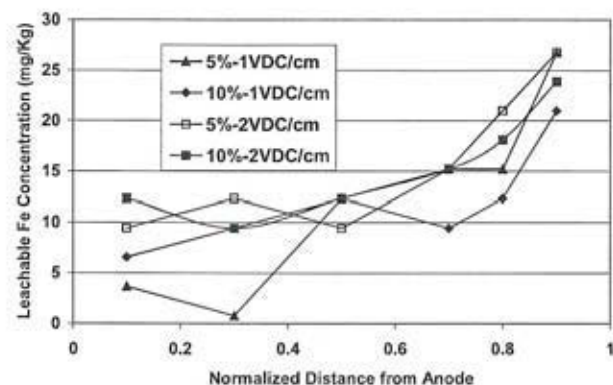


Figure 8(a)
Total iron distribution in the soil after testing



Leachable iron distribution in soil after testing

after treatment is shown in Figure 8(b). The leachable Fe concentration increased gradually from anode to cathode in all the tests. In case of 5 % H₂O₂ test, concentration increased from 4 to 27 mg/kg of soil from anode to cathode. In case of 10 % H₂O₂ test, concentration increased from 7 to 21 mg/kg of soil from anode to cathode. These results show that a considerable amount of native iron was readily available for Fenton-like oxidation, and that it was not the limiting factor for phenanthrene oxidation. The reason for low oxidation of phenanthrene may be due to low electroosmotic flow, leading to insufficient amount/concentration of H₂O₂ available for the oxidation of phenanthrene. Other factors such as soil-H₂O₂-phenanthrene interaction and solubilization may also have affected the overall oxidation of phenanthrene.

Conclusions

The main objective of this research was to assess the effects of voltage gradient on the remedial efficiency of integrated electrochemical remediation of mixed contaminants in low permeability soils. This remedial strategy aims to deliver H₂O₂ by electroosmosis to achieve Fenton-like oxidation of PAHs and to remove heavy metals simultaneously by electroosmosis and electromigration. Generally clayey soils possess high amount of iron and it is hypothesized that the native soil iron can serve as catalyst in Fenton's reaction. A series of bench-scale experiments performed on kaolin soil spiked with nickel and phenanthrene under two different voltage gradient conditions resulted in the following conclusions:

Significant migration of nickel occurred from anode to cathode in all tests. However, nickel precipitated near the cathode due to high pH conditions. Increasing the voltage gradient to 2 VDC/cm reduced pH throughout the soil as compared to that of 1 VDC/cm test resulting in greater migration of nickel towards the cathode. However, nickel precipitated in the close vicinity of cathode even in 2 VDC/cm test due to high pH conditions. The presence of phenanthrene and the use of hydrogen peroxide solution in anode did not appear to influence the soil pH distribution and transport of nickel in the soil.

A significant amount of phenanthrene was oxidized with the injection of hydrogen peroxide from anode into the soil. Phenanthrene oxidation is mainly attributed to the reactions with the hydroxyl radicals resulting from the oxidation of H_2O_2 in the presence of native iron in the soil. The native iron in the soil acted as an effective catalyst for the oxidation reaction. Clayey soils possess high iron content with sufficient fraction existing in easily leachable form available for Fenton-like reaction. The increased voltage gradient induced slightly higher electroosmotic delivery of hydrogen peroxide leading to increased phenanthrene oxidation.

The removal of both nickel and phenanthrene from the soil was very low. Although phenanthrene removal is not desirable, nickel removal is essential for the effective remediation. In order for integrated electrochemical remediation to be viable, strategies to enhance both phenanthrene oxidation and removal of nickel should be explored. These strategies may encompass increased delivery of H_2O_2 , better interaction of H_2O_2 with phenanthrene, and solubilization of precipitated metals near the cathode.

Acknowledgements

The authors are grateful to *Amid Khodadoust* and *Kranti Maturi* for assisting in performing the experiments and to the STAT Analysis Corporation and the University of Illinois at Chicago for providing financial support.

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Some geotechnical aspects of environmental protection in view of climate change

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Abstract

Changing rainfall patterns detrimentally affect geotechnical systems which are intimately linked to the quality of the environment. This paper outlines concerns for the future stability of landfills, tailings and tips, particularly those systems which were not subjected to rigorous analysis and design. Outline

geotechnical analysis is presented to show that even systems designed to comply with existing standards may not function properly in the future as a result of changes to groundwater regimes caused by climate change.

1 Introduction

Climate is controlled by the long-term balance of energy of the Earth and its atmosphere. A variety of natural events cause short-term, dramatic changes in climate, for example; large volcanic eruptions put tiny particles in the atmosphere that block sunlight, resulting in a surface cooling of a few years' duration,

El Niño events (periodic warming of the central and eastern tropical Pacific Ocean) typically last one to two years and change weather patterns around the world, causing heavy rains in some places and droughts in others.

