Passive treatment of net-acidic mine waters: field experiences in the UK.

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Abstract: Passive treatment of mine waters has been introduced to the UK gradually since 1994, and the country now has the largest number and diversity of such systems in Europe. While central government-sponsored projects have mainly focused on treatment of net-alkaline mine waters with simple aerobic "reed bed" systems, a number of private and local government bodies have been more adventurous, in applying compost-based passive systems to strongly net-acidic mine waters. A brief review of UK experiences in this theme to date reveals both the potential and some limitations of the approach. Anaerobic wetlands and Successive Alkalinity Producing Systems are currently in use in the UK, and construction of a permeable reactive barrier for acidic spoil drainage is also planned. Examples from across England and Wales are used to illustrate some of the problems inherent in the application of these types of passive treatment to systems with "flashy" and acidic runoff.

1 INTRODUCTION

Application of passive treatment technologies for remediation of discharges from active and abandoned mines is now widespread in the United Kingdom (UK). At the beginning of the new millennium there were 23 full-scale passive treatment systems, and a further 5 pilot systems, operational in the UK (Younger, 2000). The majority of the full-scale systems treat net-alkaline mine waters from abandoned coal mines. The key objective of these systems (typically aerobic wetlands, or reed-beds) is the oxidation of ferrous iron to ferric iron, and subsequent precipitation of ferric (oxy)hydroxides within the confines of the wetland. An alternative approach to passive treatment of net-alkaline waters, entailing rapid oxidation and accretion of ferrous iron on high surface area media, is also under investigation at the pilot-scale (Jarvis and Younger, 2000a).

Generally the selection of these discharges for treatment has been correctly made on the grounds of their impact on receiving watercourses. The impact assessment methodology used to order discharges according to their impact is detailed by Davies *et al.* (1997), and is critically reviewed by Jarvis and Younger (2000b). However, as these priority discharges are addressed it is becoming apparent that there is an increasing need to now attend to the more challenging task of ameliorating acidic discharges. Although the extent of net-acidic waters is less than their alkaline counterparts in the UK their impact on the aquatic environment can be far more severe, even if it is more localised. Typically the metal ion content of acidic waters is not limited to iron (see below), and Jarvis and Younger (1997) among others have illustrated how the acidity component of these discharges has severely detrimental effects on the ecology of the receiving watercourses.

After outlining the typical chemical nature of net-acidic waters in the UK, and discussing the principles of passive treatment with regard to acidic discharges, some field experiences of the application of passive treatment to acidic waters are discussed. Although attempts to treat acid waters have met with considerable success, significant problems still remain, and these are highlighted in the following discussion.

2 CHEMICAL CHARACTER OF NET-ACIDIC MINE WATERS

Table 1 presents chemical quality data for 5 acidic discharges across the UK. Whitworth A is one of the few long-established discharges from an abandoned deep mine which remains acidic. This is one of a series of discharges in the region that arise from drift workings abandoned in the 1960s (Edwards et al., 1997). The discharges at Aspatria and Nailstone #5 are similar in that they are both generated

Table 1 Chemical characteristics of selected net-acidic mine and drainage waters in the United Kingdom

Determinand	Whitworth A (South Wales)	Shilbottle (Northumberland)	Aspatria (Cumbria)	Nailstone #5 (Leicestershire)	Quaking Houses (County Durham)
Grid Reference	SS 800792	NY 220079		SK 430088	NZ 178509
Date (sampled)	30/03/99	25/06/99	11/02/00	20/11/97	11/02/00
Flow-rate (L/minute)	774				39.6
Temperature (°C)	10.8	14.6			
pH	5.94	4.09	2.9	3.2	5.8
Conductivity (μS/cm)	804	9450	3100	1570	7110
Alkalinity (mg/L as $CaCO_3$)	95	0	0	0	50
$Acidity_{calc}$ (mg/L as $CaCO_3$)[1]	171	3273	495	293	66
Calcium (mg/L)	76	474	265	114	250
Magnesium (mg/L)	47	1743	86	65	88
Sodium (mg/L)	15	382	16	11	1108
Potassium (mg/L)	12	90	6	8	188
Iron (total) (mg/L)	94.20	712.00	198.10	47.70	6.07
Manganese (mg/L)	1.15	234.00	14.45	7.00	4.26
Zinc (mg/L)	< 0.01	3.40	0.86		0.68
Aluminium (mg/L)	< 0.01	282.00	9.18	29.40	8.60
Sulphate (mg/L)	485	11485	2034	757	774
Chloride (mg/L)	13	456	171	168	1976

