Performance of a Vertical Flow Reactor Treating Acid Mine Drainage of an Abandoned Coal Mine near Carolina, Mpumalanga, South Africa

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ABSTRACT

Mine water pollution in South Africa is known to originate mainly from abandoned and operating coal and gold mines, primarily as acid mine drainage (AMD). Two types of treatment are commonly used to treat mine water: active treatment and passive treatment. Active treatment requires high maintenance for aeration, power generation for pumping and the addition of chemicals to treat the water. Passive treatment methods are typically relatively inexpensive to operate, do not require the addition of chemicals for treatment, and are usually low maintenance systems. An experimental passive treatment Vertical Flow Reactor (VFR) has been installed at an abandoned coal mine site near Carolina, Mpumalanga, South Africa (Fanie Nel Discharge). The site is located approximately 8.6 km from the Boesmanspruit reservoir, which was previously believed to be contaminated by coal mine water discharges from the surrounding area. A VFR was installed with the objective of remediating the acidic mine water and investigating its ability to operate under relatively low pH conditions. This was achieved by utilising an intermediate bulk container (IBC) and a gravel bed, with raw mine water flowing vertically through the container by gravity. The objective was to remove Fe (≈ 230 mg/L) by an autocatalytic process with ochre precipitation, to aerate the water, and to study the chemistry of the mine water, which has an electrical conductivity of 2.1 mS/cm. The pH of the discharged mine water is approximately 3, and the redox potential approximately 560 mV, indicating that ferrous iron predominates. Consequently, the anticipated Fe removal rate of 65–75% was not achieved, and the mine water could not be treated as intended. A second VRF was installed, and the influent water to this VRF was treated by an additional open limestone drain. This raised the pH by buffering and increased the Fe removal rate from 35% to 69%. This result was unexpected when compared to the Cwm Rheidol VRF in Wales, which treated mining-influenced water (MIW) with a similar chemical composition. Nevertheless, the results demonstrate the necessity of either the presence of crystallisation nuclei of iron oxyhydrates in low pH mine water or, alternatively, higher pH values that support the formation of these crystallisation nuclei for a VFR to function effectively. In the case of the Fanie Nel Discharge, neither of these conditions were present, resulting in the trial's outcomes falling short of initial expectations. Consequently, a full-scale VFR was not installed at the site.

Key Words: acidic mine drainage, passive treatment, South Africa, vertical flow reactor (VFR)

INTRODUCTION

The treatment of mine water at remote or abandoned mine sites is frequently constrained by the lack of reliable energy or the supply of chemicals. This limits the applicability of active mine water treatment technologies, which require electricity, chemicals, and quasi-continuous maintenance. While the electricity demand of active plants could be met by using renewable energy sources, the substitution of chemicals and workforce is not a straightforward process. With the growing awareness of the need to protect watercourses from contaminated and acid mining influenced water (MIW), the demand for low-maintenance water treatment solutions emerged approximately four decades ago in Northern America and the United Kingdom (Kleinmann et al. 2021; Younger et al. 2002). The first passive water treatment plant, which also treated mine water, was constructed in Germany (Brix, 1987; Wolkersdorfer, 2022). This technology was rapidly disseminated globally, and it is currently employed in approximately two dozen countries.

For the treatment of net alkaline or circum-neutral ferruginous MIW, numerous passive treatment technologies are available, while for net acidic, low pH MIW, passive treatment is still challenging or even not possible. Among the various passive MIW treatment technologies available, the vertical flow reactor (VFR) is one of the simpler ones. It employs the long-known process of autocatalysis, precipitation and co-precipitation of iron oxyhydrates, also referred to as ochre accretion (Barnes, 2008; Florence et al. 2016; Hayton et al. 2022; Sapsford et al. 2006; Sapsford and Williams, 2009). Although these processes frequently result in adverse outcomes, such as pipe or technical installation clogging, when employed as a passive MIW treatment method, they have demonstrated promising results in both small-scale and full-scale installations, particularly in net alkaline MIW (Blanco et al. 2018). At the former Cwm Rheidol Pb-Zn mine site in Wales (Jones, 1922), researchers have previously attempted various passive treatment technologies (Edwards and Potter, 2007; Jones and Howells, 1975), including a small-scale, intermediate-bulk container (IBC) based VFR to treat the MIW from the lower #9 adit (Florence et al. 2015, 2016). However, the MIW at this site is net acidic, with a pH of ≈ 2.8 and unfiltered Fe_{tot} concentrations of approximately 95 mg/L.

