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Review

Acid mine drainage (AMD) contamination in coal mines and the need for extensive prediction and remediation: a review

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Abstract

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Keywords: AMD coal mines contamination remediation Globally, the major source of environmental pollution as a result of mineral exploitation and processing is acid mine drainage (AMD). AMD refers to outflowing streams of acidic constituents from pyrite-bearing ore mines. The exposure of pyrite (FeS₂) in coal waste dumps to atmospheric oxygen and water in the presence of microbial communities promotes the formation of sulphuric acid which leaches out the inherent heavy metals into the mine discharge, a phenomenon called pyrite oxidation. AMDs are usually characterized by a convoy of toxic heavy metals, most of which are transition elements (copper, nickel, zinc, etc.) and arsenic at concentrations higher than the limits permitted by environmental regulations. The impact of this acidic discharge from coal mines on downstream/underground waters and farm lands within the corresponding mining zones have been severally reported by previous researchers, but not so much have been discussed on extensive prediction and remediation. It is in view of this that the current paper reviews the need for extensive prediction and remediation approach for coal mines under the following subheadings; General introduction, AMD sources identification, representative sampling, adoption of a prediction model, determination of AMD potential and quality via static and kinetic tests and the development of an economically sustainable remediation strategy. It is thought that this article would be useful to academia as well as policy makers that are responsible for the development and implementation of environmental regulations in coal mines.

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Introduction

Mine waste water and its management constitute sources of an environmental problem for the global mining industry, not just for the now but also for the future. Its quality and quantity are a function of the ore and mine type (mainly sulphidic ores and open casts). This creates serious environmental damages if not properly managed. Globally, waste generated by mining per year is estimated at 15 billion tonnes which exceed annual global municipal wastes generated by

ten folds (Jeffery, 2004; Soni and Wolkersdorfer, 2016). In India alone, 2.6 million people were displaced by mining between 1950 and 1990. Between the years 2002 and 2013, about 450 conflicts were recorded between mining companies and their individual host communities (Gankhuyag and Gregoire, 2018). All of these have significantly smeared the image of the industry through the intense media and civil society criticisms. Thus, the need for remediation of contaminated sites cannot be

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overemphasized. Contaminated site herein refers to a place where soil, groundwater, sediments and air have been impacted by toxic substances above the limits recommended for specific use which pose a risk to the wellbeing of the residing human, plants and animal population. The total measure of corrective steps taken to mitigate or eliminate the contamination is called remediation (UNEP-BEP Barcelona convention. https://www.unenvironment.org/ topics/chemicals-waste/what-we-do/emerging issues/ lead-and-cadmium). The mining of coal as; a traditional road construction raw material, an additive in ceramics manufacturing process and a reducing agent in the blast furnace process for iron and steel manufacturing, be it conventional or artisanal, leaves behind these foot prints of contamination on water bodies in mining zones as a result of oxidation of pyrite and the consequent leaching of heavy metals present in the ore into the waste streams.

Within academia, field services practitioners, state environmental regulatory bodies and global environmental policy makers such as the United Nations Environment Program (UNEP), there has been a quest for the development of research interfaces that can be widely beneficial to the mine environment. That is, the building of research bridges that could link the fields of geology, mining and hydrometallurgy, the latter which uses relevant technologies for tailings/waste water remediation (Pongratz, 2004; Bhargava et al., 2016). All of these require rejigging of local environmental laws, for which Swart (2003) brought to focus, in the case of South Africa, the roles and responsibilities of the Government, the extractive industry and host communities.

Mine closure programs which ordinarily should be designed by the miner, as a major precondition for license approval, seek to accomplish any of the following goals; neutralisation of the acid generated, prevention of exposure of the waste rock to atmospheric oxygen and flowing/percolating and the prevention of bacteria in the likes of Acidithiobacillus ferrooxidans from catalysing pyrite oxidation (http://www.americangeosciences.org). The current article discusses the following; AMD sources identification, representative sampling characterisation, determination of AMD potential and quality via static and kinetic tests, the development of a prediction model and the development of economically sustainable remediation strategies. Odagbo coal mine has also been discussed as a case study.

