Performance review of an operational Reducing Alkalinity Producing System (RAPS) treating coal mine waters at Tan-y-Garn, Wales

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Abstract
The Coal Authority’s passive coal mine water treatment scheme (MWTS) at Tan-y-Garn in Wales incorporates a Reducing Alkalinity Producing System (RAPS). A long-term performance review has been undertaken after 10 years of continuous operation without any major maintenance. Over the life of the MWTS, raw mine water quality has gradually improved due to flushing. Tan-y-Garn RAPS continues to generate alkalinity and raise mine water pH but the alkalinity generation efficiency has declined over time. Alkalinity generation within the RAPS is predominantly due to limestone dissolution. Sulfate-reducing bacteria are present in the RAPS but net redox transformations of sulfur are interpreted to be of minor significance to the current mine water treatment process. The RAPS removes a significant proportion of iron from the mine water. Short-term variations in iron removal efficiency reflect variations in flow rate and iron loading. Longer-term decline in iron removal efficiency is also occurring despite decreasing iron concentrations in the raw mine water. The performance of the Tan-y-Garn RAPS has declined over the past decade but the system continues to achieve the desired treatment goals. RAPS technology is considered a viable option for the long-term passive treatment of coal mine waters.

Key words: mine water, passive treatment, coal, alkalinity, iron

Introduction
The Coal Authority’s coal mine water treatment scheme (MWTS) at Tan-y-Garn in Wales, commissioned in 2006, incorporates a Reducing Alkalinity Producing System (RAPS). A performance review of the long-term water treatment efficiency of the Tan-y-Garn RAPS has been undertaken after 10 years of continuous operation.

Construction and Operation of Tan-y-Garn RAPS
Tan-y-Garn Colliery was a small drift mine at Garnswllt, Ammanford, Carmarthenshire, Wales, UK. The colliery worked the Ynysarwed seam, part of the Westphalian Upper Coal Measures of the South Wales coalfield. The mine was worked from 1876, abandoned in 1990 and allowed to flood to surface. This resulted in pollution of the nearby watercourse, Afon Cathan. A passive MWTS was constructed by the Coal Authority at Tan-y-Garn to remediate the mine water pollution in the river. The MWTS, commissioned in January 2006, comprises primary RAPS treatment, followed by three small settlement lagoons and a small constructed wetland. The overall size of the MWTS was constrained by the availability of suitable land. The treated water is discharged to the nearby watercourse.

The RAPS has a surface area of around 280 m² and is oval in plan view (approximately 30 m by 10 m). The RAPS is designed to produce alkalinity through the dissolution of limestone and by sulfate reduction within the organic materials (Hedin et al. 1994). Anaerobic conditions created by microbial activity in the compost in the RAPS help prevent blinding of the limestone surface by iron precipitation.

The RAPS at Tan-y-Garn MWTS comprises:

- Up to 300 mm maximum depth of supernatant water, with depth controlled by overflow pipes;
- 100 mm thick basal layer of limestone gravel;
- 600 mm thick layer of mixed compost and limestone gravel (mixed 50:50 by volume);
- 100 mm cover layer of compost;
- 225 mm thick underdrainage system, comprising pipes surrounded by silica gravel.

The compost used in the RAPS system met the requirements of the Specification for Composted Materials BSI PAS100:2005. The compost was provided by Swansea City Waste and was classified as a Civic Amenity Green waste. The compost was specified to have a minimum pH of 6 and not to be derived from peat. The limestone was local calcitic Carboniferous Limestone with a maximum clast size of 40 mm. Although the mass of material installed in the RAPS was not accurately recorded, it is estimated that around 180 tonnes of limestone were installed. The compost-limestone mix was designed, building on previous research by Amos & Younger (2003) and Bowden et al. (2005) to optimise the residence time within the RAPS for alkalinity generation and to maintain sufficient hydraulic conductivity to maintain throughflow. No slurry or similar materials were used in order to limit early performance spikes from enhanced sulfate reduction, and also to limit the chance of elevated DOC or nitrogenous concentrations affecting the receiving watercourse. Silica gravel was used in the underdrainage system to help retain structure and hydraulic conductivity, unlike the sacrificial limestone gravel within the reactive media.

Water monitoring has been undertaken at Tan-y-Garn since prior to MWTS commissioning. Water quality monitoring was undertaken ~twice monthly for the first 5 years and then ~monthly thereafter. In situ measurements have been undertaken using calibrated field instruments. Laboratory analysis has been undertaken in accordance with procedures approved by the UK Accreditation Service.

