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Integrated treatment of acid mine drainage using BOF slag, lime/soda ash and reverse osmosis (RO): Implication for the production of drinking water

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ABSTRACT

Acid mine drainage has been an issue of prime concern to international scientific communities. This is due to the magnitude, nature and extent of its environmental impacts. Acid mine drainage (AMD) contains hazardous and toxic chemical species that require removal prior discharging mine water to different environmental compartments. A number of mine water treatment technologies have been developed but they were reported to contain certain benefits and drawbacks. However, there is a need to come-up with environmental friendly and zero-liquid-discharge technologies. The purpose of this novel study was to produce drinking water and recover valuable minerals from acid mine drainage using an integration of Basic Oxygen Furnace (BOF) slag, lime, soda ash and Reverse Osmosis (RO) system. The process can produce very pure water and recover valuable minerals such as hematite, goethite, gypsum, and limestone. Furthermore, brine will be taken to free desalinator for further recovery of salts. To achieve the goals of this study, semi-pilot experiments were done in the laboratory using the aforementioned integrated approach. Interaction of BOF and AMD increased the pH of mine water to ≥ 8. $>$ 99% metals and 75% sulphate were also removal using BOF slag. Residual sulphates and hardness were reduced using lime and soda ash respectively. Gypsum and brucite were recovered as valuable minerals in the lime reactor. Ca as hydrated lime and limestone were recovered in the soda ash reactor. The recovered minerals could be sold as valuable minerals to metallurgical houses and off-set the process/running cost. Reverse Osmosis (RO) was used to further clean the water to meet drinking water quality. A single pass RO system was simulated in Reverse Osmosis System Analysis (ROSA). The produced water meets the requirements of the South African National Standard (SANS) 241 Drinking Water Specifications. As such, it can be concluded that this integrated technology has shown that drinking water and valuable minerals can be recovered from AMD.

1. Introduction

The unique combination of geography, climate, population distribution and the scale of the mineral deposits has led South Africa to immensely depend on mining of its natural resources [1]. This has significantly contributed to the reinforcement of the Gross Domestic Products (GDP) [2]. Even though mining industry contributes significantly to the economy of a given country the twilight of gold and coal mining in SA has resulted to serious environmental impacts that needs urgent attention [1,3,4]. Mining activities exposes sulphides bearing mineral such as Marcasite (FeS_2), Pyrrhotite (Fe_xS_x), Chalcopyrite ($CuFeS_2$) and Arsenopyrite (FeAsS) to oxidising conditions [1,5–8]. This has led to the formation of a very acidic and metalliferous mine drainage [9]. The formation of AMD using pyrite as an example can be denoted by the following equations [2].

$$FeS_2 + 3.5\,O_2 + H_2O \rightarrow Fe^{2+} + 2SO_4^{2-} + 2H^+ \quad (1)$$

$$Fe^{2+} + H^+ + \tfrac{1}{2}O_2 \rightarrow Fe^{3+} + \tfrac{1}{2}H_2O \quad (2)$$

$$FeS_2 + 3.75O_2 + \tfrac{1}{2}H_2O \rightarrow Fe^{3+} + 2SO_4^{2-} + 2H^+ \quad (3)$$

$$Fe^{3+} + 2H_2O \rightarrow Fe(OH)_2^+ + 2H^+ \quad (4)$$

$$FeS_2 + 3.75O_2 + 2.5H_2O \rightarrow Fe\,(OH)_2^+ + 2SO_4^{2-} + 3H^+ \quad (5)$$

$$FeS_2 + 3.75O_2 + 2.5H_2O \rightarrow Fe(OH)_{3(s)} + 2SO_4^{2-} + 4H^+ \quad (6)$$

These reactions are also mediated by micro-organisms [10–12]. As a result, the secondary drainage is acidic (pH $<$ 3) and rich in Sulphate,

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Fe, Al, Mn and traces of other constituents [3,13]. Acid mine drainage also contain high total dissolved solids (TDS) and electrical conductivity (EC) due to the dissolution of elements from the surrounding geology [12,14,15]. Toxicological and epidemiological studies revealed that elements in AMD are hazardous to living organisms on exposure [2]. For the interest of the public and environmental protection, this mine water needs to be treated prior to discharge to different receiving environments and compartments [16]. South Africa has advanced significantly in terms of mine water treatment and a number of its technologies have reached the pilot to implementation phase [17]. Mainly, they rely on the use of raw limestone and lime as pre-treatment options [18,19], and this have been successfully proven in mining industries. The limitation of limestone is partial removal of pollutants and increasing the pH to ≤ 7 [2]. Lime is very effective but its cost factor limits its application because it is generated from calcination of limestone [19]. Moreover, the generated sludge contains hazardous materials that can degrade the environment and pose serious health risk to aquatic and terrestrial organisms [20]. Also, environmental regulations do not deem lime a viable option [9]. However, Masindi [21], proposed a MASROE process which is similar to the proposed technology. His technology evaluated the treatment of AMD via sequential and fractional recovery of valuable resources such as pigments, hematite, geothite, magnetite, gypsum, limestone and drinking water.

There are two technologies that used for steel making and they include: the basic oxygen furnace (BOF) and the electric arc furnace (EAF). South African companies use the BOF route for steel making. In BOF process, iron ore, scrap iron, coke, oxygen and limestone are used as feed materials. The BOF production process happens when iron ore, coke and limestone are added on top of blast furnace while pre-heated as ingress from the bottom of the reactor. The hot air ignites the coke and result to the formation of carbon monoxide which reduces iron oxide to iron, in a similar manner, limestone is calcined to lime that react with impurities of silica [22]. Ca, Al, Mg, Fe and Si become the slag and they float on top of the molten iron where it is discarded on periodic basis [22,23].