It is now widely acknowledged that human activities have the potential to cause major climatic changes. The accumulation of greenhouse gases in the atmosphere as a result of human activities (consumption of fossil fuels, dumping of wastes in landfill sites, etc) will change the climate by enhancing the natural greenhouse effect, leading to an increase in the Earth's average surface temperature (Anon, 2001). At the same time, air pollution leads to high concentrations of small particles in the atmosphere that block sunlight and in certain regions of the world this 'dimming' effect may partially offset warming due to the greenhouse effects. Unfortunately, because most greenhouse gases remain in the atmosphere for a long period of time, even if emissions from human activities were to stop immediately, the effects of accumulated past emissions would persist for centuries. The globally-averaged temperature of the air at the Earth's surface has warmed between 0.3 and 0.6 °C since the late nineteenth century. The expected rise of globally-averaged surface temperature (relative to 1990) is 1 to 3.5 °C by the year 2100, with continued increases thereafter (Anon, 2006). As the Earth warms, more precipitation is expected and it is likely to fall over shorter intervals of time, thereby increasing the frequency of very heavy and extreme precipitation events. Figure 1 shows the recorded increase in the proportion of the USA which is subject to an unusually large amount of annual precipitation percentage arising from extreme weather events (defined as more than 50 mm of rainfall in a 24-hour period).

With 50-70 % of the global human population currently living in coastal areas, future sea level rise, alterations in storm patterns, and higher storm surges could have significant effects. However, it has to be realised that that flooding/sea level rise is

not necessarily the most serious concern – numerous people living inland are potentially at risk due to; ground instability at former and existing 'polluted' sites, enhanced mobility of pollution and the resultant negative impacts on eco-systems and drinking water supplies.

Higher surface temperatures are expected to cause an increased portion of precipitation to fall in the form of rain rather than snow. This is likely to increase both soil moisture and run-off. Increased heating will lead to increased evaporation and this will decrease the availability of soil moisture needed both for natural vegetation and agriculture in many places. Consequently the general concerns with regard to climate change and ground conditions can be summarised as:

- Saturation and instability of slopes.
- Drought conditions and loss of vegetation cover on slopes.
- Overland flows causing erosion, transport of solids, exposure of underlying materials.
- Saturation and detrimental effects on vegetation rooting systems.
- 'Flooding' of waste masses leading to polluted seepage, wash-out of contaminants, excess pressure on containment systems.

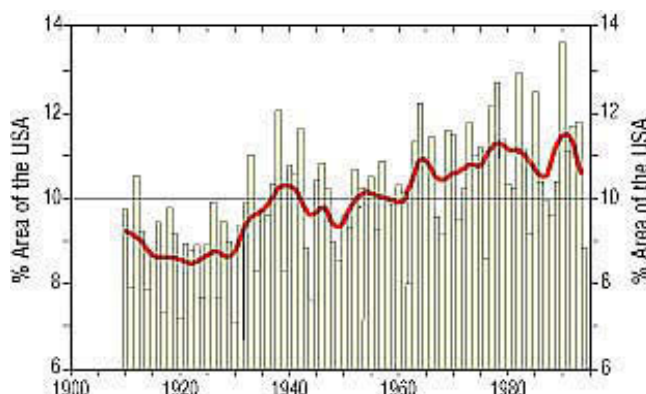


Figure 1
Occurrence of extreme rainfall events in the USA

2 Consequences

Climate change affects both shallow and deep groundwater and therefore affects the condition and stability of existing slopes, both natural and man-made. Whilst the traditional perception of many organisations is that landslides are mainly natural hazards (Anon, 2004), this is far from the truth in a developed society with an industrial heritage. Within the infrastructure there will be numerous embankments and cuttings associated with; roads and railways, spoil tips and heaps, engineered refuse disposal sites, covered deposits and lagoons of contaminated and polluted materials, repositories for sewage and dredging sludges, water reservoirs and drainage systems, floodbanks, etc. Furthermore, increased urbanisation and population growth has resulted in many people (and dwellings) being in close proximity to a variety of man-made slopes, many associated with environmental contaminants/pollutants. *Figure 2* is an aerial view of a sewage treatment works. In the centre of the picture is a lagoon containing processed sewage (up to 8 m deep) which is heavily contaminated with heavy metals. The lagoon embankments were not designed or built to any engineering standard. If any of the embankments become unstable then residents living adjacent to the site (in the dwellings to the right of the lagoon) and the local groundwater system (a major water-course is visible in the left-hand bottom corner of *Figure 2*) will be very detrimentally affected. Any region with a long industrial heritage will have a mixture of old facilities, modern facilities and future ones that can seriously affect the environment if they fail to work properly. Climate change is important for these facilities in different ways. *Table 1* contains an outline of some of the potential threats.



Figure 2
Large sewage sludge lagoon adjacent to residential accommodation

Table 1
Potential threats from slope instability

Case	Effects
Movement of geological materials from natural slopes	Changes in flow patterns, blockages, sedimentation, scour around structures, flooding. Damage to infrastructure, loss of land, damage to drainage systems. Loss of life.
Movement of surface materials on waste tips	Exposure of waste materials to precipitation – generation of polluted run-off water, penetration of water into wastes leading to greater instability, loss of vegetation and bio-diversity, loss of land
Movement of covers over landfilled waste	Exposure of waste to precipitation – generation of leachate, overload of leachate extraction and treatment systems, collapse of waste slopes and creation of flow slides
Movement of the face of tailings 'dams'	Increase in egress of polluted water through the face, potential for instability of the whole facility (leading to widespread ground and water pollution).
Instability of dyke slopes	Reduction of capacity of flood banks, blockage of land drainage systems, increased sediment in water courses

Natural slopes evolve as a result of natural processes such as erosion and movement over a long period of time. Stable slopes result from the soil having sufficient shear strength to resist gravitational forces and the slope acquiring a suitable geometry which may change gradually/incrementally to accommodate progressive changes in prevailing ambient conditions. On the other hand, man-made slopes are imposed on nature and to provide stability they are designed for a specific combination of geometry and strength at their most critical/vulnerable period. The design ensures that the slope is not too steep or high for the available shear strength of the soil and that there are no unforeseen or unrecognised weak planes, bands or layers in the ground. For man-made structures climate change can drastically change groundwater regimes from that anticipated by designers with the result that the actual Factor of Safety is less than that anticipated and it may even be so low that slope failure should be expected. If the ambient conditions (such as climate) change after the construction works have been designed and built this can negate the proper functioning of the system.

