Tracer Testing to Investigate Hydraulic Performance of a RAPS Treating Mine Water in South Wales[1]

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ABSTRACT

The Coal Authority commissioned a Reducing and Alkalinity Producing System (RAPS) at Tan y Garn abandoned coal mine, South Wales, UK, in 2006 to treat the acidic mine water discharge. Monitoring of the full-scale, operational scheme over 2½ years has demonstrated the RAPS has performed well in terms of sustained alkalinity production and also iron removal, despite a progressive decrease in permeability of the vertical flow bed. In order to understand the residence time of mine water within the system a series of ten tracer tests using lithium bromide and sodium chloride were conducted. Analysis of the salt tracer tests yielded a quantitative relationship between the naturally varying flow and the resulting residence time. The measured hydraulic performance is compared with the design expectations. This improved understanding of RAPS hydraulic and chemical performance confirms the appropriateness of published design for this passive mine water treatment method. As a result, The Coal Authority will continue applying the technology to other priority mine water discharges across the UK.

Additional Key Words: Passive Treatment, Alkalinity Production, Iron Removal

INTRODUCTION

Acidic mine drainage affects the quality and potential uses of water bodies in coal and metal mining regions worldwide (PIRAMID Consortium, 2003). Remediation by passive means, i.e. avoiding the use of power and chemicals, is normally desirable. Reducing and Alkalinity Producing Systems (RAPS) provide one option for passive treatment, summarised in the PIRAMID Design Guidelines (PIRAMID Consortium, 2003). RAPS have typically been applied to iron laden, net-acidic mine waters.

RAPS are usually configured as vertical flow reactors with the water passing through compost and limestone, these being layered or mixed media. The compost acts to provide an organic substrate with reducing conditions, removing any dissolved oxygen (D.O.) in order to minimise formation of aluminium or ferric iron precipitates within the media, which could armour the limestone and decrease media permeability. The compost may also encourage sulphate reduction, which generates a proportion of the alkalinity. The limestone is dissolved, and this is normally the dominant alkalinity producing process in

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the RAPS media. To allow sufficient time for these processes to occur, a minimum residence time of about 14 hours has been recommended (Younger et al., 2002), thus, the system hydraulics need to be designed for this.

In terms of the hydraulics of RAPS there may be three layers: 1) a top, supernatant water layer to distribute the flow laterally across the media, 2) the reactive, porous media in which vertical flow, usually downwards, occurs; 3) an underdrain, collection system. There must be sufficient driving head across the porous media to permit the downflow, otherwise part of the discharge will overflow. To allow for variable flow rates, and decrease with time of media permeability, adjustable outflow pipes have typically been employed to control the water levels (and, thus, head gradient) across the media (Younger et al., 2002).

Sapsford et al. (2007) observed that iron commonly precipitates as ochre, i.e. ferric oxy-hydroxides, on top of the RAPS media, and thus the top layer of supernatant water contributes to iron removal of the whole system. In a field trial they demonstrated that as an ochre layer thickens by self-accretion, its conductance decreases. Thus, in RAPS it appears likely that decrease in overall average permeability is due to the growth of the supernatant ochre layer, as well as changes within the media such as physical settlement and compaction, mineral precipitation, or bio-clogging.

Hydraulic performance appears to be problematic with many systems reported as having persistent overflows (e.g. Bhattacharya et al., 2008; Cravotta and Ward, 2008; Fabian et al., 2006). However, if the treated downflow is recombined with the overflow, and the result is a net-alkaline discharge, then the RAPS can provide effective treatment even with an overflow.

In order to understand system hydraulics and residence time, hydraulic tracer testing has been undertaken (Diaz-Goebes and Younger, 2004; Wolkersdorfer et al., 2005). Their breakthrough curves typically showed a sharp initial rise to peak concentrations, followed by a long tail. Diaz-Goebes and Younger concluded that matrix diffusion was the most likely process causing the observed tailing, although it is noted here that this pattern could also be due to (i) a log-normal distribution of vertical permeability across the horizontally extensive RAPS or (ii) progressive dilution of the remaining tracer in the supernatant water, and most likely all three effects play a role.