Case Study

Odagbo is an agrarian community near Ankpa in Kogi State, middle-belt Nigeria and is situated at an altitude of 275 m above sea level. It is located between latitudes 7° 28′ 30″ N and 7° 29′ 00″ and longitudes 7° 43′ 30″ E and 7° 44′ 0″ E spanning 4 km² area (Omali and Egboka, 2014; Momoh et al., 2017). The

deposit was described by Nigeria's Ministry of Mines and Steel Development (2014) and Ameh (2013) as medium quality coal with moisture and sulphur contents of 10.30% and 0.65%, respectively and a surface mine with a proven reserve of 100 million metric tonnes. Mines within the Odagbo coal measures have acid mine drainage owing to the availability of SO₄ during coal deposition in combination with Fe and organic material which promote pyrite formation. Open cast mines are characterised by Al- and trace element-laden acid mine drainage compared to underground mines. These reactions that promote acid mine drainage formation release trace elements and Al at high and complete rates in open cast mines where waste rocks particles possess higher reactive surface areas compared to underground mines wastes. The coal mine drainage chemistry is also influenced by hydrogeology where the flooded surfaces release many acid-rich drains owing to the abundance of atmospheric oxygen needed to react with pyrite (Pope et al., 2010; UNEPFI, 2020). Effluents from Odagbo coal mine are characterised by high levels of toxic metals such as nickel, copper, lead, arsenic, iron and have impacted water bodies around the mine zone (Ameh et al., 2014; Momoh et al., 2017). The pH values obtained for discharge waters, streams and boreholes in a recent field study (Ojonimi et al., 2020) around the mine range between 1.2-2.3, 3.5-4.5 and 5-6, respectively. Thus, the ongoing artisanal mining in the community, as well as previous conventional activities in the area, have led to AMD contamination which could constitute serious threats to the health of the host community. A detailed map of the study area is shown in Figure 1.

AMD Sources Identification

This section provides a roadmap to the identification of potential AMD sources in coal mines. These sources may be described as primary or secondary, depending on the rate of acid generation. The rate of acid generation is determined by factors such as microbial activity, pH, exposed surface area of metal sulphide, concentration of oxygen in the water phase and temperature (John et al., 2017). Below is a brief description of the sources;

Mine waste dump (primary source): Uncovered pile of rocks that are deemed unprofitable for further processing. Considering that these materials may be used for backfilling at the end of the project, polluted acidic water, is formed from their long relatively long-term exposure to oxygen and water.

Tailings impoundments (primary source): A demarcated area to which mineral processing tailings in form of sludge are traditionally discharged. From this facility, there is a possibility of ground water contamination through leakage of the toxic content. Dam breakage can also cause releases of toxic contents into receiving streams.

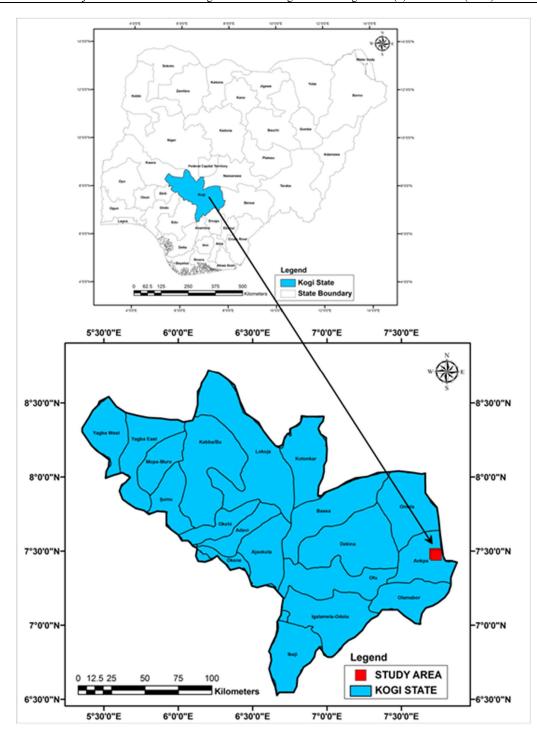


Figure 1: Map of the study area [After, Momoh et al. (2017)].