A comparable water chemistry is observed at a MIW discharge location in Carolina, Mpumalanga, South Africa. There, the pH is also ≈ 3.0, and Fe_{tot} concentrations are twice as high at ≈ 230 mg/L, indicating that the MIW is acid mine drainage (AMD). To ascertain whether a VFR might also be a solution for this discharge, called Fanie Nel Discharge (FND, the Council of Geoscience commonly uses the name Witkranz Discharge), a 1 m^3 IBC was installed and constructed in accordance with the methodology described in Wolkersdorfer (2022). The trial was conducted for a period of 22 weeks, during which the flow rate and pre-treatment were adjusted in order to evaluate various pre-treatment options and VFR set-ups. Due to substantial vandalism, the trial had to be abandoned after improvement and modifications following November 2015.

At the time of writing this paper, the Council for Geoscience operates a RAPS (reducing and alkalinity producing) system (Dube et al. 2021), which is based on the studies published by Mashalane et al. (2018) and the field trial of Novhe et al. (2016).

MATERIALS AND METHODS

Study Site Description

The area discussed in this paper falls within the geological realm of the Ermelo Carboniferous. This geological epoch is characterised by its rich coal deposits, prominently displayed within the Vryheid Formation of the Ecca Group, a major unit within the Karoo Supergroup. Within this geological tapestry, coal seams emerge as notable features within the Vryheid Formation. While the Vryheid Formation manifests itself predominantly as sandstone, it also hosts subordinate layers of shale, contributing to its geological diversity and complexity (Snyman, 1998). The Discharge location itself is located on the farm Witkranz 53 IT, Portion 11, 12 km SSE of Carolina (Figure 1), where the AMD discharges 200 to 300 m West of an abandoned underground coal mine at an altitude of 1690 masl. Additional site information or information about the mine’s history were not available, but an illegal open pit coal mine meanwhile opened at the location of the abandoned coal mine.

A characteristic of the discharge site is the optically clear water, which most likely flows out from a fracture in the rock. To the south, for a distance of about 140 m, several seeps with the same low pH as the main discharge appear and are collected in a type of gully which discharges westwards into a 1.3 km long rivulet and eventually flows into the Boesmanspruit river at an altitude of 1630 masl. A small shaft of unknown depth and history is located a few metres north of the site. At the time of the field trial, the AMD was to be treated with lime from an inoperative self-dosing device and past “treatment” flowed into a 900 m^2 settling pond, after which the Fe_{tot} concentration decreased by about 50%. From there, the AMD flowed down a slope into a 550 m^2 polishing pond divided by lime bags meant to raise the pH. These lime bags were observed to rapidly fuse together, allowing the AMD to overflow the barriers and render them ineffective. However, most of the water from the settling pond bypassed the polishing pond. All of the AMD ended up in said rivulet and in the Boesmanspruit. As a result of the hydrolysis of the iron in the AMD, the entire site was stained with iron precipitates (Figure 2), and the pH dropped by 0.4 from the time of discharge into the said rivulet.