The objective of this study is to investigate the timescale of chemical transport through an operational RAPS system at Tan y Garn in the UK. Hydraulic tracer tests have been applied to provide information relating to residence time, and also the behaviour of the tracers in the system. The evolution of the system hydraulics over time is investigated. An improved understanding of system hydraulics, residence times, and expected system lifetime will assist mine water remediation engineers to design new RAPS schemes.

THE TAN Y GARN SITE

Tan y Garn Colliery was a small drift mine at Garnswllt, Ammanford, Carmarthenshire, Wales, UK (British National Grid SN 26313 20973). It worked the anthracite of the Ynysarwed Seam (with a typical thickness of 0.76m), part of the Westphalian Upper Coal Measures of the South Wales coalfield. The mine's footprint covers an area of 0.3 km^2. The mine was worked from 1876 and was finally abandoned in 1990 and was allowed to flood to surface. The resulting mine water discharge had a low flow (mean 2 l/s), but a fairly high iron content around 50 mg/l. The receiving watercourse, Afon Cathan, was polluted by the anoxic ferruginous mine water, with the bed of the river being coated in ochre deposits for 1.3 km up to its confluence with the Afon Loughor. Consequently, the discharge was included in the Environment Agency's priority list for remediation of mine water sites. In the UK the Coal Authority is funded by central government to implement a programme of remediation schemes at coal mines abandoned prior to privatisation of the industry in 1994. Around 50 schemes are now operational, 5 of which are alkali-dosed, and, ultimately, the programme will provide treatment for all mine waters on the priority list.

The chemistry of the raw untreated mine water is summarised in Table 1 which shows the arithmetic mean values of data collected from early 2006 to mid 2008. In situ measurements were made using calibrated field instruments. Laboratory results were analysed with procedures approved by United Kingdom Accreditation Service. Metals were analysed using ICP-OES. Ferrous Iron (Fe^{2+}) was analysed by discrete colorimetric method using 1-10 phenanthroline hydrate. Hydrazine reduction followed by the Fe^{2+} method was used to determine total iron. Full details as per McAllan et al. (2009).

Water quality is steady with virtually no change in physico-chemical parameters despite an order of magnitude fluctuation in discharge flow rate (0.5 l/s up to 5 l/s) in response to rainfall. The mine water discharge has a temperature 11.0^{o}C, pH of 6.2, Dissolved Oxygen 1.3 mg/l, Eh -13 mV, the latter two indicating the oxygen depleted, reduced nature of the water. The total iron is 46.6 mg/l, of which 46.2 mg/l is ferrous iron. Sulphate is the dominant anion at 254 mg/l. The elevated iron and sulphate show the importance of pyrite oxidation in the genesis of this mine water. The alkalinity in the mine water (mean 85.2 mg/l as $CaCO_3$ or 1.7 meq/l) is approximately sufficient to buffer the protons released by oxidation and hydrolysis of the ferrous iron (mean 46.2 mg/l as Fe(II) or 1.65 meq/l). Acidity has been measured on both cold, fresh samples in the field, and boiled samples in the laboratory, yielding results for cold acidity and hot acidity respectively. The elevated cold acidity, 316 mg/l as $CaCO_3$, includes contributions from the "mineral acidity" and dissolved carbon dioxide, PCO_2, as discussed by McAllan et al. (2009). The calculated PCO_2 using PHREEQC geochemical model is 10-1.3 atm. which is approximately two orders of magnitude higher than atmospheric PCO_2 of $10^{-3.5}$ atm. The hot acidity mean of 20.1 mg/l as $CaCO_3$ is indicative of a slightly net-acidic mine water. The conventional method to determine if mine water is net-acidic or net-alkaline is to calculate the balance of the field alkalinity, pH and the hydrolysable metals (particularly Fe^{2+}, Mn^{2+}, Al^{3+}) (Hedin et al., 1994; PIRAMID Consortium, 2003; McAllan et al., 2009). The mean calculated net-acidity is 4.4 mg/l as $CaCO_3$, again indicative of a slightly net-acidic mine water.

