Experimental View of the Sub-Tropical Clayey Soils-Acid Mine Drainage Interactions

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Abstract— Issues of acid mine drainage (AMD) from mining activities is a lingering phenomenon to the present day developing South Africa as well as in most developed countries around the globe. However, the insistent rise in environmental contamination in South Africa is drastically attracting massive concerns. Vital concerns of AMD in South Africa still remain the threat to soil, surface, subsurface and ground water reserves among others, which are known to consequentially affect human and environmental health. This persistent challenge has given rise to the need for investigating the buffering efficacy of clayey mineral soils for use as natural contaminant barriers to pollutant species from AMD. Also, increasing climatic changes, growing population and depletion of natural resources from mining and related operations have further increased these environmental challenges. However, the constantly growing challenges gave bearing for this study to experimentally outline series of vital tests on three subtropical clayey soils infused with AMD for the determination of their mineral buffering efficacy towards potential use as natural containment liners in areas affected by AMD. Series of dissolution, diffusion and water sampling analysis were initiated herein on the respective soil systems leading to progressive investigations and analysis for decisive interpretations in subsequent studies. In summary, the general outcome was below the accepted criterion specified for clay liner construction in South Africa. However, the tested samples can be incorporated into the design and construction of composite barriers for acid-generating wastes.

Keywords—*Batch Sorption, Contaminants, Acid Mine Drainage (AMD), Clayey Soils, Diffusion*

I. INTRODUCTION

ACID mine drainage (AMD) occurs in sulphide-bearing mine waste due to the oxidation of iron sulphides, especially pyrite (FeS_2), pyrrhotite (FeS) and marcasite (FeS_2) causing the creation of several soluble hydrous iron sulphates, the generation of acidity and subsequent leaching of metals.

AMD have several negative human and environmental impacts [1-4] which may be alleviated by lining possible acid-generating waste rock or mine tailings impoundments with clayey or composite clayey liners. For the competent performance of these liners, it is important that their properties are not compromised over time when exposed to AMD. As such, assessment of the chemical interaction and compatibility of clay/clayey liners with aggressive AMD is a pertinent consideration in the proper management of potentially acid generating mine waste [5]. These AMD-soil interaction studies may involve dissolution, diffusion, water sampling, percolation testing, contaminant migration analysis and determination of soil chemical transformations. Although, some studies on the interaction of soils from temperate regions with AMD have been conducted [6-8] there still remains however, insufficient data on the interactive outcomes/compatibility of soils from tropical and subtropical regions with AMD. More to this, sufficient outcomes involving some key tests i.e., dissolution, diffusion etc., were not reported in most of these works which nonetheless, created a path for this study to bridge that knowledge gap. AMD usually has high acidity and is rich in sulphate and potentially toxic elements and heavy metals.

The rate of AMD generation in an environment where sulphidic material is exposed to oxygen and water, will be determined by a number of factors. The reactions will proceed rapidly in warm humid environments, while other environmental factors, such as the specific sulphide mineralogy and grain size, and the presence or absence of acid neutralizing minerals, such as carbonates will also influence the rate of AMD production and its eventual environmental impact. The major source of groundwater contamination by mine tailings have been suggested to be AMD [9]. The environmental impact of AMD as reviewed by [10] include; metal toxicity, sedimentation processes and high acidity, which usually have the potential to cause interrelated negative impacts on terrestrial and aquatic life. Since AMD has potential negative impacts on the environment, efforts are made to either prevent or control its generation and migration. As such, several measures and technologies are available as recorded by [11-14] for mitigating the environmental effects of AMD. These include co-disposal of waste rock and mine tailings and the construction of wetlands, base liners or water covers.