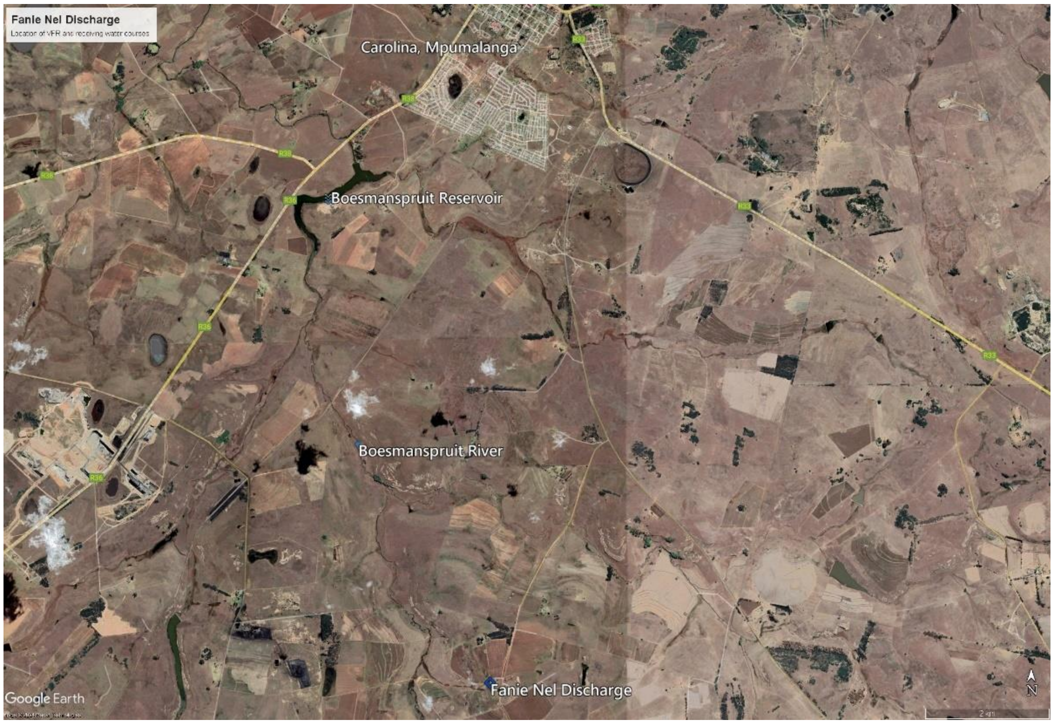

Figure 1. Location of the Fanie Nel Discharge in Mpumalanga, South Africa (Google Earth, satellite imagery 2018-06-18)

Figure 2. Overview of the discharge location (pipe below the car) with VFR on the leftmost side. The other three IBCs were used for a field trial of the Council for Geoscience. In the foreground the settling pond (photo taken on 2015-06-03).

Methods

Prior to filling the 1 m³ VFR with the substrate (slag from vanadium processing used as gravel material widely in South Africa), this material was subjected to laboratory testing for a period of 10 weeks. An aliquot of the substrate was transformed into four beakers in the laboratory, of which two were filled with distilled water and two with the FND mining-influenced water. Electrical conductivity and pH were measured on a regular basis, and at the conclusion of the test, the chemical composition of the water in the two beakers was subjected to chemical analysis. In addition, 12 pieces of the slag substrate were selected at random and analysed by XRF (Table 1).