[1] *$Acidity_{calc} = 50\ [2Fe^{2+}/56 + 3Fe^{3+}/56 + 3Al/27 + 2Mn/55 + 2Zn/65 + 1000(10^{-pH})]$, where metal concentrations are in mg/L (adapted from Hedin et al., 1994)*

from the dissolution by surface waters of metalliferous salts on the surface of a spoil heap. The discharges at Quaking Houses and Shilbottle arise from perched aquifers within spoil heaps (Amos, 1999; Younger et al., 1997). At Quaking Houses recent capping of the spoil tip with clay in selected areas, coupled with dilution of the mine water with surface runoff, have resulted in this discharge being only marginally net-acidic (Table 1).

In the UK discharges from long-abandoned deep mines (such as Whitworth A) are rarely net-acidic. Younger (1997) illustrates how discharges from deep mines that have been recently flooded may be net-acidic. However, levels of acidity (and metal ion concentrations) typically decrease once the initial "first flush" of highly contaminated water ceases. Lithological conditions permitting, long-term generation of acidity is primarily dependent upon the availability of oxygen and water, which facilitate the oxidation of pyrite, as illustrated in Equation 1, below (after Stumm and Morgan, 1996).

$$FeS_2 + 3.5\,O_2 + H_2O \rightarrow Fe^{2+} + 2\,SO_4^{2-} + 2\,H^+ \qquad (1)$$

Because these requirements are generally only met in the unsaturated zone flooded workings rarely generate high concentrations of metals and acidity over the long-term, unless hydrologic conditions permit fluctuations in the level of the water table (Younger, 1998a). Spoil heaps are invariably unsaturated however, and therefore the potential for extensive and long-term pyrite oxidation is much greater than in deep mines. Factors that determine the level of contamination emanating from the spoil heaps listed in Table 1 are likely to include the residence time of water within the spoil, the degree of crystallinity of the pyrite (Carrucio, 1975), and in particular the sulphur content of the waste rock (Younger, 1998a). Marine bands, with sulphur content in the region of 4 - 6 wt% have the greatest acid-generating capacity. The spoil heap at Shilbottle is comprised of high sulphur content shales, and this is reflected in the very high concentrations of acidity and metal ions. Indeed, the chemical quality of the Shilbottle discharge is one of the worst recorded in the UK.

3 PASSIVE TREATMENT OF ACIDIC DISCHARGES IN THE UK

Many of the passive treatment systems currently operational in the UK are based on the design guidelines of the US Bureau of Mines (Hedin et al., 1994). Whilst the design of passive treatment systems for net-alkaline waters is based upon the iron load of the discharge, acidity load is the basis of design for net-acidic discharges. Current passive treatment options for such discharges are:

1) Anoxic Limestone Drains (ALDs)
2) Anaerobic (or compost) wetlands
3) Successive Alkalinity Producing Systems (SAPS)
4) Permeable Reactive Barriers (PRBs)

In the UK there are currently no ALDs in use specifically for the generation of alkalinity. This is because there are very few discharges that meet the criteria necessary for successful long-term operation of such systems i.e. Fe (III), Al and dissolved oxygen concentrations all less than 1 mg/L (Hedin et al., 1994). Nuttall and Younger (2000) have recently demonstrated that ALDs may be successfully employed to remove zinc as smithsonite, however.