In designing treatment for the mine water it is recognised that reed bed systems can add alkalinity to the water (Hedin et al., 1994; PIRAMID Consortium, 2003), so slightly net-acidic waters can be treated as per net-alkaline waters, but with additional treatment area providing the necessary additional alkalinity input. However, at Tan y Garn there was only a relatively small area of land available for treatment, so enlarged reed beds were not feasible. Thus, it was evident that alkalinity addition would benefit the treatment process. Added alkalinity raises the pH and increases the iron removal rate, and treatment is achieved with a shorter residence time, and thus smaller areal footprint. At Tan y Garn it was decided to treat the mine water with passive alkalinity addition with a RAPS rather than active chemical dosing.

Table 1. Physico-chemical properties of mine water at Tan y Garn (arithmetic means cited).

	RAW			RAPS outflow				
Parameter, unit	mean	stdev	n	mean	stdev	n	Change	Change as meq/l
Temperature, °C	11.05	1.10	59	11.84	2.87	59	0.79	
pH	6.18	0.22	60	7.01	0.22	57	0.83	
Dissolved Oxygen mg/l	1.29	0.60	53	2.10	1.18	53	0.81	
Electrical Conductivity µS/cm	673.1	32.7	59	841.4	62.9	57	168.3	
Eh mV	-13.3	28.6	55	-92.3	43.3	54	-79.0	
Alkalinity field mg/l as	85.2	15.2	60	229.0	25.4	59	143.8	2.88
Acidity Cold, field mg/l as $CaCO_3$	316.3	99.1	60	109.0	47.4	57	-207.2	-4.14
Flow l/s	2.05	1.38	81	1.89	1.22	81	-0.17	
Sulphate mg/l as SO_4	254.2	13.1	60	227.8	33.0	61	-26.4	-0.55
Chloride mg/l as Cl	12.5	1.4	61	12.2	1.9	57	-0.3	
Calcium mg/l as Ca	42.9	2.2	61	121.8	14.2	59	78.9	3.94
Magnesium mg/l as Mg	29.1	1.6	60	29.2	2.4	59	0.1	
Sodium mg/l as Na	10.3	1.1	60	11.1	2.7	59	0.9	
Potassium mg/l as K	6.4	1.8	61	6.6	1.2	59	0.2	
Manganese mg/l as Mn	3.64	3.36	61	2.90	0.90	57	-0.73	-0.03
Aluminium mg/l as Al	0.13	0.09	59	0.03	0.06	43	-0.10	-0.01
TOC mg/l as C	0.94	0.38	51	2.89	2.46	55	1.95	
Ferrous Iron mg/l as Fe(II)	46.23	2.60	60	7.07	4.14	57	-39.16	-1.40
Total Iron mg/l as Fe	46.60	3.91	60	7.14	3.85	58	-39.45	
Net Alkalinity mg/l as $CaCO_3$	-4.40	15.80	54	213.32	25.90	55	217.72	4.35
Hot Acidity mg/l as $CaCO_3$	20.09	17.65	54	4.00	9.14	48	-16.09	-0.32
Saturation Index Calcite	-1.92			-0.24			1.68	
$\log_{10}(PCO_2)$ atm.	-1.30			-1.67			-0.37	
Iron Loading kg/d	8.27	0.47	42	1.16	1.05	42	-7.11	

The scheme was constructed comprising primary RAPS treatment, followed by three small aeration and settlement ponds and finally a small reed bed. The RAPS media was 80cm deep, and comprised 10cm of compost on top, then 60cm mixed 50:50 by volume compost and limestone gravel, then 10cm of limestone gravel. The compost was municipal compost with no peat. The limestone was local Carboniferous Limestone, this being calcitic, rather than dolomitic, (Cravotta and Ward, 2008) with a maximum clast size of 40mm. The mass of material installed in the RAPS media was not accurately recorded, but it is estimated 180 tonnes limestone were installed. The surface area of the RAPS is around 280 m^2 as an oval shape approx 30m by 10m. Water is distributed across the RAPS top surface by flow through a layer of supernatant water above the compost-limestone media. The inflow pipe is submerged to limit oxygen ingress. The level of the supernatant water is controlled by manually adjusting the level of the outflow pipe. Thus, the water level depends on the head gradient, permeability of the media, and the flow entering the system. Maximum supernatant water depth is controlled by overflow pipes which are about 30cm above the RAPS media. Water is collected in the underdrain, comprising silica gravel and embedded drainage pipes, the layer having a design thickness of 22.5cm to the base of the embedded pipes. The drainage pipes feed the water into the outflow pipe. Since the inflow pipe is at the opposite end to the outflow pipe, water passing through the system travels ~1m downward and ~30m horizontally.