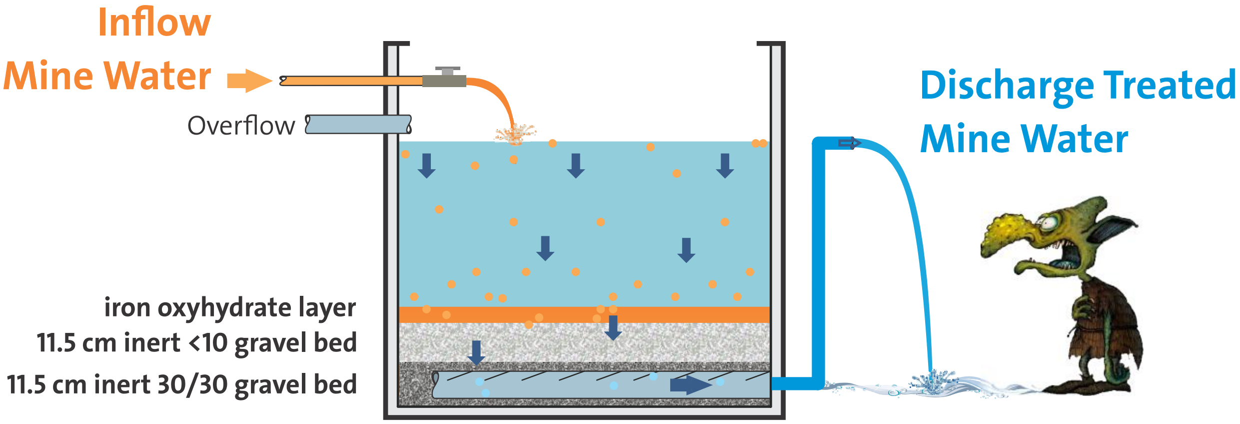

Figure 3. Scheme of a vertical flow reactor with inflow from the left and outflow at the right side. From Wolkersdorfer (2022), © Stollentroll Walter Moers.

Figure 4. Setup of the VFR in a 1 m³ IBC. From left to right: spiral-laid vacuum hose, 15 cm high slag layer, filling of the VFR with AMD, field trial VFR in operation

For the treatment of the AMD in the VFR (Figure 3), an aliquot of 0.49 L/min of the mine water discharge was diverted into a 1 m³ IBC. The VFR was filled with a 15 cm thick layer of 2–2.5 cm slag material above a spiral-laid 10.8 m long, perforated swimming pool vacuum hose (diameter 32 mm). A gooseneck hose was installed to adjust the hight of the AMD above the vacuum hoses. Subsequently, the fine material of the substrate was flushed with the AMD, after which the IBC was filled with AMD to a height of 60 to 65 cm, equalling 600 to 650 L (Figure 4) and resulting in a nominal residence time of 20–22 h.

On-site parameters (temperature, electrical conductivity, pH, redox, O_2, HACH HQ40d) were monitored monthly over a five-months period. Field measurements also included the flow rate (volumetric method, 2 L beaker, stopwatch), field iron concentrations (Fe_{tot}, Fe^{2+} filtered and unfiltered, HACH colorimetric method, DR900) as well as acidity and alkalinity (field titration, cold method, HACH digital titrator).

Additional filtered and unfiltered samples were analysed in the laboratory for Al, Mn, SO_4, SiO_2, NO_2, and NO_3 (HACH colorimetric method, DR900).

Table 1. XRF results of 12 slag samples used in the leaching experiment in the laboratory. $\overline{x}$: arithmetic average, *s*: standard deviation of the sample, *s*, % sample standard deviation in %.

Element	**7-2**	**7-1**	**6-3**	**6-2**	**6-1**	**4-1E**	**3-3**	**3-2**	**3-1**	**2-3**	**2-2**	**2-1**	$\overline{x}$	***s***	***s*, %**
Ca, %	5.5	5.63	6.47	4.37	6.71		5.7	4.93	5.66	7.68	6.01	4.91	5.78	0.93	16
Cr, %	6.14	6.37	6.48	8.58	7.69	8.67	6.11	5.88	7.09	3.8	7.58	9.15	6.96	1.49	21
Fe, %	1.95	2.2	2.32	4.69	3.21	2.67	2.24	2.33	2.52	1.7	2.67	3.2	2.64	0.78	30
Zr, ppm	152	115	146	118	142	119	133	134	126	121	131	105	129	14	11
Zn, ppm	128	128	184	119	147	101	124	134	155	126	142	307	150	54	36
Ti, ppm	3768	3969	2297	4719	49261	2062	3962	3002	4316	3381	3811	3785	7361	13.22	180
Sr, ppm	107	136	143	123	114	93	154	109	132	183	104	89	124	27	22
S, ppm	4063	7783	3364	0.0162	0.0134		8626	3468	4534	0.0217	0.0146	4654	3317	3.10	93
Rb, ppm	13.1	22.3	15.5	5.6	13.3	15.6	22.2	11.2	16.7	18.3	10.1	7.3	14.3	5.3	37
Ni, ppm	130	141	135	244	183	152	128	162	147	103	148	172	154	35	23
Mn, ppm	2248	2242	2297	2812	2743	2540	2320	1942	2567	2126	2689	2789	2443	286	12
K, ppm	5238	8892	6390	4791	6518		8441	5323	6062	6202	5066	4528	6132	1.42	23
Hg, ppm	11	11					10			10	8		10	1.22	12
Se, ppm		3			4.7		4	3	2.9	3.1	3.7		3.5	0.7	19
Ba, ppm		2000	1745	2957	1823	3572	1885	2330		1706	2457	3068	2354	649	28