3 Slope stability

In general slope instability results from the in-situ shear stresses exceeding the available shearing resistance of the ground and the major factor influencing in-situ ground strength is the groundwater regime and associated pore-water pressures. These are affected by extreme rainfall and drainage pattern changes because they alter the weight of the potential sliding mass and decrease the shear strength of the slope material. The stability state of a slope may be assessed by calculating the ratio of the resisting force or moment (due to the shear strength of the slope material) to the disturbing force or moment (due to self weight of the slope material). This defines the Factor of Safety (FOS) and a value above unity is required for stability. The flatter is a slope the higher is its Factor of safety and the more stable it will be. However, practical considerations of cost (due to amount of land occupied by a slope, the volume of material within the slope, etc) determine that engineered man-made slopes are designed with low Factors of Safety, typically in the range 1.15 to 1.50 (Perry *et al*, 2001). These values are used despite the fact that the ground and the ambient conditions it experiences are inherently more variable and majorly less controllable than those relating to the lifetime performance of a steel or concrete structure – for which global design Factors of Safety will be in the region of 1.7 (steel) and 2.7 (concrete). Furthermore, any slope which has undergone some movement and has then become stable will have an overall Factor of Safety very close to unity. Hence, it is evident that in any developed region there will be numerous ground slopes that can be destabilised by changes in the severity of rainfall patterns.

Instability of slopes (both small and large scale ground movements) can be put into three categories:

- Surface erosion whereby loss of surface soil leads to formation of rills and gullies and loss of vegetation.
- Shallow, translational slides wherein a relatively thin veneer of soil (up to 2 m thick approximately) slides down the slope parallel to the surface of the slope.
- Deep-seated rotational movement of a large volume of soil which travels as an integral mass.

3.1 Surface erosion

Soil erosion is the removal of surface layers of soil and it involves a process of both particle detachment and transport by the disturbing agencies. Erosion is initiated by drag, impact or tractive forces acting on individual particles of soil. The two most common agents of erosion are rainfall and wind. This erosion is controlled by a number of soil, climatic and topographical factors including intensity and duration of precipitation, ground roughness, length and steepness of slope, inherent soil strength and type and extent of cover (Rao and Balan, 2000). Surface erosion by water commences when the kinetic energy of rainfall is transferred to individual soil particles, breaking the bonding between particles and moving the particles upwards and laterally. If the energy of the falling rain can be absorbed or dissipated by vegetation or impacting on some other soil cover or surface obstruction energy transfer to the soil particles which results in detachment will be reduced and there will be a consequent reduction in soil erosion.

If rainfall exceeds the infiltration capacity of a soil continued precipitation results in overland flow which begins at a shallow depth. The runoff water collects in small rivulets which may erode very small channels (rills). These rills may eventually merge into larger and deeper channels (gullies). Figure 3 shows gullies formed by water running down the surface of a tip of waste material (which is a difficult medium on which to establish good vegetation cover). If the soil is permeable and has a favourable structure, infiltration will be enhanced and overland runoff will be reduced. Slowing the flow of water downslope reduces the soil transport capacity of the thin sheet flow thereby minimising the displacement of dislodged soil particles.



Figure 3
Surface erosion of a spoil heap

Ground erosion is frequently the result of human activity, e.g. formation of cuttings and embankments when roads are built, creation of waste tips, removal of trees and other vegetation from slopes, placement of final caps over engineered landfill sites. In the latter case the establishment of vegetation on the final caps over landfill sites is essential for long-term effectiveness of the cap as it settles and distorts and undergoes cyclic wetting/drying and heating/cooling.

It has been estimated that soil erosion by water and wind is responsible for about 56 % and 28 %, respectively, of worldwide land degradation (Wibisono, 2000). Furthermore, the US Army Corp of Engineers has estimated that in the USA alone the damage caused by soil erosion costs at least £140M annually (Wibisono, 2000).

3.2 Translational slide

Translational slides within slopes are usually relatively shallow with the failure surface being more or less parallel to ground surface. Typically the sliding surface is within 2 m of the ground surface, with most failures occurring at depths between 0.5 m and 1.5 m. This type of failure frequently occurs in natural slopes with adverse geological history and in man-made slopes where ground saturation has occurred. The very form of construction in some man-made slopes, e.g. the presence of geomembranes in landfill caps, may promote translational sliding. For a long slope the steady state groundwater regime consists of a phreatic surface (water table) more-or-less parallel to the ground surface with groundwater seepage essentially parallel to the ground surface. After failure the saturated materials of the slipped



Figure 4
Exposure of geomembrane due to translational slide of covering soil layer

mass may easily degenerate into a mudslide which sloughs off the slope leaving behind the same unstable geometry. The slide may also result in the exposure of man-made materials to adverse conditions, e.g. the exposure of polymeric geomembranes to UV attack (Figure 4).