The first anaerobic wetland for treatment of net-acidic mine water in the UK was constructed at Quaking Houses, County Durham. The design and construction processes for this 400 m^2 wetland, which was completed in November 1997, are described in detail by Jarvis and Younger (1999). This full-scale system was pre-dated by a 40 m^2 pilot wetland, which successfully removed acidity at a rate of 9.6 $g/m^2/d$ (Younger et al., 1997). The full-scale system removes acidity at a mean rate of 6.4 $g/m^2/d$, though removal rates are variable, ranging from -11.2 $g/m^2/d$ to 50.4 $g/m^2/d$. The exact reasons for the variation in removal rates are unclear, yet there is no obvious relationship between the acidity removal rate and any single influent physical or chemical variable. Flow-rate to the wetland is highly variable due to dilution by surface runoff. Although no quantitative proof is available it seems likely that flow-rate is an influential factor in the overall performance of the wetland, due to its controlling role on influent loading rates and residence time within the wetland. Despite the inclusion of islands and baffles within the wetland at Quaking Houses (Jarvis and Younger, 1999) hydraulic short-circuiting may well limit the efficiency of the system, particularly at high flow-rates. Again, however, demonstrating this point quantitatively has proved difficult. Robins (1998) undertook tracer tests at the site, and established a residence time of 40 hours for an influent flow-rate of 67 L/minute. Although Robins (1998) concluded that the wetland appeared to be a well mixed system, it remains unclear whether this is the case at the full range of influent flow-rates (13.9 L/minute to 420 L/minute). Nevertheless the Quaking Houses wetland continues to successfully reduce concentrations of acidity, iron and aluminium.

Operationally difficulties have been encountered with the maintenance of wetland systems across the UK. After 28 months of operation influent pipes (100 mm internal diameter) are starting to become blocked with iron precipitates and other sediment at the Quaking Houses wetland. In other cases accretion of iron in pipes is occurring rapidly, and is perhaps one of the most significant maintenance issues with regard to treatment wetlands (Adrian England, International Mining Consultants Ltd, UK, personal communication). Although the results of on site pilot-scale investigations have proved scientifically insightful and invaluable from a design perspective (e.g. Jarvis and Younger, 2000a; Younger et al., 1997) such systems are particularly prone to problems of metal accretion in pipes because pipe diameters tend to be much smaller (typically 20 mm internal diameter).

The wetland at Quaking Houses is comparatively small. In contrast a proposed scheme at the Aspatria site would involve the construction of one of the biggest compost wetlands in the world - approximately 15 000 m^2 (David Laine, International Mining Consultants Ltd., UK, personal communication). One of the main difficulties in undertaking the design and construction of treatment schemes for discharges such as Aspatria and Nailstone #5 is accommodating the highly variable flow-rates. Discharges arising from spoil heap surface runoff may increase in volume by an order of magnitude during a storm event. Because metal ion concentrations are derived from the dissolution of salts on the spoil surface, a concomitant decrease in metal concentrations as flow-rate increases may not be evident. At Nailstone #5 both the magnitude and the direction of changes in metal concentrations with flow-rate are highly unpredictable. As a consequence selecting an appropriate design loading rate is very difficult. Table 2 illustrates this point. Three separate physico-chemical analyses of the Nailstone #5 discharge are illustrated. It is clear that metal and acidity concentrations do not have a predictable relationship with flow-rate. The result is that calculations of theoretical wetland size required illustrate that wetland areas may vary by as much as two orders of magnitude depending which physico-chemical analysis is used. Two alternatives may be adopted to overcome this difficulty:

Table 2 Variation of acidity load and theoretical compost wetland area requirements at Nailstone #5, Leicestershire.

Date	**21/04/98**	**18/06/98**	**07/10/98**
Flow-rate (L/minute)	3	1200	20
pH	4.55	2.93	3.01
$Acidity_{calc.}$ (mg/L as $CaCO_3$)	166	332	709
Fe (total) (mg/L)	4.10	32.30	120.32
Mn (mg/L)	9.90	10.03	20.86
Al (mg/L)	25.00	35.50	73.28
SO_4 (mg/L)	1660	1473	2266
Acidity load (g/day)	717	573 696	3 465
Wetland area required (m^2)[1]	**102**	**81 957**	**495**

[1]*for illustrative purposes the design acidity removal rate of 7.0 $g/m^2/d$ is used (after Hedin et al., 1994).*

1) Construction of an appropriately sized balancing tank prior to any passive treatment system. In some cases such a unit may also act as a useful primary settlement lagoon, reducing the overall size of subsequent passive treatment systems. However, for acidic discharges precipitation of metals in a settlement lagoon may be minimal unless alkali dosing is undertaken.