RESULTS AND DISCUSSION

Based on the XRF analysis, the slag contains higher concentrations of Cr, Fe, Ti, S, and the pieces exhibit a high degree of variability in all parameters, as evidenced by the high relative standard deviation (*s*%). It was demonstrated that the material is inert and does not release additional potential contaminants into the AMD, as evidenced by the $\Delta R_{FND}/\overline{x}$ and RDW results (Tab. 2). Average pH in the mine water beakers increased from 3 to 4 and the average electrical conductivity stayed constant at around 2040 µS/cm. Consequently, it was determined that the slag material could be utilized for the FND VFR without adding contaminants into the AMD.

During the period of the field trial, the average flow rate at the FND was relatively constant at 25.5 L/min with a minimum of 22.7 L/min and a maximum of 28.5 L/min ($n = 9$). pH ranged from 2.3 to 3.2 (arithmetic mean 3.0, $n = 13$), temperature from 14.4 to 19.8 °C (mean 14.8 °C), redox (SHE) from 540 to 510 mV (mean 550 mV), electrical conductivity from 2001 to 2183 µS/cm (mean 2097 µS/cm) and O_2-saturation from a low 1.2 to 4.3 % (mean 2.4 %). Filtered Al concentrations are around 29 mg/L, As below detection limit, SO_4 around 1220 mg/L and Mn around 63 mg/L. Unfiltered Fe_{tot} ranged from 208 to 281 mg/L (mean 242 mg/L) with an average of 60% ferrous iron and filtered Fe_{tot} ranged from 227 to 274 mg/L (mean 245 mg/L) with an average of 64% ferrous iron. The iron data indicates that the iron at the discharge is predominantly dissolved and in reducing conditions, which explains the optically good quality of the AMD. In addition to the above constituents, the total REEs (rare earth element) concentrations reached 1.3 mg/L. In an on-site aeration experiment, it could be shown that the AMD can be aerated to a 90% O_2 saturation after 14 min, whilst the pH stayed stable around 3.1.

The parameters analysed revealed that only Fe, Al and Mn exhibited notable removal rates (Figure 5). Removal rates of sulfate were inconclusive, and the electrical conductivity decreased by approximately 3%, which is consistent with the removal of Fe and Mn. During the trial, the average pH remained constant at 3.0 and did not exhibit a substantial increase for individual sampling dates (≈ 3%). Oxygen saturation increased from an average of 2% to 34%, and the redox potential (SHE) increased from 550 mV to 630 mV. Although alkalinity ($k_{B4.3}$) and acidity ($k_{A8.2}$) were not anticipated to undergo substantial changes, both parameters exhibited slight alterations from the VFR inflow to the outflow. Alkalinity increased from an average of 1.51 mmol/L to 2.35 mmol/L, while acidity decreased from 14.91 mmol/L to 13.12 mmol/L. Nevertheless, these parameter changes were insufficient to enhance the reaction rate of iron precipitation, resulting in the VFR consistently underperforming in comparison to the initial expectations.

Table 2. Chemical analysis results of the discharged AMD before (FND) and after (VFR) the inflow through the VFR. R represents the water after the end of the leaching experiment with FND for the discharged water and DW for distilled water. $\Delta R_{FND}/\bar{x}$ gives the increase or decrease of the below element after the leaching experiment.

