

Development of high-rate passive sulphate reduction technology for mine waters

William Pulles¹, Laetitia Coetser¹, Ralph Heath¹, and Ritva Muhlbauer²

¹Pulles Howard & de Lange Incorporated, PO Box 861, Auckland Park, 2006 South Africa

²BHP Billiton, Johannesburg Technology Centre, 200 Hans Strijdom Drive, Randburg, 2125, South Africa

Introduction

The South African mining industry is facing major problems with regard to the management and treatment of contaminated mine water. These problems exist with regard to operational mines and, importantly, they also exist for mines which have ceased operations and which have long-term water quality problems.

Currently available effluent treatment technology for dealing with water quality problems is primarily of a chemical or physical nature. Although this technology is generally effective, it typically has very high capital and operating costs and intensive, ongoing, long-term maintenance requirements. This is a particular problem for those mines that have ceased operations and where it is not practical or cost-effective to construct an active treatment plant that requires constant supervision and maintenance.

An urgent need was, therefore, identified to develop low cost, self-sustaining, low maintenance passive treatment systems to address the problems of acidification and salinisation (in terms of sulphate) at operating, defunct and closed mines in South Africa, particularly as sulphate levels in discharged mine waters are regulated in South Africa. For this purpose, passive treatment is defined as follows:

A water treatment system that utilises naturally available energy sources such as topographical gradient, microbial metabolic energy, photosynthesis and chemical energy and requires regular but infrequent maintenance to operate successfully over its design life.

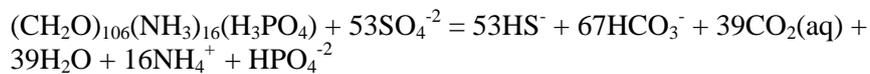
A major 10-year research programme was undertaken by Pulles Howard & de Lange Incorporated together with various other research institutions,

with funding from various agencies, including the South African government (Department of Arts Culture, Science and Technology's Innovation Fund), the Water Research Commission, Anglocoal, Eskom and all the major coal and gold mining groups in South Africa (Pulles et al. 2003). This research commenced in 1995 and went through three major and distinct phases, resulting in the registration of a patent application in 2001 (Pulles & Rose, 2001).

Background to the passive sulphate reduction process

The biological reduction of sulphate to sulphide, using an organic carbon source as the electron donor, is the process upon which the passive sulphate reduction technology rests. This process is used in both active and passive technology configurations, with the primary distinguishing feature of the passive technology being that the carbon source takes the form of a solid lignocellulose. This lignocellulose may be provided in various forms, including manure, straw, hay, sewage sludge, wood chips and a whole variety of agricultural solid residues.

While there are different ways of constructing the chemical equation for the biological degradation of lignocellulose in the reduction of sulphate, a useful one is the following:



The above equation is a simplified one, as the actual process within a biological reactor is a complex step-wise one, where the solid lignocellulose is first hydrolysed and solubilised, whereafter it is progressively fermented to simpler organic compounds which can ultimately be utilized by the sulphate reducing bacteria. Previous work (Pulles et al, 2003) undertaken in South Africa clearly showed that the hydrolysis of the complex lignocellulose was undoubtedly the rate-limiting step in a passive sulphate reducing reactor.

While this insight might seem trivial, a review of the literature on passive sulphate reduction suggests that many researchers were not aware of this phenomenon as the literature abounds with optimistic results for passive sulphate reduction, derived from relatively short-term laboratory studies. The many long-term reactor studies undertaken by Pulles Howard & de Lange clearly indicated a common time-related performance trend for

passive sulphate reduction reactors, typified by the performance curve shown in Figure 1 below.

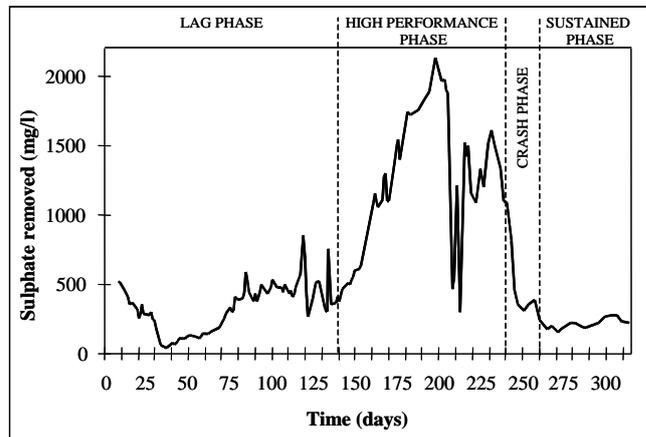


Figure 1. Typical sulphate reduction performance for passive reactor

The curve shown in Figure 1 is typical of a standard passive sulphate reduction reactor with regard to the following performance phases:

1. A lag phase with a typical duration of around 90 days (closer to 150 days in the case of Figure 1) during which time the reactor bacterial populations are adapting to the environment and sulphate reduction is relatively low.
2. A very efficient high performance sulphate reduction phase, typically not lasting longer than 8 months after reactor startup, during which time the readily available and hydrolysable energy in the lignocellulose is very efficiently utilized in sulphate reduction.
3. A sharp decline or crash in reactor performance, typically in the 8th or 9th month after reactor startup, resulting from the exhaustion of the readily hydrolysable lignocellulose. The onset of this phase may be retarded if the reactor is operating at excessively long hydraulic retention times.
4. A sustained but sharply reduced rate of sulphate reduction that will last for a period of 5 to 6 years if no replenishment of lignocellulose occurs.
5. Eventual cessation of sulphate reduction capability due to inability to hydrolyse the remaining lignocellulose material

Phases 1 to 3 have been observed numerous times in the hundreds of reactor studies undertaken by Pulles Howard & de Lange over the ten year

period from 1995 to 2004. Phases 4 and 5 have been observed in long-term field pilot scale studies undertaken by Pulles Howard & de Lange at the Vryheid Coronation Colliery in Kwazulu-Natal, South Africa. The monitoring programme is still ongoing on this pilot plant where 6 different sulphate reduction reactors have been in operation since 1996. With more than 600 000 data points, this pilot plant is undoubtedly the world's longest and best monitored passive sulphate removal plant and has revealed very valuable information about the long-term performance of these reactors. This research is being written up in the form of a research report to be published early in 2005 by the Water Research Commission.

Much of the research published in the literature is for studies that have not been undertaken for long enough to move through the crash phase into the low level sustained sulphate reduction phase and the results of such research should be used with caution and should not ever be used to inform reactor design exercises. Based on our research, we have found that the typical low level sustained sulphate reduction rate will be of the order of 300 millimoles per cubic metre per day ($300 \text{ mM/m}^3/\text{d}$) with lower levels being obtained if hydraulic short-circuiting is prevalent. This corresponds well with results reported in the PIRAMID Consortium report (2003):

Tentative design values for SO_4^{2-} removal in such systems range from 300 millimoles per cubic metre per day ($\text{mM/m}^3/\text{d}$) (Gusek, 1998; Lamb et al., 1998) to approximately $800 \text{ mM/m}^3/\text{d}$ (Willow and Cohen, 1998).

It is not clear whether the value of $800 \text{ mM/m}^3/\text{d}$ reported in the above extract from the PIRAMID report refers to data obtained from the high performance Phase 3 of the reactor or for the low level sustained sulphate reduction found in Phase 4.

It is furthermore relevant to note that the research that has been undertaken suggests that the energy contained within a lignocellulose carbon source can be divided into the following components:

- Firstly, there is the readily soluble fraction that is typically mobilised and washed out of the reactor within the first 2 weeks. This fraction comes available when the reactor is still in its lag phase and gives rise to the initial reasonable sulphate reduction performance within the first 2-3 weeks of the reactor. However, a large fraction of this energy is lost and the passive reactor typically exhibits a very high COD load in the first 2 weeks.

- Secondly, there is the readily hydrolysable fraction that requires bacterial population adjustment during the lag phase and is then subsequently utilized during the high performance sulphate reduction in Phase 2 to drive high rates of sulphate reduction. Sulphate reduction values in excess of 7000 mM/m³/d have been repeatedly observed. The exhaustion of this fraction of the lignocellulose is dramatically shown by the crash in reactor performance at around the 8th month of operation.
- Thirdly, there is the difficult to hydrolyse fraction that represents the bulk of the energy contained within the lignocellulose. This fraction represents the cellulose and hemicellulose that is protected by the lignin biopolymer that coats the lignocellulose fibres. This energy is partially available in a standard passive sulphate reduction reactor and will support a sulphate reduction rate of 300 mM/m³/d for a period of 5-6 years. Based on research undertaken by Pulles Howard & de Lange it has been shown that a standard passive sulphate reduction reactor will not mobilise all the potential energy represented by this fraction.
- Fourthly, there is the recalcitrant energy fraction which is simply not available to biological metabolic pathways but which would be released using aggressive chemical, physical and/or thermal action.
- Finally, there is the ash fraction that represents the non-reactive portion of the lignocellulose.