Soil covers and liners may also be used as hydraulic barriers for containing AMD leakage at mine sites. When used for this purpose, it is usually pertinent to determine whether the soil liner can buffer contaminants or is compatible with AMD. Consequently, several studies have been conducted on the compatibility of some soils from temperate regions with AMD

[15-18]. However, as earlier pointed out, there is need to investigate the interaction of AMD with tropical/subtropical soils as there is insufficient data and knowledge in this area of AMD-soil compatibility interest whose gap requires bridging. South Africa's mining history has created vast economic benefits and still plays an important role in safeguarding the country's position in the global eyes. Despite such benefits, mining sector operations as elsewhere in the world, have resulted in serious environmental consequences, notably in respect of poor environmental and water management and, in the case of the gold mines of the Witwatersrand, AMD. Pyrite (FeS_2), sphalerite (ZnS), galena (PbS), arsenopyrite (FeAsS) and occasionally chalcopyrite ($CuFeS_2$) are associated with gold mineralization in operating gold mines in South Africa [19-21]. Mining activities and processing from these sulphidic ores have led to several tonnes of mine waste that oxidize and lead to the widespread generation of AMD in and around areas where such operations occur in South Africa.

AMD is known to occur in a number of areas in South Africa, mostly in the Witwatersrand Gold Fields, the country's various Coal Fields and the O'Kiep Copper District. Thus, potable sources of water in mining areas, some of which are densely populated, are severely at risk due to AMD contamination. The key factors which differentiate the developing problem in South Africa from international scenarios is the degree of interconnection of large voids, the sheer scale of the Witwatersrand operations and the fact that many of the problem areas are located in or close to major urban centres. Most mining centres are also covered by the natural habitats of some rare and endangered species. Bioaccumulation of metals from AMD adversely affects flora and fauna in South Africa and this has led to the loss of food sources and elimination of some sensitive species [22-25]. The surface flow or decant of mine water is also of concern to the environment as the water, in accordance with well-known and researched chemical and geochemical reactions between the mine rock strata, wastes and oxygen, readily becomes acidic, characterized by elevated concentrations of salts, heavy metals and radionuclides. The immediate concern as recorded by [26] is the threat posed by decanting mine water in the Witwatersrand area, which poses potentially severe environmental and safety impacts on the receiving water environment and associated surface areas. In this light, mitigating the effects of AMD contamination of the South African mine environment is imperative. Currently, with the sulphidic ore exploitation in South Africa, mine tailings dams and acid water containment ponds are being constructed for the storage of mine waste being generated from mining operations, and assessment of the suitability of local construction materials is therefore essential. In an earlier stage of this study, the chemical and geotechnical properties, and the mineralogical composition of three sampled soils, prior and subsequent to AMD percolation were determined and the results were presented.

However, the study herein experimentally outlines the clayey soils-AMD interaction with respect to vital or prerequisite approaches in the forms of dissolution, diffusion and water sampling analysis on AMD permeation through three compacted clayey soils sampled around the City of Johannesburg (CoJ), South Africa, where waste disposal activities occur. To assess the impact of these contaminant migration processes on the buffering efficacy and performance of the respective soils as lining materials, these key tests were prime and are subsequently discussed herein. The outcomes forms part of a larger progressive study towards very decisive analysis and interpretations of the soils-AMD interaction for the determination of their buffering efficacy for potential use as natural earthen lining materials (locally available clayey mineral liners) for the containment of mine waste and associated pollutants.

II. RELATED BACKGROUND

The contaminant mass transport mechanisms in natural and compacted clayey soils have been extensively investigated by several researchers over time [27-32]. Compacted clays usually have low permeability and as such, groundwater flow through them is usually slow. Consequently, the mass of solute transport by flowing contaminating solutions (advection) is likely minute, whereas it is usually significant in solute transport due to concentration gradients (diffusion). As such, dissolution, diffusion and percolation tests are commonly conducted to determine retardation factors, effective diffusion coefficients and hydraulic conductivities respectively. Hence, the study found the need to experimentally assess the principles involved in dissolution, diffusion and water sampling analysis of the transport mechanisms towards determining the buffering efficacy of the respective sampled subtropical clayey soils.

III. DISCUSSION OF FINDINGS

A. Dissolution Tests

To determine the dissolution degree, S_o, or distribution coefficient, K_d, of potassium, sodium, zinc, nickel, cobalt, chloride and sulphate the batch sorption test was conducted in conformance to [33]. The dissolution degree, S_o, is a measure of the extent to which the AMD permeant dissolves contaminant species from the respective soils. However, for species that were sorbed on the soils from the AMD permeant, the distribution coefficient K_d, was then determined from the batch sorption tests. Before the sorption tests were conducted, representative soil samples were air-dried, disaggregated and passed through the No. 40 sieve. 20 mm of AMD was added to 5 g of sieved soil in plastic centrifuge bottles to obtain a soil: AMD ratio of 1:4. The use of AMD in place of single ion solutions was to allow for potential competitive sorption between ions for available soil exchange sites.