Underground and open cast workings (primary source): The openings made for mineral exploitation. Owing to limited availability of oxygen as well as low Al/Fe ratio, AMD generation rate in underground mines is comparatively slower than in open cast mines. The wide distortion of open cast coal sediment allows for greater exposure of the surfaces of the waste rocks to secondary reactions between the AMD generated

sulphuric acid and alumino-silicates minerals such as feldspar present in the catchment. This further introduces more acidity into the system by the release of higher concentrations of aluminium ion (Al³⁺) (Baker and Benfield, 1999).

Underground discharged water (primary source): The volume of water discharged daily from underground mines into natural water bodies is

enormous as the activity requires significant volumes of water, especially in the extraction and processing phases. For example, to achieve the production of a ton of gold, about 716 m³ of water is required, most of which is used for the following purposes; release of minerals from host rocks, washing and transportation of materials, dust control and cooling of drilling machinery (Hutson et al., 2004; Mudd, 2008; Ray and Dey, 2020).

Abandoned pits and mine workings (secondary source): Any place in the mine previously opened and ventilated for miners to work or travel through during the active days of the project (https://www.coaleducation.org). They represent a very significant environmental liability, as long after mine closure, AMD created in these pits can continue over several decades if they are not properly covered.

Tailings pipes (secondary source): in some cases, mines may release contaminated water in a controlled or uncontrolled manner into nearby streams. In places where environmental regulations are porous, riverine tailings disposal is practiced in which case, pipe network is designed to discharge tailings directly into rivers

Pit walls (primary source): These are made during open cast construction and often have increased surface area for the exposure of sulphide-bearing rock to forces responsible for AMD occurrence. There is a likely release of more acidity in mine drainage from pits walls in the first one year and a sharp decline as years of mine sites increase. The impact of AMD from pit walls on water quality can span over 64 to 136 years. Fe, Al and H⁺ constitute the most abundant components of the acids produced from pit walls at sites of ages 0-1, 3-20 and 20-138 years, respectively (Pope et al., 2018).

Ore stockpiles (primary source): These are heaps of materials kept for further processing and are considered as containing lean amounts of valuable metal. They constitute centres for AMD initiation as they are often uncovered.

Representative Sampling for AMD Assessment

Data on mine water discharge and protocols of waste management are either under reported or not reported and this imposes a barrier to understanding the risk. If ever reported, the reliability of results from analytical tests can be viewed via the mirrors of the accuracy of the mode of collection, type and nature of samples. Prior knowledge of the geology of the area, costs and timeline for conducting the tests are factors responsible for the sampling regime. The sampling strategy, therefore, provides a lead to the appropriate analytical method (s) for adoption (Miranda and Sauer, 2010). Having considered the mentioned aspects, corrective measures can be adopted to mitigate or

remediate the contamination. Within coal mines, this article suggests an extensive sampling campaign that would yield about 150 solids (Matsumbo et al., 2018) accounting for bulk and drill-core which should provide a comprehensive description of the waste rocks/deposited material and their characteristic properties for testing. Up to 150 water samples collected from tailings ponds may be analysed accounting for both composite and individual samples. Mineral characterization and quantitative analyses are also recommended to predict AMD occurrence in coal mines.