Minimal maintenance activities have been undertaken at Tan-y-Garn MWTS since construction. Near-surface hydrous ferric oxide (HFO) sludge has been removed occasionally but the RAPS compost has not been replaced. Self-seeded vegetation is now growing on top of the RAPS (Fig. 1).

Figure 1 Photograph of Tan-y-Garn RAPS; note accumulation of HFO at the surface and vegetation growth
There is not continuous flow monitoring at Tan-y-Garn MWTS but spot readings of flow rates have been obtained between 2006 and 2015 using thin-plate weirs. Mine water flow rates at Tan-y-Garn vary seasonally and in response to significant rainfall events; typical flow rates are in the range 1-2 L/s with peak flows up to 6 L/s recorded. During high flow events, a proportion of the mine water flow bypasses the RAPS with the overflow re-entering the treatment scheme further downstream in the treatment system, where it mixes with the water that has flowed through the RAPS.

Tracer testing at Tan-y-Garn MWTS by Watson et al. (2009) allowed estimation of a 4.4 hour residence time in the reactive media of the RAPS, dropping to less than 2.2 hours residence time during peak flow conditions. These residence times are considerably lower than the minimum residence time of 15 hours within the limestone layer recommended by Watzlaf et al. (2004). However, work by others has also confirmed that alkalinity can be generated more rapidly than suggested by original design guidance; e.g. Trumm et al. (2008) reported that an 8-hour residence time was adequate for a pilot RAPS. Taylor et al. (2016) reported that residence times at Tan-y-Garn RAPS decrease proportionally to increasing flows. The hydraulic conductivity of the Tan-y-Garn RAPS is interpreted to have reduced over time due to compaction and biogeochemical activity (Watson et al. 2009).

Changes in Raw Mine Water Quality at Tan-y-Garn

Raw mine water quality at Tan-y-Garn is gradually improving due to flushing of the shallow mine workings by rainfall. Net acidity has declined over time and the mine water is now only marginally net acidic. The annual average (and minimum) pH of the raw mine water has increased from 5.9 (5.5) in 2006 to 6.5 (6) in 2015. Since MWTS construction, annual average concentrations of total iron have decreased from 47 to 36 mg/L. Similarly, total concentrations of manganese, nickel and zinc have gradually decreased in the raw mine water since 2006. The long-term performance of the Tan-y-Garn RAPS needs to be considered in the context of improving raw mine water quality.

Alkalinity Production within Tan-y-Garn RAPS

Within the RAPS, there is a net gain in alkalinity and mine water pH increases to around 7. The magnitudes of the pH rise and net gain in alkalinity within the RAPS have gradually declined over time (Fig. 2). Despite the decline in RAPS alkalinity generation efficiency over the past decade, the RAPS performance remains sufficient to neutralise the net acidity of the raw mine waters.

Alkalinity generation within the Tan-y-Garn RAPS is predominantly due to limestone dissolution. Alkalinity production due to limestone dissolution is assumed to be proportionally related to the measured increase in calcium concentrations; a 1 mg/L increase in calcium concentrations is assumed to be equivalent to 2.497 mg/L of alkalinity as CaCO₃ (Watzlaf et al. 2000). Calcium concentrations increase within the RAPS by around 70 mg/L, equivalent to around 175 mg/L of alkalinity as CaCO₃ (Fig. 3). Dissolution of the limestone is interpreted to be occurring at around 7842 kg/annum, based on the average flow rate of 1.42 L/s. The limestone within the RAPS is thus predicted to last around 23 years (e.g. for another 13 years), which is broadly in line with the 25-year design life of the MWTS.

Alkalinity can also be generated within RAPS by microbial sulfate reduction. The microbial community present at Tan-y-Garn MWTS includes bacteria and archaea involved in iron and sulfate cycling, including sulfate-reducing bacteria (Falagan et al. 2016). Sulfate concentrations have decreased within the RAPS on average by 6%, suggesting that some sulfate reduction is occurring. A 1 mg/L decrease in sulfate concentrations is assumed to be equivalent to 1.042 mg/L alkalinity as CaCO₃ (Watzlaf et al. 2000). Equivalent alkalinity generation due to sulfate reduction within Tan-y-Garn RAPS has gradually declined over time (Fig. 4). Net redox transformations of sulfur are interpreted to be of minor significance to the current mine water treatment process.
Figure 2 long-term variations in RAPS performance in relation to pH and total alkalinity (as CaCO$_3$).