The real challenge in obtaining high rate passive sulphate reduction is in finding ways of liberating the difficult to hydrolyse fraction at a higher rate than 300 mM/m³/d and at a high conversion efficiency where the bulk of this fraction is utilized and only the fourth and fifth lignocellulose fractions are left.

Development of the novel passive treatment technology

This realization that passive sulphate reduction reactors were constrained by lignocellulose hydrolysis rates led to the initiation of an in-depth research programme to find ways of overcoming this rate-limiting step, thereby unlocking the energy available within the lignocellulose for sulphate reduction, at a rate high enough to make the technology economically viable (>600 mM/m³/d). This research pursued the following two different paths:

- Pretreatment (aerobic) of the lignocellulose to make the lignocellulose less resistant to anaerobic bacterial action.
- Understanding of the mechanisms of anaerobic lignocellulose hydrolysis in order to develop a process that could optimize this step.

The first leg of the research was undertaken in collaboration with the University of Pretoria and involved the fungal degradation of lignocellulose material. The objective of this research was to use various white rot fungi to weaken the lignin structure before the lignocellulose was packed into the anaerobic reactors, thereby affording the anaerobic bacteria better access to the cellulose and hemicellulose fractions. An offshoot of this research was an investigation into the utilization of the concentrated organic liquor leaching from the fungal reactors as an energy source in the sulphate reduction process. This leg of the research, while showing technical promise, was ultimately terminated on the basis of preliminary economic evaluations that indicated that even using best case data, the costs of such a process would far outweigh the benefits.

The second leg of the research was undertaken in collaboration with the Environmental Biotechnology Research Unit at Rhodes University and is reported on at this conference in a paper presented by Prof P Rose. This leg of the research was successful and the understanding obtained from this research led to the development of a novel reactor called the Degrading Packed Bed Reactor (DPBR) that was designed to optimize the hydrolysis of lignocellulose. This concept is novel insofar as the primary design and operating purpose of the reactor is lignocellulose degradation as opposed to sulphate reduction. The performance of the DPBR is measured not only in the rate of sulphate reduction, which is exceptionally high for a passive reactor, but also in the ability to produce an effluent high in organic loading (measured as COD) to be utilized in subsequent sulphate reducing reactors.

In terms of the typical passive sulphate reduction performance curve shown in Figure 1, the effect of overcoming the lignocellulose hydrolysis rate-limiting step is to significantly reduce the effect of the crash phase and to lift the sustained sulphate reduction phase to a higher level than the default 300 mM/m³/d efficiency found in a standard passive reactor.

The outcome of the research is the development and patenting of a new integrated and managed passive treatment process, known as the IMPI Process. The essence of the IMPI process is the subdivision of the overall

treatment process into individual units, each designed and optimized to perform a key function. This integrated process is shown in Fig. 2 below.

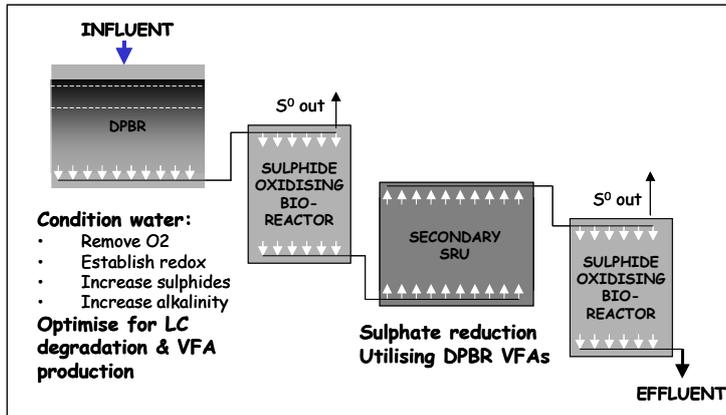


Fig. 2. Schematic of the IMPI process

The purpose and essential features of the 4 different stages can be summarised as follows:

Reactor 1: Degrading Packed Bed Reactor (DPBR): This reactor is packed with multiple layers of specially selected carbon sources (electron donors) and also receives regular inputs of readily available carbon. The primary functions of this unit are to rapidly condition the influent by removing dissolved oxygen, establishing the desired redox conditions and producing elevated levels of sulphides and alkalinity in the first portion of the reactor. The remainder of the reactor is devoted to the optimized hydrolysis of lignocellulose material and the production of volatile fatty acids (VFA). The effluent from this reactor will contain reduced levels of metals and sulphate and elevated levels of sulphides, alkalinity, VFAs and nutrients.