Sample	**FND**	**VFR**	**$\bar{x}$**	**R_{FND}**	**R_{DW}**	**$\Delta R_{FND}/\bar{x}$**
Sample Date	2015-06-05	2016-03-10				
pH, –	3.07	3.22	3.1	3.02	6.69	–
Al, mg/L	31	25	28	38	0.132	10
Ca, mg/L	162	148	155	171	3	-7
Co, mg/L	1.59	1.20	1.395	1.27	<0.010	-0.12
Cr, mg/L	0.014	0.010	0.012	2.14	<0.010	2.13
Fe, mg/L	265	196	230.5	77	0.297	-154
K, mg/L	7.2	9.9	8.55	8.95	<1.0	0.4
Mg, mg/L	48	50	49	57	<2	8
Mn, mg/L	20	23	21.5	19	<0.054	-2.5
Na, mg/L	8	10	9	10	<2	1
Ni, mg/L	2.66	2.81	2.735	1.97	<0.010	-0.77
Si, mg/L	24	22	23	35	1.1	12
Zn, mg/L	2.32	1.80	2.06	1.85	0.010	-0.21

These results can be explained by the geochemical behaviour of the VFR system. All the FND samples have pH and redox values that fall into the Fe^{2+} field of the Pourbaix diagram (red symbols in Figure 6), which has been proven by the Fe species analysis. During the AMD passage through the VFR, both the pH and the redox potential slightly increase and approach the boundary to the goethite predominance field (blue symbols in Figure 6). Nevertheless, the iron oxidation kinetics at these pH values are too low, and the AMD residence time in the VFR is insufficient to form ferric iron, resulting in insufficient particle formation for subsequent autocatalysis.

After 22 weeks of the field trial, the VFR was emptied to visually identify the quantity of sludge that had formed over the course of the trial and in order to prepare the next trial set up. It was observed that the inert slag was only slightly covered with sludge, and that most of the slag substrate had only a thin sludge coating, which could be easily removed mechanically (Figure 7). This visual result corroborates the chemical result with relatively low Fe removal rates. In comparison to other VFRs, the quantity of sludge formed is notably low, indicating that the AMD treatment for Fe removal at that site was not effective.

Figure 5. Total (top) and ferrous (bottom) iron (filtered, unfiltered) removal rates during the VFR field trial