The full scale treatment system was commissioned in January 2006. A layer of ochre, i.e. iron oxidised and precipitated, formed rapidly on top of the compost in the supernatant water layer. Due to the rapidly changing flow rates the water level fluctuated significantly between maintenance visits, scheduled every two weeks. Thus, in dry periods the ochre often became exposed to the air, whilst in wet periods, the ochre was submerged, and the overflows operated. The supernatant water layer appears to be characterised by oxidation and ochre formation. The submerged inlet pipe does not appear to inhibit the oxidation processes. This is considered to be analogous to a recent trial of a vertical flow reactor relying on heterogeneous oxidation of iron. This trial, also in South Wales, treated an unaerated mine water with low D.O. by accretion of iron to a growing ochre bed (Sapsford et al., 2007).

Table 1 shows the chemistry of the treated water, as well as the computed changes for key parameters. The data confirms that the RAPS system is reducing and is alkalinity producing. Reducing conditions are shown by the drop in Eh to strongly negative values, together with sulphate reduction averaging 0.55 meq/l and a strong H_2S smell in the RAPS effluent chamber. Alkalinity production is evident from the increase in calculated net alkalinity of 4.35 meq/l. This increase is due to both the increase in measured alkalinity, 2.88 meq/l, and the removal of iron, 1.40 meq/l. The alkalinity is principally generated by calcite dissolution and is represented by a Ca^{2+} increase of 3.94 meq/l. The approximate balance of the net alkalinity generated, i.e. 4.35 meq/l, is presumed to come from the sulphate reduction 0.55 meq/l. If we assume that the ferrous iron is stoichiometrically removed with sulphate by precipitation of FeS, then 0.55 meq/l of Fe^{2+} are removed in the RAPS media, whilst the remaining 0.85 meq/l Fe^{2+} removed are probably accounted for by precipitation as ochre in the supernatant water. The mean iron removal rate is 24 g/m^2/day which is similar to ranges stated elsewhere (Younger et al., 2002; PIRAMID Consortium, 2003). The removal rate increases with higher loadings, and higher flows, and decreases with lower, limited loadings at times of low flow. Calcite saturation index approaches zero in the effluent. Thus, it appears that even under the very highest flows the limestone can dissolve sufficiently rapidly to reach saturation in the RAPS media. The timescales of the various reactions are poorly known, since the effective porosity of the media is not known.

METHODS

In order to assess the evolution of system hydraulics with time, a series of tracer tests were conducted over a period of 18 months and with a range of flow conditions. The RAPS permeability was estimated based on measurements of the difference in water level across the media, i.e. pond water level minus outflow pipe water level, and the flow rate from the outflow pipe.

The tracer tests involved injecting between 10 and 20kg of sodium chloride into the influent mine water pipe, and breakthrough curve was detected at the outflow pipe with a datalogger recording EC (CTD Diver, Van Essen Instruments). The first tracer test used 0.5 kg lithium bromide which was detected with an autosampler.

RESULTS

Changes in permeability over time

Hydraulic conductivity was simply estimated by dividing the measured flow rate by (i) the head gradient between the supernatant water and the outflow pipe and (ii) then by the effective area of the RAPS. As expected, the permeability has decreased with time, as per Figure 1. The earliest data are not reliable since the difference in water levels was so small. Thus, the starting value at commissioning was in excess of 5 m/d, after 1 year it was around 1 m/d, after 2 years 0.5 m/d, falling then to around 0.2 m/d.