Reactor 2: Primary Sulphide Oxidising Bioreactor (PSOB): This reactor contains very little or no carbon source and has the primary function of oxidizing sulphides to elemental sulphur for removal from the reactor while minimizing changes to the VFAs, nutrients and redox conditions.

Reactor 3: Secondary Sulphate Reducing Reactor (SSRR): This reactor contains a specially selected single carbon source rather than a multiple

layer, multi-carbon source. The primary function of this reactor is to utilize the VFAs produced in the DPBR and to remove additional sulphate down to the design level. The effluent from this reactor would contain reduced levels of metals, sulphate, VFAs and nutrients and elevated levels of sulphides, and alkalinity.

Reactor 4: Secondary Sulphide Oxidising Bioreactor (SSOB): This reactor contains very little carbon source and has the primary function of oxidizing sulphides to elemental sulphur for removal from the reactor.

If required, a final aerobic polishing stage could be added, primarily to remove residual levels of VFAs and nutrients. The individual units could be combined in a tapered –up or tapered-down configuration, i.e. one DPBR to many SSRRs or vice-versa, depending on the design duty of the reactors.

The longest operating DPBR has continued for a period of 3 years and is currently still operating. Its performance in terms of sulphate removal rate is shown in Figure 3 below.

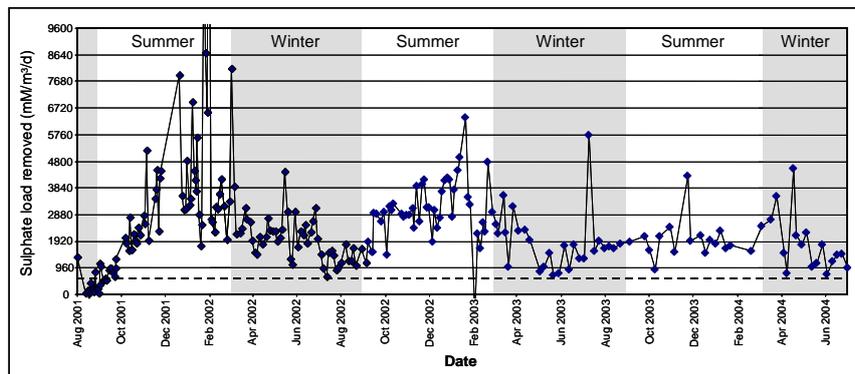


Figure 3. Long-term performance of DPBR in terms of sulphate load removal

The data presented in Figure 3 does indicate that the DPBR is able to sustain the long term sulphate reduction or removal rate at or above 2000 mM/m³/d, representing a 7-fold increase in performance over the standard sulphate reduction reactor with a removal rate of around 300 mM/m³/d. Figure 3 also indicates that the removal efficiency fluctuates with the winter and summer seasons and that there is a gradual decrease in the removal rate due to the consumption of the lignocellulose material. The

operating strategy for a DPBR is therefore to incorporate an annual replenishment of lignocellulose material to replace the fraction that has been consumed. While this has not been applied to the reactor depicted in Figure 3, this action can be expected to exert an upward force on the performance curve to counteract the gradual decline.

Figure 4 presents sulphate removal (in mg/l), alkalinity production and sulphide production data for the same long-term DPBR reported in Figure 3. Figure 4 shows a much more marked seasonal effect on alkalinity production than on sulphate reduction or sulphide production. It is also of interest to note that the reactor effluent sulphide concentrations do not exceed 400 mg/l and are more typically at around 300 mg/l, suggesting that the maximum sulphate reduction that can occur in a single stage passive sulphate reduction reactor is around 900 to 1000 mg/l sulphate.