BOF slag is a heterogeneous mixture of a large quantity of Ca, Si, P, S and Mn formed during the refining of the liquid steel [24–26]. These elements react with the dissolved CaO to form the slag. Poorly moltened Fe residues are also incorporated to BOF slag matrices [24]. Steel production process results in the production of large quantities of BOF slag [22,23]. Research studies proclaimed that, for every ton of crude steel produced, there is approximately 100–200 kg of BOF slag generated as a by-product. This may also be attributed to the feed material and catalysts been used [27,28]. After production, BOF slag is partially reprocessed and beneficiated, most steel plants dump this by-product to landfills, retention ponds and lagoons due to low market needs and demands. Beneficiation of BOF slag include: Road construction [29–33], soil amendments [34], water treatment [35] and metallurgical uses [34]. Its application in mine water treatment was in a lab-scale and the water quality was for discharge. This study therefore want to take its application further, to a large scale mine water treatment and produce the water that meet the drinking quality requirements and specifications.

Reverse Osmosis (RO) has been widely used in the treatment of wastewater emanating from mining houses, industrial and metallurgical process; natural water and point of use treatment modular [36,37]. However, this process has a disadvantage of producing large volume of brine that is expensive to dispose considering environmental regulations and requirement [12]. It also demands high energy intensity in highly polluted water [36,37]. Brine treatment technologies were also developed and the include freeze desalination and eutectic freeze [38–40]. These technologies require certain amount of pollutants for them to be effective [39,41]. Moreover, an RO system is very sensitive to fouling and scaling when subjected to highly concentrated solution and heavy metals [37]. This usually inflates the treatment cost thus requiring pre-treatment options to prolong the lifespan of a membrane and its efficiency. Several studies have opted for the pre-treatment of water prior purification by the RO [21,42–45]. Gashi, et al. [1] evaluated the removal of lead, zinc, cadmium and copper from effluents of battery industry, fertiliser industry, zinc electrolysis, sulphuric acid production and flotation plant by precipitation, adsorption and reverse osmosis. In their study, the authors reported that the rejection 91–98% rejection of heavy metals from treated effluents was achieved. Bouguerra, et al. [46] and Feng, et al. [47] also evaluated the treatment of acid mine drainage using a pre-treatment step and polishing in a secondary phase. The authors claimed that, this will aid in the pro-longing the life span of a treatment system. Pre-treating with soda ash will leave only Na for an RO feed hence making the RO circuit to much effective since it is very resistant to monovalent ions [37].

Several technologies have being developed for the treatment of mine water and they include passive [48–51], active [49,52] and integrated processes [21]. Due to the drawbacks in solitary technologies, research studies have being focusing on integrated and multi-disciplinary approaches for mine water treatment [18,21]. The principal idea is to complement the weaknesses in individual technology and come-up with a synergetic solution. The governing principle is that Fe and Mn which are present at high concentration in AMD are poisonous to the membrane materials on contact hence a pre-treatment option is required to remove these metals prior interacting with the membrane [53]. Lime and calcined cryptocrystalline magnesite can be used as pre-treatment agents to increase the pH of AMD and remove all the heavy metals [54] that have the potential to foul the membrane [37], however, this is not a very cost effective solution since BOF slag on the other hand, is a by-product produced in the steel industry which has the dual benefit in relation to other alkaline agents because (1) it can effectively increase the pH of AMD and remove all the metal species and (2) reduces the environmental footprint in the steel industry [35].

In this paper, the removal of inorganic contaminants from AMD by BOF slag, lime, soda and RO were meticulously embraced and evaluated. This novel study explored the chemistry of BOF slag, the resultant products and the quality of processed water thereof. BOF slag is rich in Fe and Ca hence making it much easier to recover magnetite and gypsum [55], considering that AMD is rich in Fe and sulphate. BOF slag will remove pollutants from acid mine drainage by dissolving Ca and Mg into solution hence increasing the pH of the resultant water (Eqs. (7)–(10)).

$$CaO + H_2O \rightarrow Ca(OH)_2 \tag{7}$$

$$CaO + H_2O \rightarrow Ca(OH)_2 \tag{8}$$

$$Ca(OH)_2 \rightarrow Ca^{2+} + OH^- \tag{9}$$

$$Mg(OH)_2 \rightarrow Mg^{2+} + OH^- \tag{10}$$

An increase in pH will lead to the precipitation of metal species (Eq. (11)).

$$M^{n+} + nOH^- \rightarrow M(OH)_n\downarrow \tag{11}$$

Lime and soda ash will be used to reduce the water hardness and to remove the residual pollutants, i.e. calcium as gypsum and magnesium as brucite whereas soda as will remove calcium as limestone and hydrated lime. The removal mechanisms of inorganic contaminants will be investigated by the chemical composition of the feedstock and the resultant sludge after each stage. RO system will then be used to upgrade the produced water to meet drinking water quality standard as stipulated by the South African National Standards (SANS 241).

2. Materials and methods

2.1. Feedstock

Basic Oxygen Furnace (BOF) slag was collected from a Harsco Metals and minerals industry in South Africa, Pty (Ltd). Commercial