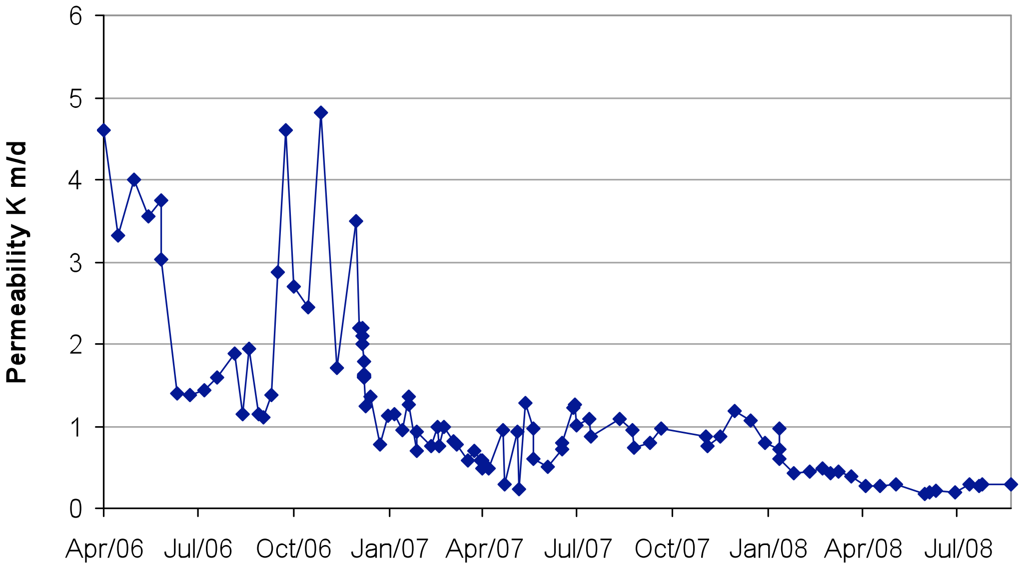

Figure 1. Estimated change in vertical hydraulic conductivity with time for RAPS.

Hydraulic tracer tests

Ten tests were conducted between December 2006 and June 2008, see Table 2 and Taylor (2008). Figure 2 illustrates the typical results observed, the data starting at the time of injection. The influent background EC is relatively steady. The small downward spikes were due to rainfall runoff running directly into the injection chamber and the RAPS, although this is not considered to have any significant effect overall. The effluent breakthrough curve has the expected shape of a steep rise to peak, followed by a long tail. In most of the tests the background outflow EC had changed through the test, so accurate mass recovery calculations were not possible, although approximations suggested generally greater than 80% was recovered. In assessing the results it is here assumed that the peak time, Tp, represents the dominant advection process, and further this is considered here to reflect the residence time in the whole RAPS system, including supernatant water, porous media, and underdrain. It is recognised that whilst the tracer may be affected by matrix diffusion, the background minewater may be affected less since the concentration gradient between flowing and stagnant pores approaches equilibrium over time.

Table 2. Summary of tracer tests

Test No.	Date	Tracer	Flow (l s^{-1})	Tp (hours)
1	Dec-06	LiBr	4.7	5
2	Jan-07	NaCl	4	6.25
3	Apr-07	NaCl	1.53	16.2
4	May-07	NaCl	1.5	21.3
5	Jul-07	NaCl	1.64	18.25
6	Aug-07	NaCl	1.62	21
7	Jan-08	NaCl	3	10
8	Mar-08	NaCl	1.86	14.75
9	Jun-08	NaCl	1.02	28.5
10	Jun-08	NaCl	0.9	22.83