The tests were conducted at laboratory temperature of 22 ± 2°C and with soil-AMD interaction times of 5, 10 and 15 days before been agitated in a ball bearing electric laboratory shaker. For each 5 day quota of the interactive period, the mixture was agitated for 16 hours, meaning that, for batch sorption tests lasting over 10 and 15 days, the mixtures were agitated for periods of 32 hours and 48 hours respectively. Interaction times of 3, 7 and 11 days were reported by [34] however for the purpose of this study, longer soil-AMD interaction times were pursued. Agitation was terminated a day prior the expiration of the interaction/contact time to allow the mixtures to settle. For instance, a test with interaction time

of 10 days meant the mixture was agitated for a total period of 32 hours, 16 hours within the initial 5 days and another 16 hours of agitation from day 6 to day 9. Agitation did not take place on day 10 of the interaction time to allow for settlement of the mixture. On expiration of the interaction times, each mixture was centrifuged at 10,000 g for 20 minutes in a high speed centrifuge. The supernatant was then decanted and sampled for chemical analysis. The distribution coefficient, K_d, of each nominal contaminant species was determined using the relationship in (1).

$$K_d = \frac{C_i - C_f}{R} \times \frac{V_s}{M_a} \qquad (1)$$

Where: C_i = Initial concentration of species in leachate (mg/L); C_f = Concentration of species in decanted solution at the expiration of test (mg/L); V_s = Volume of leachate added to the soil (mL); and M_a = Mass of air-dried soil (g).

The contaminant species retardation factor expression as discussed in a previous study was then used to calculate the retardation factors, R, of the various species. More to this, the relationship expressed in (2) was used to calculate the dissolved amount of a species (dissolution degree, S_o) per unit area of air-dry mass of soil.

$$S_o = \frac{C_f - C_i}{C_i} \times \frac{V_s}{M_a} \qquad (2)$$

Where: S_o = Dissolution degree (mL/g).

B. Diffusion Tests

For the soil porewater concentration profiles of the chemical species diffusing through the saturated soils to be obtained, the diffusion test was performed. The diffusion cell used for the study conformed to the bespoke device used by [34] which comprised of a cylindrical stainless steel mould with an internal diameter of 54 mm and a height of 70 mm. It had a stainless steel base plate and top cap fitted tightly together by steel rods and screws. Grooves were created in the top and bottom of the cylinder for placement of O-rings and silicon sealant to keep the top and bottom caps tightly sealed to the cylindrical mould. Two ports were provided in the top cap. The first port held a 9 mm diameter Teflon stirring rod with paddle connected to a low-speed stirrer. While the second port meant for periodic sampling of 1 mL of permeate from the chamber for ion analysis during the test had a diameter of 2 mm closed with a screw cap.

Before the laboratory experimental test, an approximate duration of the test for the nominal ions was determined by using the relevant soil parameters and values of diffusion coefficients based on literature for species of interest inputted in *POLLUTEv6*, a computer program that implements a solution to the one-dimensional advection-dispersion equation by [35]. Fig. 1 shows the schematic of the bespoke diffusion cell culled from [34]. A Harvard miniature compactor was used to directly compact the soil samples in the diffusion cell. The thickness of the compacted specimen was trimmed to about 40 mm and the height of permeant chamber was 30 mm. A water reservoir was created on top of the compacted soil for a few days to increase the degree of saturation after which the water was then carefully emptied out and replaced with AMD. The duration of the test was 14 days and AMD (source

solution) was continuously stirred at 5 rev/min throughout the test. Table I summarizes the properties of the compacted soil used for the diffusion tests.
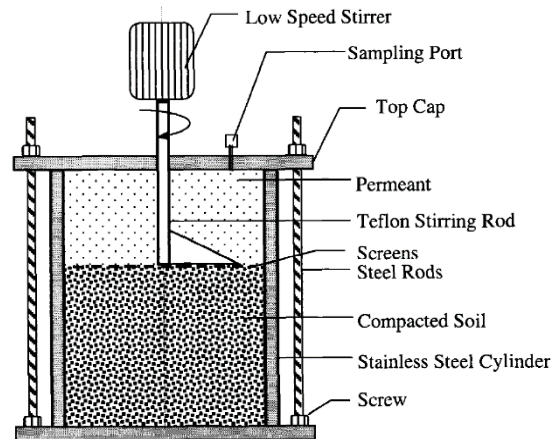