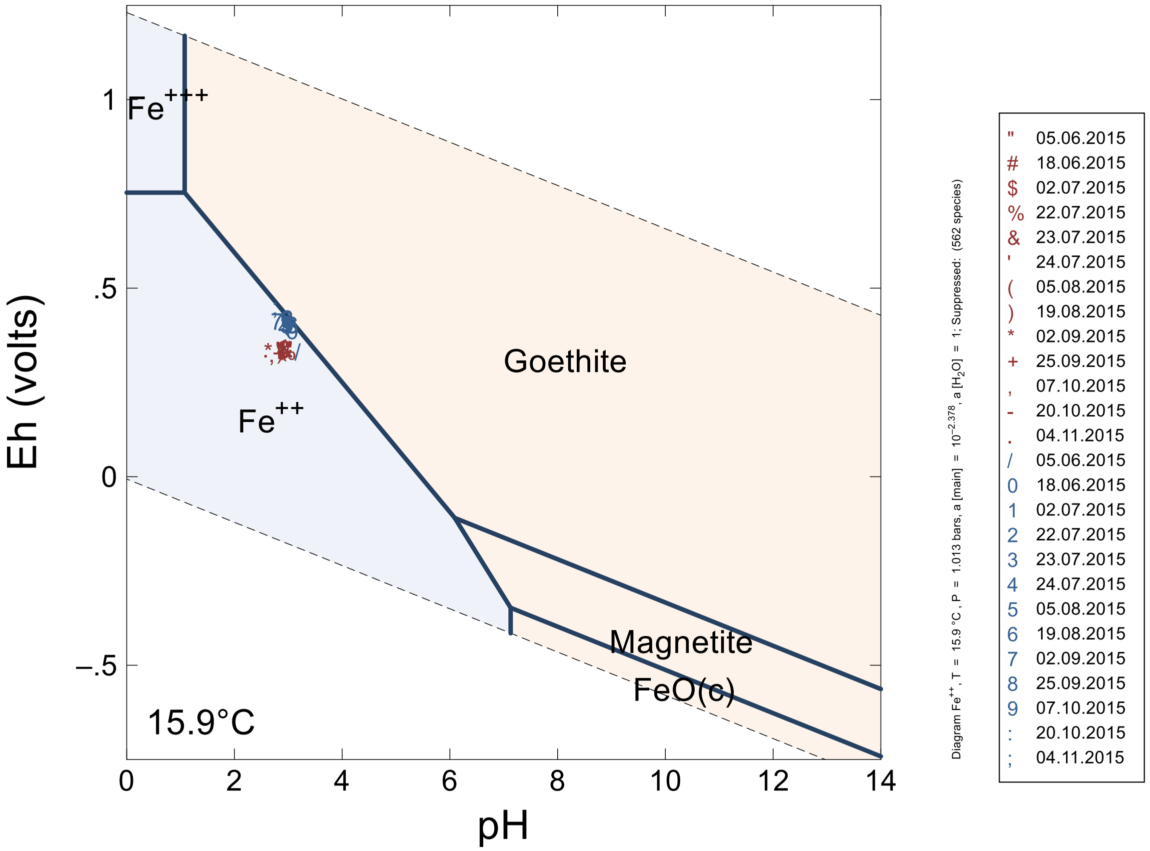

Figure 6. Pourbaix-diagram of the AMD (unmodified image from Geochemist's Workbench). Red symbols: inflowing water, blue symbols: outflow from the VFR. [Fe] = 234 mg/L, temp = 15.9 °C, *p* = 1.013 bar

Figure 7. Sludge formation on top of the inert slag substrate of the VFR at the FND site

CONCLUSIONS

It could be demonstrated that the circumstances of the Cwm Rheidol VFR were distinctive in that the mine adit–pipe–galley system constituted a pre-treatment that allowed the Cwm Rheidol MIW to be treated in the experimental VFR. However, at the Fanie Nel Discharge, although the quality of the mine water was comparable to that of Cwm Rheidol, this unique “pre-treatment” was not available, resulting in only low removal rates for iron. These unfiltered Fe-removal rates never exceeded 51% for Fe_{tot} (range: -22–51%, average 19%) and for Fe^{2+} (range: 1–61%, average 32%) and are therefore very low (Figure 5). The reason for that behaviour is that the pH of the AMD is very low and most of the Fe is ferrous iron, which didn’t have enough time to oxidise between the discharge location and the VFR. Therefore, no particles were available on which the Fe could sorb, precipitate and result in autocatalytic Fe removal.

In conclusion, the Fanie Nel Discharge VFR demonstrated a substantial underachievement, and a VFR at this location was therefore not a viable option. Furthermore, the experiment demonstrated that net acidic MIW cannot be treated reliably with a VFR.

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REFERENCES

Barnes, A. (2008). The rates and mechanisms of Fe(II) oxidation in a passive vertical flow reactor for the treatment of ferruginous mine water [PhD, Cardiff University]. Cardiff.

Blanco, I., Sapsford, D.J., Trumm, D., Pope, J., Kruse, N., Cheong, Y.-w., McLauchlan, H., Sinclair, E., Weber, P. and Olds, W. (2018). International Trials of Vertical Flow Reactors for Coal Mine Water Treatment. Mine Water Environ., 37(1), 4-17. https://doi.org/10.1007/s10230-017-0491-z

Brix, H. (1987) The applicability of the wastewater treatment plant in Othfresen as scientific documentation of the root-zone method. Water Sci. Technol., 19(10), 19-24. https://doi.org/10.2166/wst.1987.0093

Dube, G., Mello, T., Vadapalli, V., Coetzee, H., Tegegn, K., Lusunzi, R., Moja, S., Malatji, M., Sinthumule, M.E. and Ramatsekisa, R. (2021) Full-scale Reducing And Alkalinity Producing System (RAPS) For The Passive Remediation Of Polluted Mine Water From A Flooded Abandoned Underground Coal Mine, Carolina, South Africa. In, Mine Water Management for Future Generations, Cardiff, Wales, United Kingdom.