A translational slide is analysed on the basis of force equilibrium along a potential sliding plane. If we consider the stability of a small element of the potential translational slide material wherein the element has weight W , the ground surface has a slope angle of β and the resistance to sliding along the potential slip plane is T , then the Factor of Safety (FOS) against sliding is given by;

$$\text{FOS} = \frac{T}{W \sin \beta} \quad (1)$$

The estimation of W is relatively straightforward but because of the thinness of the sliding mass the value of W is, in a geotechnical engineering context, relatively small. Thus the value of FOS is rather sensitive to the absolute value of T – this term is dependent on a number of parameters that are difficult to assess (such as effective cohesion) and that are very sensitive to groundwater regime (because of the low value of total stress).

3.3 Rotational failure

Deep-seated failure occurs by a soil mass sliding on an approximately circular failure surface (the most common case) or on a failure surface composed of a series of straights. Such

failure has a variety of consequences; damage/disruption of systems and structures within and on the ground, exposure of underlying materials (both natural and man-made, loss of containment and retention functions, release of contaminated semi-fluid masses, etc. Rotational instability is usually a deep-seated failure mechanism with a large volume of material being involved in the movement. This mass is destructive by virtue of its size. Furthermore, the failed slope may be unable to resist forces from material within or behind it (such as water within a reservoir dam or slimes within a tailings facility) and a large scale catastrophic collapse could ensue.

The stability of a given situation is assessed in terms of the moment equilibrium and the Factor of Safety is written as

$$\text{FOS} = \frac{M_R}{M_D} \quad (2)$$

M_R is the total resisting moment (generally due to the shear strength of the ground) and M_D is the total disturbing moment (due to the self-weight of the unstable mass) and both are large in magnitude. M_D is a direct function of the unit weight of the sliding mass which is relatively insensitive to likely changes in groundwater regime – most of the material will already have a high degree of saturation (around 80 % or more) and a change to full saturation will only increase the unit weight by 4 to 5 %. The greatest potential change is to M_R due to uplift pressures and reduction in frictional resistance.

Many structures composed essentially of waste, particularly those formed some time ago, were not subject to rigorous analysis or design but because they were stable after construction and still appear stable today there is tacit assumption that they will remain stable regardless of environmental changes. However, the stability may be a result of fortuitous beneficial effects and the incorporation of hidden Factors of Safety by discounting effects which are difficult to quantify in an engineering way. The variations in patterns of rainfall/precipitation, ambient air temperature and evapo-transpiration activity due to global climate change will have significant effects on the groundwater regime within soil and the vegetation that it supports. This will cause changes to the state of the soil (degree of saturation, total and effective stress level, brittleness, etc) that directly control its stability and effectiveness as a technical component of an engineered system.

4 The shear strength of soils

In geotechnical engineering the shear stress (τ) needed to cause sliding failure of a soil has two components, i.e. friction and cohesion. The frictional resistance originates from the relative sliding of solid, rough grains within the soil mass and, by analogy with the 'traditional physics model' of one solid material sliding over another solid material, this resistance has been defined as being proportional to the stress pressing the rough surfaces together. Cohesion is some inherent strength which is independent of the current stress between solid particles and was initially postulated to explain the general behaviour of clays, i.e. soils which are apparently cohesive – it is now recognised that only heavily overconsolidated clays exhibit some form of true cohesion. However, when considering the stability of the near surface region of soils there are

two other effects that need to be taken into account, i.e. soil suctions and soil reinforcement by vegetation. Thus for an examination of the shearing behaviour of soil at very shallow depths the shear strength (τ) should be written as;

$$\tau = f + c' + s + r \quad (3)$$

Where;

f = sliding resistance due to contact between hard surfaces which are pressed together

c' = inherent cohesion resulting from the stress history of the soil

s = sliding resistance due to suctions between particles in a partially saturated soil

r = sliding resistance due reinforcement by the roots of vegetation

4.1 Frictional resistance

In soils the materials being pressed together are not solids, they are a collection of grains with a variety of shapes with spaces (voids) in between the grains – the volume of a soil which comprises voids is typically in the region of 35 to 50 % of the total volume. Furthermore the voids contain a mixture of air and water. The 'traditional physics model' does not account for the presence of water on the sliding interface between the solid materials. *Terzaghi* (1966) accounted for this difference by introducing the concept of effective stress (σ'_n), i.e. a notional stress which is directly proportional to the stress transmitted across the actual contact points between of the solid materials which are sliding. Thus, in general, the total stress (σ_n) acting across a sliding surface has two components – the effective stress (which generates frictional shearing resistance) and the pressure (u) in the pore fluid (water and/or air) which cannot generate directly any shearing resistance.

$$\text{Thus} \quad \sigma'_n = \sigma_n - u \quad (4)$$

$$\text{and} \quad f = \sigma'_n \tan \phi' \quad (5)$$

The term $\tan \phi'$ is in effect the friction coefficient between solid materials on either side of the sliding surface. The solid materials on either side of the sliding surface may comprise particles of geological materials but in some cases (such as in engineered landfill sites) one side of the sliding surface may be a solid, non-geological material, e.g. polymeric lining material used in the base or cap of an engineered refuse disposal site. If a protective layer (such as a polymer geosynthetic) is used in conjunction with the foregoing type of liner then the materials on either side of a potential sliding surface could be solid, non geological material. For soils, ϕ' ranges from about 20° (for clays) to around 50° (for gravels), i.e. a friction coefficient of between 0.4 and 1.2 approximately. If changes in groundwater regime mean that a soil moves from an unsaturated state to a fully saturated state then σ'_n may be effectively halved.