The Adoption of a Prediction Model

This section brings to bear, typical AMD prediction model. At this point, it is essential to introduce the basic AMD chemistry as it provides the theoretical frame work for its occurrence (Ferguson and Erikson, 1998):

$$2\text{FeS}_2(s) + 2\text{H}_2\text{O} + 7\text{O}_2 \rightarrow 4\text{H}^+ + 4\text{S}_4^{2-} + 2\text{Fe}^{2+}$$
 [1]

In the reaction (Equation 1.0), pyrite is oxidized in the presence of water to form four molecules of hydrogen ion and sulphate which are the products of sulphuric acid dissociation in solution. Provided the pH values are low, further oxidation of the soluble ferrous ion (Fe^{2+}) to ferric ion (Fe^{3+}) occurs slowly (Equation 2.0).

$$4Fe^{2+} + O_2 + 4H^+ \rightarrow 4Fe^{3+} + 2H_2O$$
 [2]

As pH rise to levels within the range, 3.5 and 4.5, a filamentous bacterium known as Metallogenium forms a microbial community that facilitate oxidation. The same reaction (Equation 2.0) occurs at pH levels below 3.5 but, in this case, promoted by *Thiobacillus ferrooxidans*, an iron bacterium. Equation 3.0 proceeds if the ferric ion (Fe³⁺) formed is in contact with pyrite. It goes to dissolve the pyrite to generate more acids, evidenced by 16 molecules of H⁺.

$$2\text{FeS}_2$$
 (s) $+14\text{Fe}^{3+} + 8\text{H}_2\text{O} \rightarrow 15\text{Fe}^{2+} + 2\text{SO}_4^{2-} + 16\text{H}^+$ [3]

Equation 4.0 indicates the precipitation of Ferric iron as hydrated iron oxide which can be identified as an amorphous yellow to reddish-brown substance deposited under ponds and rock surfaces in mines (Figure 2).

$$Fe^{3+} + 3H_2O \leftrightarrow Fe(OH)_3 (s) + 3H^+$$
 [4]

Generally, the rate of chemical reaction is driven by the prevailing pH and it progresses slowly at 2-3 pH values. Higher intensity of the formation of hydrated iron oxide and acid (Equation 4.0) can facilitate the accentuated removal of dissolved Fe without necessarily removing sulphate (SO₄²⁻), meaning that their relationship in water is reaction kinetics dependent. Therefore, high sulphate concentration does not necessarily translate to high Fe concentration (Campaner et al., 2014).



Figure 2. Precipitation of ferric iron as hydrated iron oxide.

Natural weathering processes which occur slowly are responsible for oxidation of sulphide minerals. Acid released from the undisturbed rocks, may not cause serious damages on the ecosystem as availability of carbonates within the catchment provides a buffering system at such conditions (Good Practice Guidance for Management of Acid and Metalliferous Drainage, 2020-2025; http://www.mrt.tas.gov.au). However, exploitation and beneficiation of the ore bodies increasingly promote the rate of these chemical reactions owing to the exposure of large volumes of sulphide rocks and waste dumps to water and oxygen. The rate at which these reactions occur varies from one sulphide mineral to the other. For instance, marcasite oxidises faster than crystalline pyrite (King, 2020). There is wide variability of this AMD chemistry in coal mines, with pH ranging from 1.2-2.5 and heavy metal and other chemical concentrations ranging within and above the limits permissible by UNEP convention on mine water management. These variations are influenced by factors including; mine type, hydrogeology and geology of the area. AMD prediction seeks to achieve the following objectives; (i), to establish the extent to which acid will be generated from a discrete volume of waste materials, (ii), to predict the quality of the drainage based on the rate of acid formation measured (USEPA, 1994). The first step towards implementing a mine closure program is to extensively examine the source, extent, type and amount of prevailing contamination. This is followed by determining how and the extent to which the environment and human population is impacted.

AMD Potential and Quality via Static and Kinetic Tests

There are several important factors that must be considered when evaluating the acid generation potential of rock material. These include; amount of acid generating (sulfide) minerals present in the ore [pyrite (FeS₂), chalcopyrite (CuFeS₂), sphalerite