Figure 3 Calculated equivalent alkalinity generation due to limestone dissolution based on change in calcium concentrations within the RAPS.
Alkalinity generation patterns observed at Tan-y-Garn MWTS are consistent with those reported by Watzlaf et al. (2000), with alkalinity production due to sulfate reduction declining as the RAPS ages whereas alkalinity production due to limestone dissolution continues. The decrease in hydraulic efficiency previously reported by Watson et al. (2009) is interpreted to explain the observed gradual decline in overall alkalinity generation efficiency.

Mesocosm experiments by Bangor University for the Coal Authority (Falagan et al. 2016) showed that providing an additional bioavailable source of organic carbon (glucose) stimulated bacterial sulfate reduction in incubating compost and HFO samples from Tan-y-Garn RAPS under anaerobic conditions. Therefore, if RAPS performance were to require boosting in future, providing an additional source of bioavailable carbon could stimulate additional alkalinity generation by microbial sulfate reduction. However, the longevity of such stimulus would need to be assessed and the cost-benefits of potential multiple stimuli compared with regeneration of compost/limestone media.

**Metal Removal within Tan-y-Garn RAPS**

Although RAPS are designed primarily to generate alkalinity and raise pH, the Tan-y-Garn RAPS also removes a significant proportion of the total iron from the mine water (Fig. 5). Iron remains predominantly (93%) in the ferrous state at the RAPS outlet, confirming that reducing conditions are maintained inside the RAPS. Mesocosm experiments by Falagan et al. (2016) showed that ferrous iron concentrations in Tan-y-Garn compost samples increased on incubation under anaerobic conditions, whether or not additional carbon (glucose) was provided.

Iron removal efficiency within the RAPS is temporally variable. Short-term variations in iron removal efficiency correlate with fluctuations in flow rates and thus iron loading (Fig. 6). Long-term decline in iron removal efficiency has also been observed. Despite the decline in influent concentrations, total iron concentrations at the outlet of the RAPS have gradually increased (Fig. 5). This longer-term trend of declining iron removal efficiency is interpreted to reflect the decline in microbial sulfate reduction as well as decreasing availability of sorption capacity.
Figure 5: Long-term variation in total iron concentrations at Tan-y-Garn MWTS

Figure 6: Variations in iron removal rates and flow rates at Tan-y-Garn MWTS RAPS
Iron oxidation is occurring near the surface of the RAPS lagoon. HFO build up on the surface of the RAPS has an adverse influence on lagoon hydraulics (Watson et al. 2009). Similar issues have been reported at other RAPS (e.g. Skousen & Ziemkiewicz 2005). Incorporating a settlement lagoon before the RAPS, as was suggested by Watzlaf et al. (2000), might have reduced HFO build up on the top of the RAPS. This might have benefited the long-term hydraulic performance of the RAPS. Incorporating a pre-RAPS settlement lagoon may also help manage short-duration fluctuations in flows following rainfall events, potentially decreasing the proportion of flows bypassing the RAPS.

In addition to removing iron from the mine water, the RAPS also reduces concentrations of other metals. A significant proportion of the aluminium is removed within the RAPS. The average total aluminium concentration at the RAPS outflow is 0.03 mg/L compared with 0.12 mg/L in the raw mine water. Manganese concentrations at the RAPS outflow are 7% lower on average than in the raw mine water. As was observed for total iron, total manganese concentrations at the RAPS outflow exhibit greater variability than is observed in the raw mine waters. RAPS are not usually expected to be effective in removing manganese (Kepler & Mc' Cleary 1994). Zinc and nickel concentrations are also lower (typically below the reporting limits of the analytical method used, <0.01 mg/L) at the RAPS outflow than in the raw mine water, where average concentrations of zinc and nickel are 0.02 mg/L and 0.03 mg/L respectively. It is considered likely that manganese, nickel and zinc are predominantly removed through sorption or co-precipitation with iron, although zinc and nickel may also precipitate as sulfides within the RAPS.

Conclusions

The RAPS system at Tan-y-Garn MWTS has been operating for 10 years with relatively little maintenance. The RAPS remains functional although performance has gradually declined over time. Alkalinity generation within the RAPS is primarily due to limestone dissolution. Sulfate reduction is currently of negligible importance. The RAPS also removes a significant proportion of iron and other metals. Due to the gradual improvement in raw mine water quality due to flushing, it is possible that it will not be necessary to replace the current RAPS once it has reached the end of its operational life. Traditional passive treatment using settlement lagoons and constructed wetlands may ultimately be sufficient. RAPS technology has been demonstrated to be a viable option for the long-term passive treatment of coal mine waters. The Coal Authority will continue to consider new RAPS as a passive alternative to chemical dosing.

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References