2) Limit influent flow-rate, and therefore loading rate, and rely on dilution by the treated water to keep pollutant concentrations within any regulatory limits at the effluent point. Whilst clearly a compromise situation such an option may be favourable if the objective of a scheme is to achieve a reasonable improvement in receiving watercourse quality, rather than meet strict regulatory standards. Such an option has been adopted at Quaking Houses, despite the fact that regulatory limits are imposed on the effluent water quality. However, in this case pollutant concentrations typically decrease as flow-rates increase.

In April 1998 a passive treatment system for remediation of the Whitworth A discharge in South Wales was commissioned. This was the first full-scale system in the UK employing a SAPS unit (Younger, 1998b). Early monitoring results suggest that the system is very effective in reducing metal and acidity concentrations. For the period July to September 1999 mean influent and effluent iron concentrations were 65.2 mg/L and 5.7 mg/L respectively, and pH increased from 6.13 to 6.98 (data from Environment Agency, Welsh Region). As at Quaking Houses, however, quantitative descriptions of the hydraulics of the system are proving difficult. Younger (2000) rightly identifies the hydraulics of passive treatment systems as a key research issue for the immediate future.

A different approach has been adopted for the remediation of the spoil drainage at Shilbottle. A recent investigation by Amos (1999) has revealed that water is draining in a diffuse manner from a perched aquifer within the spoil. Highly polluted water (Table 1) emerges as toe drainage from the south-western side of the spoil heap. Work by Benner et al. (1997) and Blowes et al. (1995) has illustrated that permeable reactive barrier (PRB) technology may be appropriate in the hydrologic conditions evident at Shilbottle. Amos (1999) has recommended the installation of a PRB measuring 170 m long, 2.0 m wide, and 1.0 m deep. The recommended substrate for the system is an equal mix of a screened slurry compost and limestone. At the time of writing negotiations to progress this work to full-scale construction are underway.

4 CONCLUSIONS

- In the UK application of passive treatment technologies for remediation of net-alkaline mine waters is both widespread and largely successful. However, less attention has been paid to the more challenging task of ameliorating net-acidic discharges with passive systems.
- There are numerous acidic discharges in the UK, which are predominantly associated with spoil heaps. Acidity concentrations vary, but in one case concentrations in excess of 3000 mg/L as $CaCO_3$ have been recorded.
- Anoxic limestone drains for alkalinity generation have found little application in the UK. However the first anaerobic (compost) wetland in Europe continues to operate successfully at Quaking Houses, County Durham. Early indications suggest that the first Successive Alkalinity

Producing System (SAPS) in the UK, in South Wales, is also performing very efficiently.

- Design of systems for waters draining spoil heap surfaces is particularly difficult because of the highly variable acidity and metal loadings associated with such discharges.
- Perhaps the single greatest operational difficulty encountered with passive systems is the accretion of material within pipes, ultimately leading to complete blocking.
- Particularly pertinent to systems treating acidic waters, where intimate contact with a reactive substrate is crucial for effective treatment, is the issue of hydraulic efficiency. Characterising the hydraulic behaviour of passive systems for acidic mine water amelioration is proving difficult, but must be accomplished if maximum hydraulic efficiency is to be attained.

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Bierne oczyszczanie kwaśnych wód kopalnianych: doświadczenia terenowe w Wielkiej Brytanii

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Streszczenie: Bierne oczyszczanie kwaśnych wód kopalnianych wprowadzano w Wielkiej Brytanii stopniowo od 1994 roku, i kraj ten ma w tej chwili największą liczbę i różnorodność takich systemów w Europie. Podczas gdy projekty finansowane przez rząd skupiały się zasadniczo na oczyszczaniu alkalicznych wód kopalnianych prostymi systemami aerobowymi (podłoże trzcinowe), kilka prywatnych i lokalnych urzędów było bardziej odważnych i zastosowały bierne systemy do oczyszczania silnie zakwaszonych wód. Krótki przegląd doświadczeń brytyjskich w tym zakresie ujawnia zarówno możliwości jak i ograniczenia takiego podejścia. Obecnie wykorzystywane są anaerobowe filtry roślinne i „Alkaliczne Systemy Produkcji", planuje się także skonstruowanie przepuszczalnej reaktywnej bariery dla kwaśnego drenażu. Pokazane zostały przykłady z terenu Anglii i Walii celem zilustrowania niektórych problemów związanych z zastosowaniem tego typu biernego oczyszczania w systemach z kwaśnym odpływem.