

Evaluation of industrial wastes as active materials in permeable reactive barrier for acid mine drainage remediation

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Abstract The aim of this work was to evaluate the use of agricultural and industrial waste materials as reactive agents into Permeable Reactive Barriers (PRB) for Acid Mine Drainage (AMD) remediation. Laboratory-scale columns containing Volcanic Scoria (VS) as porous fill material and approximately 18% w/w of industrial wastes used as reactive materials were used as PRB under up-flow continuous mode treating 1.9 L AMD d⁻¹. Drinking water sludge (DWS) clearly enhanced AMD remediation because of chemical neutralization and biological sulphate reduction, which produced metals precipitation. The removal yields obtained when using DWS as reactive material ranged from 60 to 90%. Because of these results, the use of the DWS in PRBs would allow to simultaneously carry out the management of an industrial waste and the remediation of AMD through a low-cost and environmentally sustainable procedure.

Keywords: permeable reactive barrier; industrial waste; acid mine drainage

1. Introduction

Acid mine drainage (AMD) is an industrial polluted effluent that is associated with mining activities and abandoned old mining areas. The European Water Framework Directive (WFD, Directive 2000/60/EC) established the main goal of achieving a good environmental quality for all European water bodies. Then, remediation measures should be adopted to fulfil the WFD requirements in many effluents such as the AMD generated in abandoned metal mining areas.

AMD can be treated by different types of technologies, which can be classified into two different groups: active and passive technologies (Johnson and Hallberg, 2005). The performance of the passive technologies is based on naturally occurring physical, chemical or biological mechanisms such as chemical pH neutralization (and subsequent metals precipitation as hydroxides or sulphides), microbiological sulphate reduction that also

causes pH increase, chemical reduction of metals, metal uptake by plants, and metal capture by adsorption or ion exchange. These mechanisms are developed in low-cost flow systems such as drains, open channels, sorbent or ion exchange columns, permeable reactive barriers (PRB) and constructed wetlands. PRB consist of reactive porous materials installed in the path of water migration to capture or degrade water pollutants (Rambabu et al., 2020; Shabalala and Masindi, 2022). As in most of the passive systems, different agricultural or industrial wastes or by-products have been studied as reactive materials for PRBs (Moodley et al., 2018; Masindi et al., 2022). Many of these waste materials are selected because chemical characteristics as well as their abundance or proximity to metallic mining areas.

In this context, the aim of the present work was to evaluate the feasibility of using new agricultural or industrial solid wastes as reactive materials for AMD remediation by means of PRB systems.

2. Material and Methods

Synthetic AMD was synthesized in the laboratory based on the typical composition of real AMD. The composition of the synthetic AMD is shown in Table 1.

Table 1. Characteristics of synthetic AMD.

| Metal | Concentration (mg/L) | Metal | Concentration (mg/L) |
|------------------|----------------------|-------------------------------|----------------------|
| Fe ⁺² | 600 | Zn ⁺² | 50 |
| Al ⁺³ | 450 | Ni ⁺³ | 20 |
| Mn ⁺² | 120 | SO ₄ ²⁻ | 2045 |

Volcanic scoria (VS) was used as a structural base and porous fill material for the PRBs. Four agricultural/industrial waste materials were used as reactive agents into the PRBs: sugar foam (SF), paper mill sludge (PMS), drinking water treatment sludge (DWS) and olive mill waste (OMW).

With the filling material and the wastes used as reactive materials, five PRBs made from polyvinyl chloride were configured. Each PRB presented the following dimensions: 0.2 m diameter × 0.75 m height. Each column was filled with VS (which was used as a structural solid medium offering high porosity) and one of the reactive materials, which accounted to 18% of the total weight. The final volume of the PRB was in all the cases 23.6 L volume. Additionally, a reference PRB was configured only with VS in order to be used as reference tests. According to the above description, five continuous PRB were configured. Each one was fed with $1.9 \pm 0.2 \text{ L d}^{-1}$ (hydraulic retention time approximately $7.0 \pm 1.0 \text{ d}$) of the synthetic AMD. In order to facilitate the metal removal by means of bioprocesses the PRBs configured with reactive materials were inoculated using 50 mL of sludge from the anaerobic sludge digester of a domestic wastewater treatment plant. During the steady state operation of the PRB, weekly samples (0.5 L) were taken from the outlet sampling port.