4.2 Cohesion

Effective cohesion (c') is an inherent shear strength which is dependent on the current internal state/structure of the slope material and is not directly dependent on the current stress level within the slope material. This inherent shear strength depends on the stress/strain history of the soil and how this has affected the packing of the particles within the slope material. For instance, desiccation of a clay soil creates very high suctions in the water within the voids between the particles (due to the surface tension effect in water) which pull the particles together so that they interlock to form a dense mass. Subsequent wetting of the particulate mass will reduce the suctions within the pore water but since the particles are not smooth the friction between them will maintain the interlocked nature of the mass and the particles will not return to their original packing state. In order to destroy the interlocking produced by desiccation it is necessary to apply some shear stress, i.e. the value of the effective cohesion. In effect the interlocking is readily destroyed by physical distor-

tion/straining of the soil so that particles are separated. Such distortion may be a result of the post-construction progressive development of that part of the shear strength of a soil mass which is due to friction between individual soil grains. However, when a small, 'undisturbed' soil sample is tested in a laboratory significant values of c' (around 20kN/m²) may be measured and initial stability of engineered slopes may support the existence of such large values of c' . Nevertheless it must be realised that this effective cohesion is rapidly lost due to soil deformation.

4.3 Soil suctions

When soils are partially saturated the water between individual grains behaves as if it were in a capillary tube (the diameter of which depends on the size of the soil particles and the extent to which the voids in the soil are filled with water, i.e. on the degree of saturation) and manifests surface tension effects. This induces a negative pressure within the pore water and this pressure is inversely proportional to the equivalent diameter of the capillary tube in the soil. The suction in the pore water pulls the solid particles together and thereby creates frictional resistance to sliding of the particles even without the application of an external stress pressing the particles together – it is precisely this partial saturation/surface tension effect which enables a sandcastle (damp sand) to be built with vertical faces but if the sandcastle dries out it collapses as the internal suctions are lost. *Mitchell* (1976) has quoted potential capillary heads within fine sand and silt of the order of 1.7 m and 3.5 m, respectively. These values would correspond to pore water suctions of around 17 and 34 kN/m² and would give rise to apparent effective cohesions in the range 10 to 20 kN/m² approximately. Fine-grained soils contain very narrow capillary passages and have low permeability and so exhibit very slow moisture content changes and can maintain pore suctions equivalent to tens of metres of water more-or-less indefinitely.

4.4 Vegetation effects

Vegetation is the major factor in the prevention of erosion of a soil slope through its influence on the effects of precipitation as indicated in *Table 2*.

Vegetation also affects the stability of a slope directly by providing mechanical enhancement of the strength of the soil. The roots (which have a relatively high tensile strength)

Table 2
Groundwater effects from vegetation

Effect	Comments
Rainfall interception	The volume of rainfall reaching the ground, and the rate at which it does so, is reduced (by the canopy action of plants) so that saturation of the upper soil layer is likely to be prevented
Surface water run-off	Vegetation reduces the velocity of flow so that the potential for surface erosion is less. Filtering of soil particles out of runoff is performed by stems and organic litter.

and the soil form a composite material akin to Reinforced Soil so that soil and roots act as a united mass. The soil friction angle is unchanged but there can be an apparent increase in cohesion of around 3-10 kN/m² which can extend for several metres depth in the case of tree roots (Coppin and Richards, 1990). In addition, the tap roots of many large shrubs and trees penetrate to great depths and thus into ground which

is considerably stronger than the surface material. The trunks and principal roots then act as individual cantilever pile walls providing a buttressing effect to up-slope soil masses. If the trees are sufficiently closely-spaced arching may occur between the up-slope 'buttresses'. Thus the presence of vegetation, and its type, can be crucial in determining the surface stability of both natural and man-made slopes.

5 Landfills

For the past 50 years waste disposal by landfill has been an integral part of the approach of all developed countries to waste management. Over that period there has been a huge increase in the complexity and level of sophistication of design and construction procedures. The early landfilling operations commonly involved the uncontrolled infilling of natural depressions and man-made excavations. This approach assumed that within the ground-water the concentrations of any contaminants derived from landfill would reduce to acceptable levels as they dispersed and were diluted under natural processes. Subsequently landfill philosophy moved to the objective of total containment and isolation of wastes and there was a major upsurge in the development of engineered waste disposal by landfill:

The earthworks techniques employed by the Civil Engineering industry were employed to form low permeability clay liners beneath the impounded refuse.

Man-made materials (geomembranes, bentonite mats, composite liners, etc) were introduced into the ground to provide basal barriers to the movement of leachate from the base and sides of the refuse.

Compacted clay caps, and subsequently composite caps incorporating geosynthetics and geomembranes, were placed over the top of refuse when a site was full.

The development of vegetation on completed landfills was encouraged to enhance the visual appearance of the site.

Landfilling has progressively given way to above ground disposal, i.e. 'landraising' or 'landforming' and this means that the stability of the side slopes of the refuse mound assumes even greater importance than before.

Since the active life of waste within a landfill deposit is between 25 and 40 years, it is obvious that Europe contains many thousands of active and former landfill sites of very different kinds in terms of size, type, structural configuration and composition. These facilities could seriously pollute the environment and ecosystems throughout the whole of Europe if climate change caused their containment systems to fail. The design measures that have the potential to be significantly affected by climate change are the final cap over the refuse and the basal lining system.

The final cap has several vital functions:

To minimise leachate production by preventing the ingress of rain and surface water into the underlying waste.