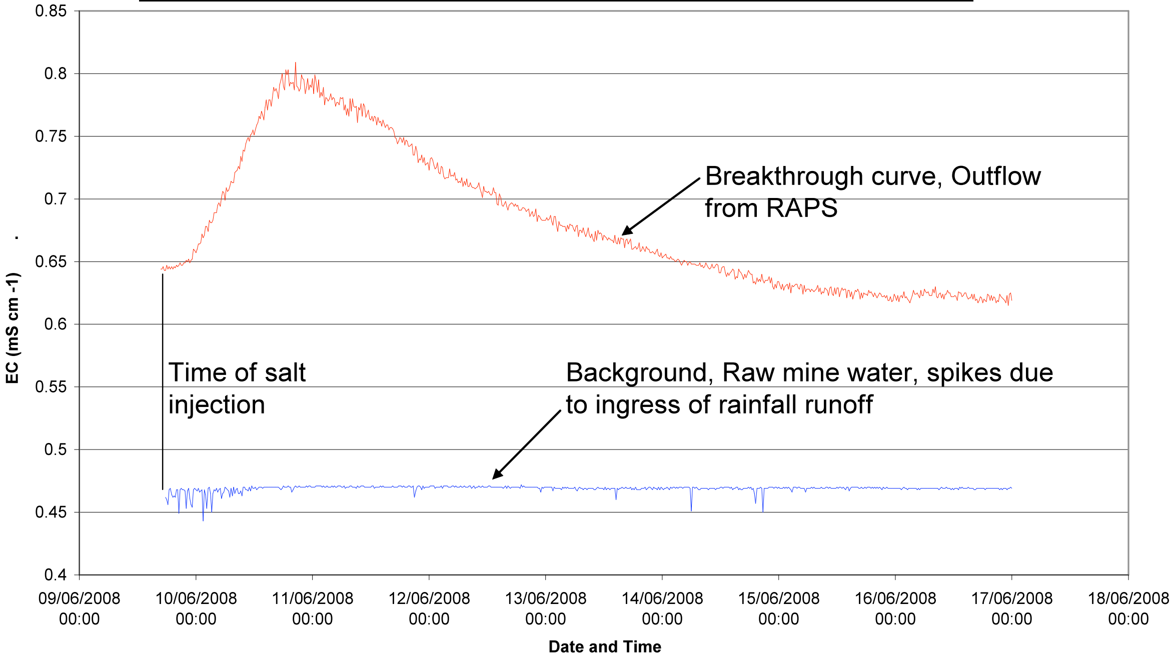

Figure 2. NaCl tracer test no 9, injection on 9th June 2008

Figure 3 shows a good correlation between the flow and inverse Tp such that the gradient, 92 m^3, represents the bulk effective volume of the whole system, averaged across all the tests. The calculated bulk effective volume for each test did not correlate well with the changing supernatant water levels. Neither was there any apparent trend of decreasing bulk effective volumes with the decreasing permeability.