(ZnS), galena (PbS), cinnabar (HgS) and molybdenite (MoS₂)], quantity and type of potential contaminants present, type of carbonate minerals such as calcite (CaCO₃) present, mineral surface area available for reaction, particle size of the waste, availability of water and oxygen and the bacterial community present (Thorsten, 2013). These factors were extensively discussed by Ojonimi et al. (2020) in a previous review of acid mine drainage in coal mines. This article suggests a robust geochemical characterization program that provides a basis for risk assessment and closure of coal mines. The primary intention of this geochemical characterization campaign is to provide a lead to understanding of the geochemical characteristics of geological materials specific to mines with the use of the conventional static and kinetic testing methods designed for addressing the potential to generate acidic drainage (Presita et al., 2015). The static and kinetic geochemical predictive tests suggested are the acid base accounting (ABA) test, Net Acid Generation (NAG) test and the standard Humidity Cell Test (HCT). A static test determines both the total acid generating and total acid neutralizing potential of a given sample. ABA analyses which determine the acid and neutralizing potentials of waste rocks are used to distinguish between source rocks and non-source rocks, with the latter described as non-acid forming (Kontopolous et al., 1996). These tests find several industrial applications and are traditionally carried out in field-based laboratories of mines. It forms the theoretical framework for a material's potential to generate net acid conditions. The method may be viewed as estimating a given material's 'total potential content of acidity or alkalinity. This method determines the nature of sulphur (sulphide sulphur and sulphate sulphur) present in the ore by measuring the amount of nitric acid-extractable sulphur and the amount of hot waterextractable and hydrochloric acid-extractable sulphur (NDEP, 2013). Neutralizing potential (NP) is measured by the use of the method adopted by Presita et al. (2015) as modified from the protocol of Sobek et

al. (1978). The Static NAG testing protocol simulates a second AMD by oxidizing and accelerating the dissolution of sulphide minerals with the use of hydrogen peroxide from which acid production and neutralization results can be directly measured while NAG value is established by two-stage titrations of the leachate with sodium hydroxide at pH 4.5 and pH 7 (Miller and Bruland, 1997). Kinetic tests represent the qualitative evaluation of the drainage emanating from the AMD sources and these take into account the geotechnical characteristics of the source rock and the prevailing metrology of the area under study. For instance, there are temperature and pH variations of AMD sources during rainy and dry seasons which consequently affect the activity of microbial communities that promote pyrite oxidation.

Although it is unclear what the position of the local environmental guidelines on these tests, researchers in other climes (Vithana et al., 2013; Dold, 2017; Matsumbo et al., 2018) have reported that there are shortcomings in the use of same for distinguishing between AMD source or non-source rocks. The challenge is that iron carbonate mineralogy as well as the effects of sulphur are not accounted for by ABA and NAG. On the other hand, the usual practice with tailings ponds as potential AMD generation sites is to carry out preliminary onsite pH investigations on samples collected from them, followed by subsequent laboratory analyses. This is not without its drawbacks as there are occurrences of seasonal pH variations when it comes to preliminary field investigations as well as further laboratory works. The usual onsite practice with samples taken from tailings ponds is to acidify them with concentrated acid, but this begins to impose limits on the accuracy of TDS, EC and heavy metals results if they are to be kept for analyses longer than three months to allow for extensive studies. Prediction of AMD occurrence via the use of analytical instruments for quantifying mineralogy and describing morphology of samples are expected to generate reliable data. The combination of traditional static and kinetic tests with the use of modern analytical methods such as X-Ray Diffraction (XRD), Inductively Coupled Plasma-Mass Spectroscopy (ICP-MS), Scanning Electron Microscopy (SEM) would greatly yield reliable data for such study.

Development of Economically Sustainable Remediation Strategies for Coal Mines

It is difficult to control AMD at source. Thus, in order to minimize its impact on the receiving water bodies, collection and treatment of the discharge represents an alternative approach. Several AMD passive treatment approaches have been developed in the last 3 decades, where the main concern is the search for an adequate alkaline remediation material. Factors such as neutralization potential, cost, proximity to the mine and quantity required provide the lead for the choice of an alkaline material (Saha and Sinha, 2018). Despite