To prevent uncontrolled escape of landfill gas from decomposing refuse.

To provide a growing medium for vegetation which will restrict water ingress into the refuse and will also give an acceptable appearance to the completed landfill site.

Consequently final caps need to be composed of several layers, each of which performs a specific function so that the complete system operates as required. Figure 5 illustrates a typical current arrangement of layers – most are relatively thin and create a number of interfaces which are orientated parallel to the surface of the cap. Such an arrangement of thin sloping layers has great potential for instability through translational sliding.

To illustrate the potential effects of groundwater regime alteration due to climate change, the following hypothetical (but typical) situation will be analysed:

A cap with an overall slope of 1 vertical to 2 horizontal, i.e. a slope angle (β) of 26.5°.

The bulk density of the soil is 2 Mg/m³ (corresponding to unit weight, γ , of 19.6 kN/m³).

The water table is parallel to the surface of the slope, i.e. there is steady-state seepage down the slope.

The upper surface of the soil is vegetated, partially saturated and has been subjected to desiccation due to surface evaporation and water extraction by vegetation.

For translational instability a conventional geotechnical analysis of stability generates the following expression for Factor of Safety against downslope sliding (Sarsby, 2000);

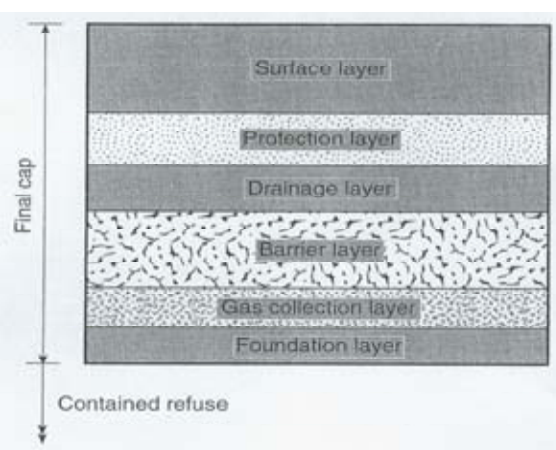


Figure 5
Multi-layered final cap over landfill

$$FOS = \frac{c'}{\gamma z \sin \beta \cos \beta} + \left(1 - \frac{\gamma_w d}{\gamma z}\right) \frac{\tan \phi'}{\tan \beta} \quad (6)$$

Where z is the depth to the failure surface, d is the height of the water table above the failure surface.

If the apparent shear strength due to pore water suction effects and surface vegetation are included (as mentioned previously) then equation (6) becomes,

$$FOS = \frac{c' + s + r}{\gamma z \sin \beta \cos \beta} + \left(1 - \frac{\gamma_w d}{\gamma z}\right) \frac{\tan \phi'}{\tan \beta} \quad (7)$$

Consider now the stability of the cap at the base of the surface layer shown in *Figure 5* which is intended for the support of vegetation and for which z would be typically around 0.3 m. Since most vegetation cannot survive in permanently saturated ground the designer of the cap would assume that the surface layer was not saturated (hence the presence of the drainage layer in *Figure 5*) so that d was equal to zero. Since the surface growing medium would need to be a fine-grained soil even a conservative designer would be unlikely to assume an effective cohesion of less than 2 kN/m² and an effective friction angle of less than 25°. For these conditions the conventional estimation of Factor of Safety immediately after construction would give

$$FOS = \frac{2}{19.6 \times 0.3 \times \sin 26.5 \cos 26.5} + \left(1 - \frac{9.8 \times 0}{19.6 \times 0.3}\right) \frac{\tan 25}{\tan 26.5}$$

$$= 0.85 + 0.94 = 1.79 \quad (8)$$

The value of 1.79 is significantly greater than unity and would be deemed acceptable. In fact, once vegetation had thoroughly established itself the overall Factor of Safety would be considerably larger (due to inclusion of pore water suction effects and root reinforcement), i.e.

$$FOS \text{ (actual)} \approx \frac{2 + 15 + 7}{19.6 \times 0.3 \times \sin 26.5 \cos 26.5} + 0.94 \approx 11 \quad (9)$$

Consequently the slope would be very stable and quite acceptable. However, because of the major contribution of the cohesion terms very little frictional resistance (about 9 % of the total which is potentially available) would need to be developed. Should the surface layer become saturated (due to increased rainfall and blockage of the drainage layer because of increased washout of fine particles) then the pore suction effect would be lost and d would become 0.3. Thus the Factor of Safety would become;

$$FOS = \frac{9}{19.6 \times 0.3 \times \sin 26.5 \cos 26.5} + \left(1 - \frac{9.8 \times 0.3}{19.6 \times 0.3}\right) \frac{\tan 25}{\tan 26.5}$$

$$= 3.83 + 0.47 = 4.30 \quad (10)$$

The new FOS is still significantly greater than unity and so the slope would still be stable and subject to very little ground movement. However, if the ground saturation persists then the vegetation will die and its reinforcing effect will be greatly reduced or even lost completely. In the latter case the Factor of Safety will become



Figure 6
Translational slide due to soil saturation and loss of vegetation

$$FOS = \frac{2}{19.6 \times 0.3 \times \sin 26.5 \cos 26.5} + \left(1 - \frac{9.8 \times 0.3}{19.6 \times 0.3}\right) \frac{\tan 25}{\tan 26.5}$$

$$= 0.85 + 0.47 = 1.30 \quad (11)$$

According to equation (11) the slope should be still stable. However the contribution that frictional resistance needs to make to achieve an overall FOS of unity has now risen to approximately 32 % of the total available and this will mean that significant deformation of the soil will be needed. This movement will cause some loss of the effective cohesion (for the reasons given earlier) and more frictional resistance will have to be developed – this means more ground movement and more loss of effective cohesion and so on until the slope fails because all of the cohesion has been lost. *Figure 6* illustrates the foregoing progressive type of failure due to soil saturation.