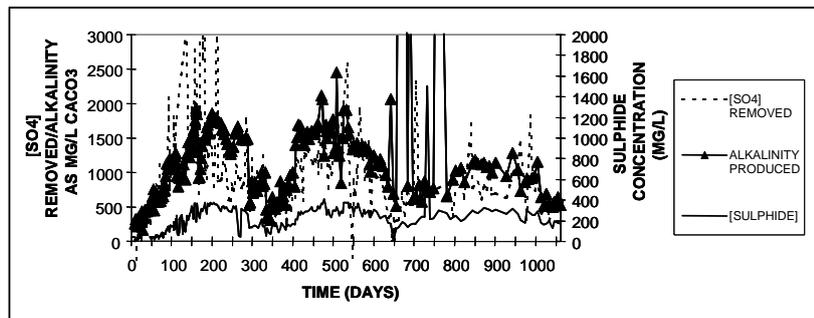


Figure 4. Long-term performance of DPBR in terms of sulphate concentration removed, alkalinity produced and sulphide produced

Design approach for the IMPI technology

The developed technology has been packaged in a suite of novel integrated passive mine water treatment technologies referred to as:

- IMPISURE:** Removal of sulphates, metals and acidity
- IMPIMATE:** Removal of metals and acidity
- IMPIPLUME:** Removal of sulphates, metals and acidity in groundwater plumes

While the research programme has resulted in the development of a detailed descriptive process model that serves as the basis for plant and process design, the implementation of the technology needs to take account of various site-specific factors. The treatment process is a

biological one and therefore needs to be customized to take account of unique site conditions, specifically the following:

1. Each mine water has a unique chemistry that affects the biological process design and the commissioning and operating procedures.
2. The technology requires a large inventory of carbon substrate on initial construction and each site will have a unique blend of carbon substrate that is sourced as close as possible to the plant.
3. Each site has unique physical features that affect the civil construction and the available hydraulic driving head.

In order to address these uncertainties, we have developed a standardized 4-phase implementation procedure that ensures that the above factors are fully defined and incorporated into the existing descriptive process model in order to produce an optimized site-specific plant design. This procedure is shown schematically in Figure 5 below.

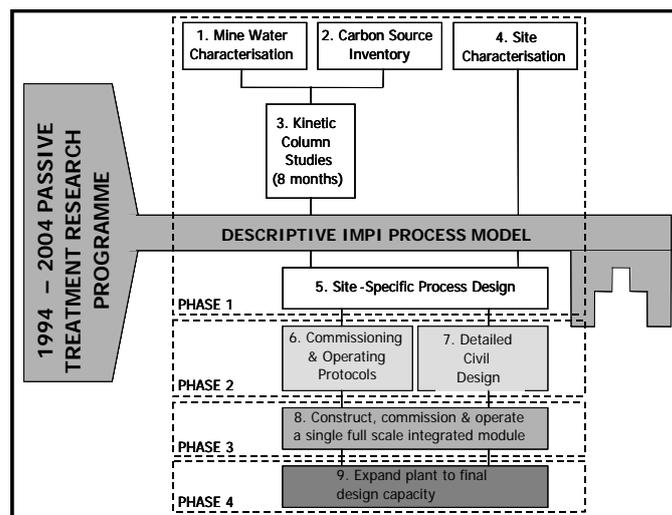


Figure 5. Implementation procedure for IMPI passive treatment technology

PHASE 1: DEVELOP SITE-SPECIFIC PROCESS DESIGN PARAMETERS

Step 1: Obtain all historical water quality records and any information that may exist on predicted future water qualities for the

anticipated design life of the plant. Evaluate these data in order to obtain a current and possibly future characterisation of the water to be treated. Establish and confirm the desired treatment objectives, i.e. which contaminants need to be removed and to what levels? Define the design treatment duty of the passive treatment plant and establish whether the IMPISURE or IMPIMATE is required.

Step 2: Based on the existing descriptive process model, undertake a first-order conceptual process design in order to establish a first estimate of the volume of carbon required as an initial inventory. Undertake a survey to establish potential carbon sources within a 25km radius of the proposed plant location and obtain representative samples of this carbon material. Undertake batch anaerobic evaluations of the carbon samples and compare to the carbon source performance database in order to obtain a ranking of carbon source suitability. Select a carbon mix or suite of mixes, to be utilized in the kinetic column studies as being representative of the desirable carbon mix for a full-scale passive treatment plant.

Step 3: Undertake kinetic passive treatment column evaluations in the passive treatment laboratory using the carbon mix selected in Step 2 and using the actual mine water to be treated. Pulles Howard & de Lange have developed and constructed a unique R1.5 million passive treatment laboratory facility that comprises 50 continuously fed passive treatment reactors and a dedicated analytical laboratory. Four full-time technical staff are employed to operate these facilities.