These samples were analyzed for: i) pH (pH-meter basic 20, Crison); ii) concentration of dissolved metals (total Fe, Al^{+3} , Zn^{+2} , Mn^{+2} , Ni^{+2}) by means of ICP-AES using a Thermo ICAP 6500 spectrometer (Thermo Electron, Cambridge, UK); iii) sulphate ion concentration by ion chromatography using an 883 Basic IC Plus chromatograph equipped with a Metrosep A Supp 5 column (Metrohm, Herisau, Switzerland); iv) Chemical Oxygen Demand (COD) measured by UV-Vis photometer; and v) Total Suspended Solids (TSS) concentration measured by dry weight according to standard methods (A.P.H.A., 1998).

3. Results

In Figure 1 it is presented the effluent metal concentration as well as the metal removal yield from the AMD during the treatment with the PRB. On the one hand, when using OMW as reactive material, a negligible metal removal was observed in most of the cases. Only when dealing with the Al, the metal removal was significant. Regardless of the mechanism that could occur in this case, it is considered that the presence of a significant amount of olive oil in the OMW can hinder the contact of the solid material with the soluble metals in the AMD. This limitation significantly reduced the possible precipitation or capture effects (Pagnanelli et al., 2009). On the other hand, it was observed that SF, PMS and DWS were the active materials presenting good performance in terms of metal removal when compared with the reference test carried out with the filling material, VS.

Regarding the material that presented the highest alkalinity, the SF, its performance was worse than expected. Theoretically, the high alkalinity should lead to higher chemical metal precipitation yields. However, the existence of parallel chemical reactions as well as other effects or mechanisms that contribute to the overall metal removal led to worse results than expected.

In general terms, when analyzing the metal removal yields, it was considered that the DWS was be the active material causing the best metal removal yield which ranged from 60% to 90%.