In the case of a deep-seated failure the Factor of Safety is defined as in equation (2) and it is the M_R term that is most prone to decrease as a result of long-term conditions being different from that assumed by a designer. For general rotational failure

$$M_R = \text{function } \{(\sigma_n - n)\alpha \tan \phi'\} \quad (12)$$

The term α is a function of the roughness of the sliding interface and for a particulate material such as refuse it is equal to unity. However, if the most critical sliding surface is a soil-on-solid surface (as could occur with a composite liner) the value of α could be around 0.5 (the early forms of geomembrane had very smooth surfaces). As a consequence, changes in water table position coupled with the presence of low friction interfaces could reduce the assumed design shear strength on the failure surface (originally estimated using standards current at a specific time and to a specification/brief applicable at that time) by up to 75 %. An example of a classical deep-seated rotational failure occurred at the Kettleman Hills



Figure 7
Movement of landfilled waste due to the refuse sliding on a geomembrane liner

hazardous waste repository (USA) which was essentially a very large, oval-shaped bowl excavated in the ground to a depth of about 30 m, into which the waste fill was dumped. It contained a highly-designed, properly-constructed composite double liner system (consisting of impermeable membranes, compacted clay layers, and drainage layers) to protect the ground and groundwater against transport of leachate out of the containment system. In March 1988 a slope failure occurred at the site with lateral displacements

of up to 11 m and vertical settlements of up to 4 m being measured. Surface cracking was clearly visible, as also were cracks and displacement on the exposed portions of the containment system (the liner) beneath the refuse. There was significant escape of polluted water. The subsequent technical investigation revealed that the failure resulted from reduction of the sliding resistance of the liner system due to the ingress of water (*Mitchell and Seed, 1990*). This type of instability is illustrated in *Figure 7*.

6 Spoil dumps from mineral extraction

6.1 Tailings dams and waste heaps

The quantities of tailings and other spoil from mineral extraction which have been disposed of on the ground surface are enormous and far exceed the volumes of materials handled by the whole construction industry. Severe environmental pollution can result from tailings dumps and spoil tips depending on the nature of the spoil and the proximity to rivers and streams.

Conventional impoundment storage is one of the cheapest methods of surface storage of processed mining waste from mineral extraction. The tailings are pumped from the processing plant and are dumped behind an initial low-height embankment. The fines settle out in a lagoon behind the embankment. Cyclones are used to separate out the sand fraction from the waste stream and this is used to raise the embankment. It is not usual for a tailings embankment to be built to its full height prior to disposal, unlike water retention dams. The idea is to progressively raise the embankment so as to avoid having a very low freeboard which can raise the phreatic surface of the embankment, thereby increasing the risk of failure. Three basic designs are adopted when constructing and raising dams, i.e. upstream, downstream, and centreline. The most common type used is the upstream design which raises the embankment towards the centre of the impoundment. The build material is thus deposited on the slimes. The centreline method raises the embankment on the slimes and also on the downstream face. For the downstream design, as the face is raised the toe of the slope moves away from the impoundment. Most older tailings impoundments around the world have been constructed by the upstream method, mainly due to the low volumes of build material and the low cost. Unfortunately much of the face (the 'dam') of the such tailings facilities sits on slimes which are likely to liquefy when they are vibrated. Consequently it is not surprising that the majority of large failures have occurred in facilities constructed by the upstream method.

With the upstream method of construction the tailings 'dam' is in reality a facing or veneer to the slimes deposit. In many cases involving slopes in tailings, shallow critical failure surfaces within the dam are found to approach the infinite-slope condition. On the other hand, for deep-seated rotational failure the circular failure surface will pass mainly through the slimes which have relatively little variation in effective shear strength parameters. The location of the phreatic surface (internal water level) within an embankment exerts a fundamental influence on its behaviour and control of the phreatic surface is of primary importance in tailings embankment design.

Whilst spoil heaps and tailings facilities have quite a lot of common features, with waste heaps the material can be regarded as more-or-less uniform throughout the tip rather than consisting of a veneer and a core. Most failures in spoil heaps have been associated with high moisture contents or water seepage. The effects of such failures have been compounded by the looseness (high porosity) of the older tips, leading to the formation of flow slides wherein the spoil mass is transformed into a liquefied state. The problem of colliery tip instability was brought into sharp focus by the disaster at Aberfan in 1966 when a slide involving some 100,000 m³ of colliery waste resulted in the death of 144 people (116 of whom were children). When the major slide occurred the height of the tip was approximately 66 m. The flowslide travelled down a 12.5° slope for a distance of about 480 m to the Junior School which it largely destroyed (*Figure 8*). Data for the discards showed that both the initial density and the volume decrease on subsequent saturation were very sensitive to placement moisture content. At Aberfan the material of the tip was loose colliery discards, tipped over the face of the spoil heap. The hydrogeology of the site was the triggering mechanism for the failure - after heavy rainfall artesian pore pressure developed in the sandstone beneath a relatively impermeable cover of boulder clay (*Bishop, 1973*). The resulting public enquiry concluded that water contributed to the disaster in four major ways:



Figure 8
Coalmining spoil tip failure at Aberfan

- Water issuing beneath the tip has caused back-sapping.
- Water under pressure acting in the tip material and in the foundation had initiated shear displacement along a pre-existing slip surface.
- Water filling the voids of the loose fill comprising the tip made it susceptible to a catastrophic flowslide.