the recognition of limestone as an abundant neutralising agent of choice for AMD treatments, previous research works on its use report that its neutralization rate is limited by high metal concentration in solution particularly by slowing down its dissolution rates. For instance, Sun et al. (2019) noted that dissolved iron in acid mine drainage not only coats limestone surfaces and hinders dissolution rates, but iron precipitation reactions increase the required neutralization time. Similarly, Crovatta and Bingham (2016) reported that the use of limestone for AMD neutralisation has faced the challenge of low solubility and slow dissolution rate compared to other alkaline reagents and decreased efficiency of neutralization attributed to its "armouring" with Fe3+ and Al3+ compounds. Armouring herein refers to strong adhesion or complete encrustation. The use of limestone as a sole neutralisation reagent is also characterised by generation of high volumes of sludge. Aside this, the near neutral conditions offered by limestone alone leaves behind substantial amounts of contaminants and therefore possess challenges that go beyond mere neutralisation (Thorsten, 2013).

The current article proposes the potential use of other abundant resources to design effective and inexpensive methods as sustainable solutions to the problems imposed on the residents of coal mining zones by heavy metal-laden waste streams. More recent studies have identified adsorption as an effective and economical method used in acid mine drainage remediation (Haidar et al., 2016). The process of adsorption offers flexibility in design and operation and has the capacity to increase the quality of treated mine drainage (Fu and Wang, 2011). An adsorbent used in adsorption technologies may be viewed as any low-cost material provided it is naturally/cheaply abundant, requiring little processing or is an industrial waste (Bailey et al., 1999). Many types of low-cost adsorbents have been used; clay adsorbents and activated carbon (Jusoh et al., 2007), all of which do not require continuous operation once applied (De Gisi et al., 2016). Recently, scholars have focused on processing solid agricultural bye products into porous materials described as activated carbon for passive AMD treatment methods, bearing in mind the fact that passive treatment could have minimal overall environmental impacts compared to active treatment technologies (Hengen et al., 2014). Activated carbon is a non-hazardous carbonaceous material and can adsorb a wide variety of substances including heavy metals. It is capable of attracting molecules to its internal surface and is so called an adsorbent. Also, rice husk may be considered a low-cost material since it is naturally abundant as a solid agricultural bye product which does not require huge energy for processing. Recent research pathways have focused on the conversion of this solid bye product into porous and active large surface substances called activated carbon for passive remediation of AMD. Nigeria presently imports large quantities of activated carbon for use in waste water treatment. Research interests in

the domain of conversion of agricultural bye products into activated carbon from local sources for AMD treatment have therefore been growing. Despite their abundance as described above, there is limited information on remediation of acid mine drainage using the combination of limestone and activated carbon. This article therefore proposes the need to investigate the potential use of rice husk activated carbon/limestone (RHAC/Limestone) composite for remediation while minimising the rate of consumption of limestone resource as well as promoting value addition to the agricultural bye-product. Bench scale studies for determining the suitability of a material such as suggested is often designed using the Taguchi Orthogonal Array (OA). It is simple, efficient and provides the best range of parameter combinations in terms of quality, performance and computational costs (Azizi et al., 2010).

Conclusion

The cost incurred in terms of the liability associated with current and future remediation projects by the global mining industry for ignoring the prediction and management of AMD is estimated at 140 billion United States Dollars (Parbhakar-Fox and Lotter Moser, 2015). In Nigeria alone, reports by the federal ministry of mines and steels development (Ministry of Mines and Steel Development: MMSD, 2014) indicate that there are over 70 identified abandoned mines requiring remediation. Aside from the number of sites officially reported, there are several other abandoned mines being reworked by artisans across the country. Most of these artisans have no scientific knowledge of the impact of their activities on the environment. This translates to a huge financial burden to bear by Africa's growing economies. With the case of the 2010 widespread lead poisoning due to artisanal mining that claimed over 40 lives in Zamfara state, Nigeria, extensive AMD prediction and remediation projects should be given top priority for action. Data obtained from such projects could give a lead to working at the prediction and remediation of other sulphide bearing mines within and outside the country. In addition, since AMD is a global challenge facing the mining industry, research works of this nature require synergy between experts in the academia, the industry and international environmental agencies across regional and intercontinental boundaries with the aim of fasttracking the realisation of the United Nations sustainable development goals six (SDGs-6: clean water and sanitation).

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