In order to customize the descriptive IMPI process model to optimally treat the actual mine water under investigation, a set of 8 columns is operated for a period of 36 weeks. The experimental programme that is applied to these reactors is aimed at establishing performance criteria specific to the mine water and carbon mix at the site. It is not appropriate to undertake such experimental work on full-scale modules that each contain 1000 m³ or more of carbon substrate and the column reactors are used for this purpose. A 36-week test period is required to overcome the initial 60-90 day commissioning period and to generate sufficient data to enable extrapolation over a 20-year design life.

The experimental programme also develops the specific commissioning and operating procedures that may be required to ensure satisfactory treatment of difficult-to-treat mine waters. As part of the experimental programme, analyses of influent and effluent are undertaken for sulphate, sulphide, COD, alkalinity, pH, redox potential, iron, aluminium and manganese.

Step 4: Undertake site evaluations in order to establish potential physical locations for the proposed passive treatment plant components. These site evaluations include establishment of the following data:

- Obtain record of flow rates of water to be treated, including information on peak flow conditions.
- Establish available static hydraulic head on the water to be treated and establish practical limits to increasing this head.
- Obtain a site survey plan accurate to 0.5m contour levels for the selected potential site.
- Establish relevant floodlines and plot them onto the site survey plan.
- Excavate test pits down to 4-5m in order to establish ground conditions. Subject soil samples to suitable laboratory tests to determine structural and hydraulic properties.
- Prepare a scoping EIA report regarding the suitability of the proposed site for construction of a passive treatment plant.

Step 5: Utilise the data generated in Steps 1-4 in order to develop a site-specific process design that can be used as the basis for a detailed civil engineering design for the full-scale plant. This site-specific process design is fundamentally based on the descriptive IMPI process model that then incorporates inputs from Steps 1-4 described above to ensure that it is optimized in terms of the actual mine water to be treated and the actual carbon substrates to be used.

PHASE 2: DETAILED CIVIL DESIGN, COMMISSIONING & OPERATING PROTOCOLS

Step 6: Based on the column studies undertaken as part of Step 4, the site-specific commissioning and operating protocols are defined and appropriate commissioning and operating manuals are

prepared. These protocols / manuals also document the management, monitoring and auditing requirements for the particular plant.

Step 7: Based on the process design prepared in Step 5, and taking account of the developed commissioning and operating protocols, a detailed civil engineering design is prepared. The design is based on the construction of a single full-scale integrated module, although all water distribution and collection systems are designed with the objective of future addition of modules (Phase 4) to handle the full flow of water to be treated. Drawings and tender enquiry documents are developed to enable tenders to be sought for the actual plant construction and for subsequent plant commissioning and operation. This step also includes the completion of a proper EIA with the necessary water use licences to enable the plant to be constructed and operated.

PHASE 3: CONSTRUCT SINGLE FULL-SCALE INTEGRATED MODULE

Step 8: Once the construction tender has been awarded, construction work is undertaken to construct the plant in accordance with the civil design. Construction work needs to be closely supervised to ensure that work meets the specifications and that static commissioning objectives are met. Once construction has been completed, full wet commissioning commences – this commissioning may take as long as 90 days. The commissioning phase will also be used to ensure that the operating contractor is fully trained to adequately operate the plant. Once the plant is deemed fully commissioned, it will revert to the standard operational sequence and the appointed operating contractor will assume his/her operational duties. It is envisaged that Phase 3 would typically have a duration of 12-18 months before moving to Phase 4.

PHASE 4: EXPAND PLANT TO FINAL DESIGN CAPACITY

Step 9: Once the operation of the single full-scale module has proceeded satisfactorily for a period of 12-18 months, a decision can be made to expand the plant (i.e. construct additional parallel full-scale modules) to treat the full mine water flow. The

performance of the single operating full-scale module will be reviewed and, if necessary, the process design prepared in Step 5 will be reviewed and modified. Steps 6, 7 and 8 will then be redone for the additional full-scale modules.

To date, Phase 1 performance or amenability studies have been undertaken on 5 different coal mine effluents – 2 in the Witbank coalfields in the Mpumalanga province and 3 in the Vryheid coalfield in the KwaZulu Natal province. Typical datasets deriving from such a study are shown in Figures 6 and 7 below.