Edwards, P. and Potter, H. (2007) The Cwm Rheidol Metal Mines Remediation Project – Phase 1. In Cidu, R. and Frau, F. (Eds.), Water in Mining Environments (pp. 181-185). Mako Edizioni.

Florence, K., Sapsford, D.J., Johnson, D.B., Kay, C.M. and Wolkersdorfer, C. (2016) Iron mineral accretion from acid mine drainage and its application in passive treatment. Environ. Technol., 37(11), 1428-1440. https://doi.org/10.1080/09593330.2015.1118558

Florence, K., Sapsford, D.J. and Wolkersdorfer, C. (2015) Mechanisms of Iron Removal during Passive Treatment of AMD in a Vertical Flow Reactor. In Brown, A., Bucknam, C., Burgess, J., Carballo, M., Castendyk, D., Figueroa, L., Kirk, L., McLemore, V., McPhee, J., O’Kane, M., Seal, R., Wiertz, J., Williams, D., Wilson, W. and Wolkersdorfer, C. (Eds.), Agreeing on solutions for more sustainable mine water management (pp. 1-11 [electronic document]). Gecamin. https://doi.org/10.13140/RG.2.1.1463.9121

Hayton, S., Trumm, D., Horton, T., Pope, J., Ross, D. and Williams, M. (2022) Passive Treatment For Elevated Iron And Arsenic In Circum-Neutral Mine Affected Water. In, IMWA 2022 – Reconnect, Christchurch, New Zealand.

Jones, A.N. and Howells, W.R. (1975) The partial recovery of the metal polluted River Rheidol. In Chadwick, M.J. and Goodman, G.T. (Eds.), The Ecology of Resource Degradation and Renewal (pp. 443-459). Blackwell.

Jones, O.T. (1922) Lead and Zinc – The Mining District of North Cardiganshire and West Montgomeryshire (Vol. 20). Geological Survey.

Kleinmann, B., Skousen, J., Wildeman, T., Hedin, B., Nairn, B. and Gusek, J. (2021) The Early Development of Passive Treatment Systems for Mining-Influenced Water: A North American Perspective. Mine Water Environ., 40(4), 818–830. https://doi.org/10.1007/s10230-021-00817-8

Mashalane, T.B., Novhe, N.O., Yibas, B., Coetzee, H. and Wolkersdorfer, C. (2018) Metal Removal from Mine Water Using Passive Treatment Technologies (Anaerobic and Aerobic) in the Ermelo Coalfields, Mpumalanga, South Africa. In, IMWA 2018 – Risk to Opportunity, Pretoria.

Novhe, O., Yibas, B., Coetzee, H., Atanasova, M., Netshitungulwana, R., Modiba, M. and Mashalane, T. (2016) Long-Term Remediation of Acid Mine Drainage from Abandoned Coal Mine using Integrated (anaerobic and aerobic) passive Treatment System, in South Africa: A Pilot Study. In Drebenstedt, C. and Paul, M. (Eds.), IMWA 2016 – Mining Meets Water – Conflicts and Solutions (pp. 668-675). TU Bergakademie Freiberg.

Sapsford, D.J., Barnes, A., Dey, M., Williams, K.P., Jarvis, A., Younger, P. and Liang, L. (2006) Iron and Manganese Removal in a Vertical Flow Reactor for Passive Treatment of Mine Water. In, ICARD 2006, St. Louis.

Sapsford, D.J. and Williams, K.P. (2009) Sizing criteria for a low footprint passive mine water treatment system. Water Res., 43(2), 423-432. https://doi.org/10.1016/j.watres.2008.10.043

Snyman, C.P. (1998) Coal. In Wilson, M.G.C. and Anhaeusser, C.R. (Eds.), The Mineral Resources of South Africa (pp. 136-205). Council for Geoscience.

Wolkersdorfer, C. (2022) Mine Water Treatment – Active and Passive Methods. Springer. https://doi.org/10.1007/978-3-662-65770-6

Younger, P.L., Banwart, S.A. and Hedin, R.S. (2002) Mine Water – Hydrology, Pollution, Remediation. Kluwer. https://doi.org/10.1007/978-94-010-0610-1