Flowslides resulting from waste heap failure are not confined to tips of colliery spoil – notable flowslides have occurred within limestone waste, China clay spoil, fly ash, gold and copper tailings (Calembert and Dantine, 1964; Jennings, 1979; Street, 1987). In all cases the general causes have been material dumped in a loose state (to come to rest at its angle of repose) which has subsequently been rapidly saturated. With new tips, where modern engineering and management practices have been introduced, it is unlikely that they will become unstable.

6.2 Instability

The Factor of Safety against shallow translational sliding within the face of a tailings dam or waste tip can be investigated using equation (7) and an approach similar to that used previously for engineered landfills. However, there will be some essential differences between the situations:

The tailings dam is predominantly formed from coarse particles so that c' will be certainly zero – the same is generally true for spoil heaps although with time some weathering may lead to the formation of a very thin crust containing fine grains.

The face of the dam or heap will have a slope angle which is approximately equal to the angle of repose of the waste because of the method of deposition, thus $\beta \approx \phi'$.

The coarse material is very permeable so that saturation is extremely unlikely to occur. Furthermore the face slope is so steep that even heavy rainfall will flow down, and run off, the face although gullying is likely to happen. Hence pore suctions will be maintained, leading to a value of s (in equation 7) of greater than zero. Mitchell (1976) quoted potential capillary heads in coarse sand and sandy gravel of 0.08 m and 0.03 m respectively – these heads would give rise to apparent effective cohesions of around 5 kN/m² and 2 kN/m², respectively.

Spoil heaps and tailings dams will not usually support any significant amount of vegetation (the bare surfaces dry out in the Summer and water flows rapidly down them in Winter) so that r will be zero.

Thus in equation (7);

$$\begin{aligned} \beta &= \phi', d = 0 \text{ (for any value of } z), c' \\ &= 0, s \approx 2 \text{ to } 5 \text{ kN/m}^2, r = 0 \end{aligned} \quad (13)$$

For most tailings sands the effective friction ϕ' is typically in the range 33° to 40° and γ ranges from 17 and 19 kN/m³ (Sarsby, 2000). Consequently the theoretical Factor of Safety against translational sliding would lie in the range 1.15 to 1.40 approximately. The worst-case, lowest value is greater than unity (however marginally) and changes in rainfall pattern



Figure 9
The slimes lagoon of a tailings dam constructed by the upstream method.

alone would not be expected to cause translational instability of old tip and tailings slopes.

For tailings facilities and spoil heaps a deep-seated rotational failure will, paradoxically, pass through uniform (in a geotechnical context) material. Furthermore the shear strength parameters will be very similar to the material in the face of the deposit. Also the slimes material will be normally consolidated and fully saturated so that

$$c' = 0, \beta \approx \phi', s = 0, r = 0 \quad (14)$$

In addition the waste materials will usually be founded on a strong stratum so that rotational failure cannot penetrate below the base of the waste. Consequently stability of the waste can be made using stability charts (such as the ones produced by Tesarik and McWilliams, 1981) as well as by conventional rotational failure analysis. Such analysis, for ϕ' values of 33° and 40°, indicates that in order for tailings dams and waste tips to have a Factor of Safety at least equal to unity the freeboard, i.e. the depth of the water-table beneath the upper surface of the spoil, must not be less than approximately 50 % of the depth of spoil. Such a low water table is feasible for the free-draining discards of most spoil heaps – however, as shown by the case of Aberfan, most old waste tips do not contain basal drainage which is guaranteed to function effectively for the whole life of the tip. For many tailings facilities the water table behind the 'dam' is nowhere near as low as that required by the preceding analysis and yet they show no signs of instability. Consequently there must be some hidden factor that is providing stability. This is probably the development of effective cohesion within the slimes due to overconsolidation as a result of desiccation during the periodic construction process. This situation is illustrated in Figure 9 where people are standing on the stiff surface of desiccated slimes whilst the newly-deposited slimes in the left hand side of the picture are drying and cracking. Reliance on this effective cohesion is a major concern, because although the overconsolidation effect will persist in the upper layers of the slimes, it will be lost in the lower layers as the total stress increases due to raising of the dam. Furthermore the effective cohesion is rapidly destroyed due to movement (as described earlier) and so the old tailings facilities must remain highly sensitive to destabilisation by any form of shock – this can be caused by sudden loading due to an extreme rainfall event as happened at Buffalo Creek in the USA and Stava in Italy.

7 Conclusions

The evidence of the detrimental effect of climate change on the stability of both natural and man-made slopes is clear to see. However, the severity of the after-effects of rainfall-induced slope instability is increasing with time and some slope structures which have been stable to date are likely to fail in the future.

The potential for large-scale destabilisation of old tailings facilities by future extreme precipitation events is very high and a matter for serious concern.

There is a need to develop national/regional integrated risk assessment management plans for man-made slopes which will:

- identify the causal mechanisms of slope instability,
- take account of the influence of human activities,
- develop an accessible method for rapid assessment of site vulnerability,
- incorporate indirect appraisal and monitoring of slope condition.

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