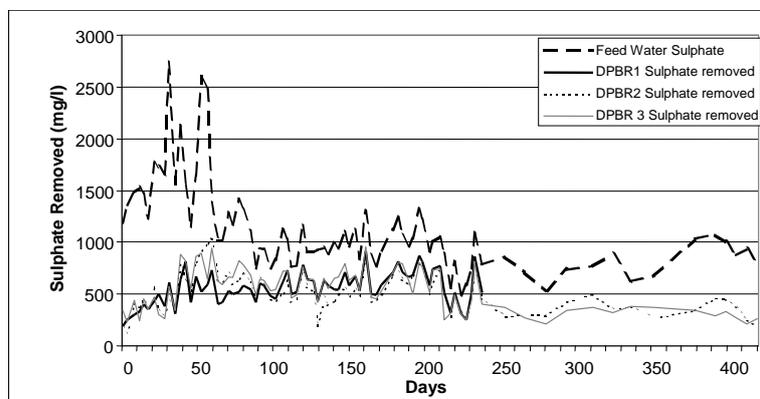


Figure 6. Design studies for a South African colliery – sulphate concentration removed

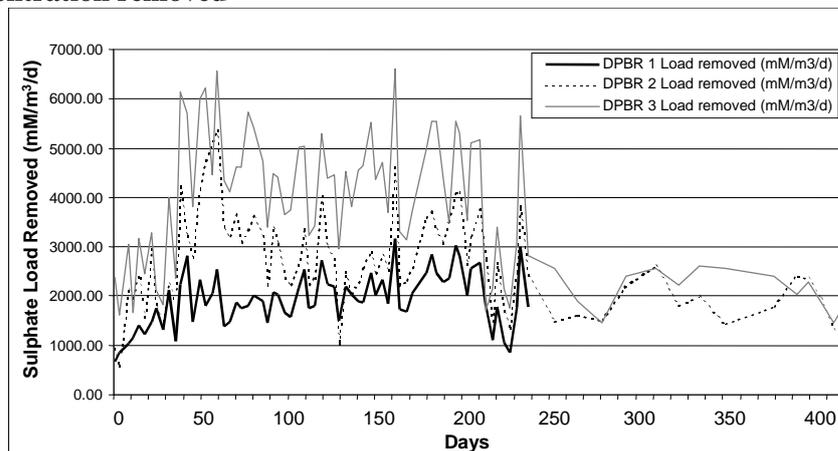


Figure 7. Design studies for a South African colliery – sulphate load removed

A proposal is currently being considered by the South African coal mining industry to construct and operate a single 200 m³/day full-scale module at a Mpumalanga Colliery and to evaluate its field performance for a period of 18 months.

Current research initiatives

A number of research initiatives are currently underway in South Africa that are relevant to the passive sulphate removal technology. These initiatives include the following:

1. Further development of the passive sulphide oxidation technology to passively convert sulphide to elemental sulphur in order to achieve passive biodesalination. This research has been ongoing for a period of around 5-6 years and is currently being undertaken by the Environmental Biotechnology Research Unit at Rhodes University with funding from the Water Research Commission. It is planned to incorporate the results of this research into a 20 m³/day sulphide oxidation field reactor to be operated as part of the 200 m³/d full scale module.
2. Evaluation of tapered configurations of the DPBR technology for specific use in treating highly acidic mine effluents. This work is being undertaken by Pulles Howard & de Lange with funding from BHP Billiton.
3. Evaluation of acidophilic sulphate reducing populations to treat strongly acidic mine effluents in passive treatment reactors. This research is being undertaken by Pulles Howard & de Lange and is being funded by BHP Billiton.

The research programme on the acidophilic sulphate reducing bacteria has been undertaken in order to develop the passive treatment technology to operate below the current pH cutoff value of 4.5, below which sulphate reducing bacteria do not perform well in passive treatment reactors. Bacterial populations were isolated from mine effluents and selectively cultured over a period of around 150 days to produce populations that have currently been treating strongly acidic mine effluents for around 300 days in duplicate reactor studies. Selected results from this research are shown in Figure 8 below.