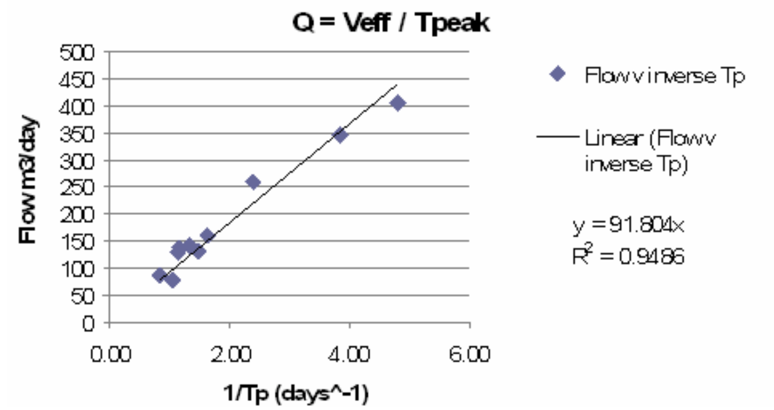

Figure 3. Flow versus inverse peak time for all salt tracer tests

Comparing the measured bulk effective volume of 92 m^3, with the overall system volume of 310m^3 which includes an assumed 20cm supernatant water depth, the bulk effective porosity is 30%. Note that this is not necessarily the same as the effective porosity in the reactive media. Since the supernatant water and the underdrain are designed to be highly porous, i.e. mainly filled with water, it is likely that these layers contribute strongly to the bulk effective volume. Thus, 30% is likely to represent the upper limit of the effective porosity of the reactive media. The bulk residence time, here assumed as Tp is the sum of the time spent in each of the layers, and the longest amount of time is spent in the layer with the highest effective volume. Although the portion of time spent in the reactive media is of interest to calculate reaction rates, this cannot be quantitatively derived from the tracer tests, as these measure the whole system, and not just the reactive media. However, if we assume the supernatant water layer is 20cm deep with an effective porosity of 80% to account for it being partly filled with ochre precipitate, its effective volume is 45m^3; then the underdrain with an assumed 30% effective porosity, accounting for some clogging with fines washed down from above, has an effective volume of 15m^3. The estimated effective volume of the reactive media layer is then about 32 m^3 (i.e. 92 – 45 – 15), or about one third of the bulk effective volume, and this gives a “minimum estimate” of the effective porosity of the reactive media of 16%. The residence time spent in each layer is proportional to the layer’s fraction of the bulk effective volume, so the residence time in the reactive media is about one third of the bulk system residence time, Tp, if we accept the “minimum estimate”. Under mean flow conditions of 2 l/s, the bulk effective volume of 92m^3 gives a predicted system residence time of 12.8 hours, whilst the estimated residence time in the reactive media is only 4.4 hours. Peak flows regularly exceed 4 l/s implying less than 2.2 hours residence time in the reactive media.

The dissolution of limestone in the reactive media, which appears to reach saturation at all observed flow rates, must be occurring relatively rapidly. Younger et al. (2002) expect a RAPS with 14 hour residence time to produce 150 to 300 mg/l as $CaCO_3$ net alkalinity. At Tan y Garn 218 mg/l as $CaCO_3$ net alkalinity is produced in as little as 2 hours, given the above assumptions.

It is also noted that the above estimates suggest a significant proportion of the residence time, perhaps as much as half, is spent in the supernatant layer, and that this is the zone where most of the iron is removed as ochre, probably by heterogeneous oxidation (Sapsford et al 2007).

Since, the effective volume appears relatively constant, independent of water levels, and does not decline with permeability, the treatment capacity of the RAPS looks set to continue to perform at present levels, which is in line with existing design guidelines and expected performance. At present dissolution rates the limestone will be exhausted in a total of 15 years. The compost was chosen to provide a long lasting carbon source, hopefully avoiding this becoming a limiting factor (Bhattacharya et al, 2008), although the mass balance suggests that sulphate reduction is not the most important process in the RAPS.

The main barrier to continued operation is the continued decline in permeability, and associated increasing overflows. At Tan y Garn it is fortunate that the water is only slightly net acidic, and thus quite large untreated overflows can be accommodated. It is not certain whether the decrease in permeability is mainly attributed to the surface ochre deposits (Sapsford et al 2007), or changes within the media or underdrain. Surface de-sludging will be necessary soon, and re-measuring permeability and tracer testing afterward will help quantify this.

CONCLUSIONS

Routine monitoring at Tan y Garn RAPS has allowed the performance of the system as a whole to be assessed. The application of a series of tracer tests provides information on the system hydraulics including residence time and bulk effective volume. Further with some assumptions, the residence time may be estimated within the supernatant, ochre producing layer, and within the reactive media. Estimates of bulk porosity of the reactive medium vary from a minimum of 16% to a maximum of 30%. If we accept the minimum estimate, most time is spent in the supernatant layer, where most iron is removed. The time spent in the reactive media can be surprisingly short, perhaps 2 hours under high flows, yet the treated discharge still approaches calcite saturation. Tan y Garn works as expected, having comparable areal iron removal rates and alkalinity production rates to published design guidelines. The system lifetime is likely to be limited by permeability, although there is a chance that permeability can be improved by desludging the surface.

The reason for the apparent fast reactions is unknown, so although in this specific case study the design criteria may have been highly conservative, this may be an exception. Therefore, current published design guidelines appear fit for purpose, and the UK Coal Authority will continue to consider new RAPS as a passive alternative to conventional active treatment.

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