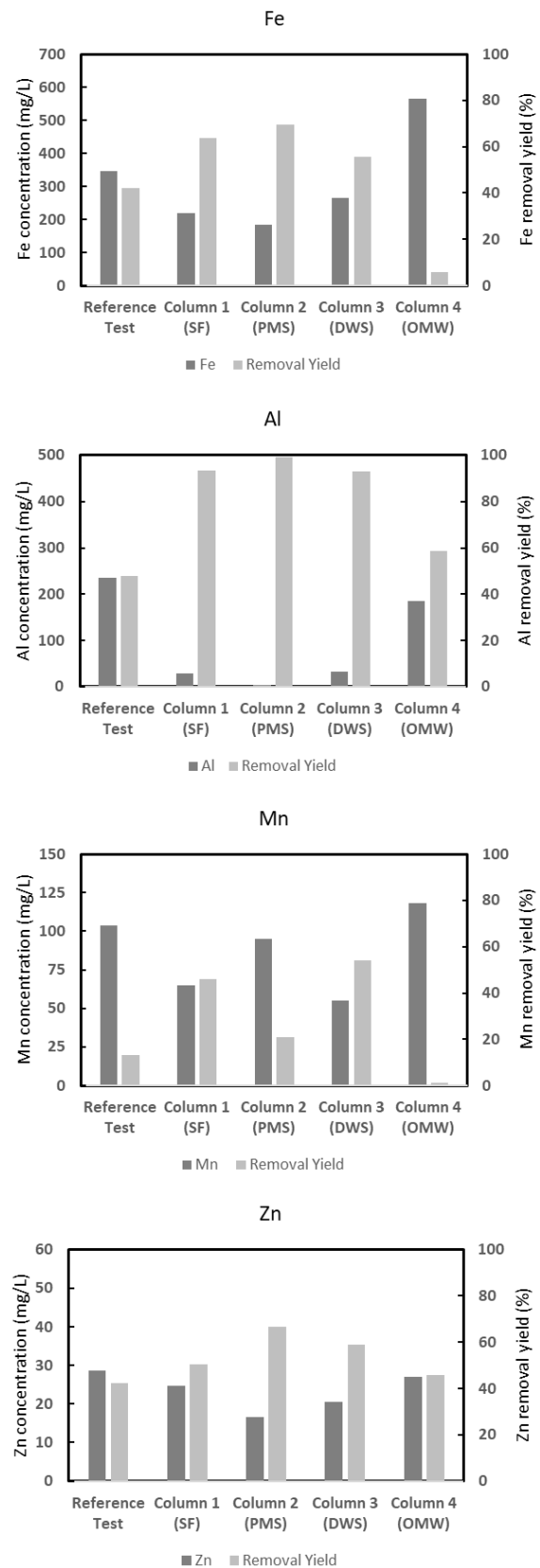


Figure 1. Evolution of the main metal parameters during the remediation of the AMD in PRBs.

4. Conclusions

According to the results obtained in the present work, it is observed that OMW is a material that can be discarded as reactive material in PRB because it presented Fe removal yields lower than 10% and a negligible Mn removal yield. The SF, despite the good initial expectations due of its high alkalinity, does not present the expected performance. Being its Mn and Zn removal yields lower than 50%. However, PMS and DWS offered very good results in terms of metal removal, ranging its removal yields between 60 and 90%, except in the case of the Mn when operating with PMS. Finally, it must be highlighted that the use of wastes as reactive materials in PRBs would allow the remediation of AMD through a low-cost and environmentally sustainable procedure at the same time that a waste is valorized.

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References

- A.P.H.A.-A.W.W.A.-W.P.C.F., 1998. Standard Methods for the Examination of Water and Wastewater, 20th ed. American Public Health Association/American Water Works Association/Water Environment Federation, Washington DC, USA.
- Johnson, D.B., Hallberg, K.B., 2005. Acid mine drainage remediation options: a review. *Sci. Total Environ.* 338, 3–14. <https://doi.org/10.1016/j.scitotenv.2004.09.002>
- Masindi, V., Foteinis, S., Renforth, P., Ndiritu, J., Maree, J.P., Tekere, M., Chatzisyneon, E., 2022. Challenges and avenues for acid mine drainage treatment, beneficiation, and valorisation in circular economy: A review. *Ecol. Eng.* 183, 106740. <https://doi.org/10.1016/j.ecoleng.2022.106740>
- Moodley, I., Sheridan, C.M., Kappelmeyer, U., Akcil, A., 2018. Environmentally sustainable acid mine drainage remediation: Research developments with a focus on waste/by-products. *Minerals Eng.* 126, 207–220. <https://doi.org/10.1016/j.mineng.2017.08.008>
- Pagnanelli, F., Viggli, C.C., Mainelli, S., Toro, L., 2009. Assessment of solid reactive mixtures for the development of biological permeable reactive barriers. *J. Hazard. Mater.* 170, 998–1005. <https://doi.org/10.1016/j.jhazmat.2009.05.081>
- Rambabu, K., Banat, F., Pham, Q.M., Ho, S.-H., Ren, N.-Q., Show, P.L., 2020. Biological remediation of acid mine drainage: Review of past trends and current outlook. *Environ. Sci. Ecotechnol.* 2, 100024. <https://doi.org/10.1016/j.ese.2020.100024>
- Shabalala, A., Masindi, V., 2022. Insights into mechanisms governing the passive removal of inorganic contaminants from acid mine drainage using permeable reactive