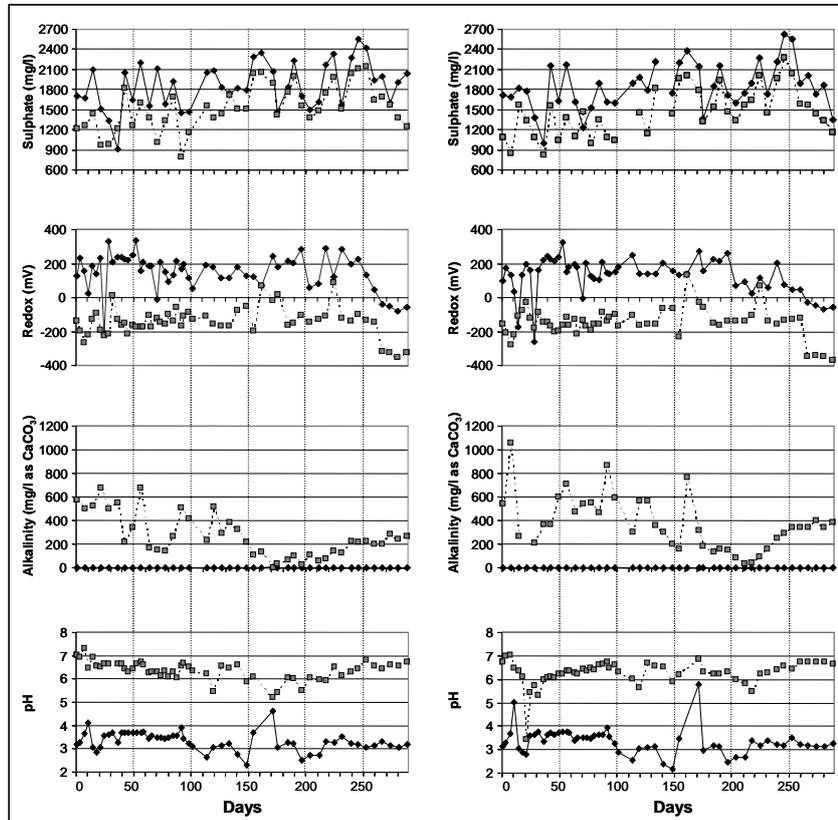


Figure 8. Performance results for acidophilic sulphate reducing populations

The original intent of the acidophilic research programme was to develop a biological reactor that could pretreat the water ahead of a DPBR by raising the pH to above 4.5 and reducing the redox to a point where sulphate reduction is possible. The data in Figure 8 indicate that this research objective is being met with influent pH of between 3 and 3.5 consistently being raised to a pH 6-7. Sulphate reduction of around 300 mg/l is being achieved and effluent redox values have been reduced to between -150 and -200 mV – suitable for introduction into a DPBR. Research on these acidophilic populations and reactors is continuing.

Conclusions

The research programme into the development of an integrated passive treatment system for sulphate removal from mine waters has been a sustained effort undertaken over a period of 10 years from 1995 to 2004 and additional evaluations and research are still continuing. We have seen progressive improvement in the performance of the system as the research has gone through the different phases. A major deviation from previous passive treatment approaches was initiated in this research programme and has resulted in the development of a novel patented system that is capable of exceeding published passive sulphate removal rates by around 700 percent. The only component of the new integrated system where development is still being finalised is the sulphide oxidizing bioreactor for which the fundamentals were developed in the research project. A number of design studies have been undertaken at South African coal mines and plans are well developed to construct the first full-scale module during 2005.

The technology developed during this research project is firmly based on two major South African biological sulphate reduction research initiatives. The first initiative has been ongoing for a period of around 10 years by Pulles Howard & de Lange and has focused on passive sulphate removal technology. The second initiative has been ongoing for a period of around 11 years by the Environmental Biotechnology Research Unit at Rhodes University. These two initiatives started off operating independently but have been cooperating with true synergy for the last 4-5 years. The collective manpower investment in these two initiatives is believed to be of the order of 60 – 80 man years and the collective budget in 2004 value is around R45 million. The leading position occupied by South African researchers in the field of passive sulphate removal technology is therefore not accidental, but is rather the product of a sustained and concerted research effort and a sustained support from the South African research funding agencies and mining industry.

Acknowledgements

The generous financial contributions of a variety of funding organisations over the last ten years is acknowledged. While all the South African gold and coal mining groups have contributed in some way to this research, particular acknowledgement must be given to the South African Department of Arts, Culture, Science and Technology's Innovation Fund

programme, the Water Research Commission, Anglocoal and BHP Billiton.

The intellectual input of many researchers and research institutions over the years needs to be acknowledged, in particular the Environmental Biotechnology Research Unit at Rhodes University and Pretoria University and their staff. Various researchers have also been involved at Pulles Howard & de Lange over the years and their input to the technology development is also acknowledged.

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