Fig. 1. Schematic of the bespoke diffusion cell

TABLE I
Summarized properties of the compacted soils initiated for the diffusion tests

Properties	ASTM Design.	Soil A	Soil H	Soil K
Natural Water Content (%)	D 2216	20	32	27
Water Content (Air-dry soil) (%)	D 2216	4.7	1.3	1.4
Optimum Water Content (%)	D 698	16.2	15.4	15.7
Bulk Unit Weight (kN/m³)	-	18.52	19.11	19.97
Compaction Water Content (%)	-	15.91	15.15	15.58
Initial Degree of Saturation	-	81.9	79.8	81.3
Final Degree of Saturation	-	99.5	99.7	99.4
Degree of Compaction (%)	-	99.1	98.9	99.3
Dry Unit Weight (kN/m³)	-	15.15	16.37	17.33
Moulded Dry Unit Weight (kN/m³)	-	15.13	16.35	17.31
Wet Unit Weight (kN/m³)	-	17.7	18.9	20.1
Void Ratio	-	0.76	0.77	0.74
Porosity	-	0.45	0.44	0.43

On termination of the diffusion test, the permeant chamber was carefully emptied out of the cell and sampled for ion analysis. The soil sample was then extruded from the mould and sectioned into nine approximately equal thicknesses. The moisture content was determined for each sectioned soil sample. Other portions were then squeezed with a pneumatic porewater squeezer to obtain soil porewater for chemical analysis. In a case where the post-diffusion water content was insufficient to generate enough volume of porewater for chemical analysis, a saturation extract was obtained and the measured concentrations were adjusted for dilution.

C. Analysis of Water Sample

As already discussed in an earlier study, while measuring the hydraulic conductivity of compacted specimens of the soils using a constant-head rigid wall bespoke permeameter device, effluents were also collected for chemical analysis in a 500 mL graduated cylinder over the test duration. Soil porewater, influent permeant and the effluents obtained during hydraulic conductivity testing were acidified by high-purity nitric acid and analyzed for cationic or heavy metal concentrations using inductively coupled plasma-atomic emission spectroscopy (ICP-AES). To accommodate for any

reagent or procedural contamination, blanks of reagent and de-ionized water were analyzed with each batch of samples. The pH and electrical conductivity of water samples were measured in triplicate with appropriate probes and the average values were recorded. Chloride and sulphate were the main anions considered during the study. These anions were detected through ion chromatography with Waters IC-Pak A Column using Borate/Glauconate Eluent. The device comprises of stainless steel anion columns that separate and quantify the anions at low (ppb) levels. Pre-test sample preparation included dilution and filtration. Fresh deionized megapure (18 megaohm) water, less than 4 hours old, was drawn and used in the preparation of standards and the dilution of samples before analyzing with the Waters 432 Conductivity Detector. This was to minimize possible contamination due to exposure to atmospheric pollutants in accordance to [36].

To prevent the precipitation of salts in the column due to the injection of samples with high concentrations of ions, proper sample dilution before the injection into the column was done with fresh megapure (18 megaohm) water. Calibrated samplers were used to collect required quantities of test sample and megapure water to obtain the diluted sample for the analysis. The samples were filtered using 0.45 µm pore size plastic syringe filters to prevent clogging of the analytical column system or its peripherals and the excessive build-up of pressure due to the particulates in the samples. The samples were introduced into the Millitrap cartridge via Luer-lock syringes with bulk of the test conforming to test procedures outlined by previous authors [37], [8], [34], [38]. The filter was rinsed at least two times with megapure water and then sequentially, 20 mL of the sample was filtered to waste. In accordance to [36] the samples were then injected at the recommended flow rate of 1.2 mL/min and at a pressure of 2.41 MPa (350 psi) which was in range with the recommended pressure threshold of 6.89 MPa (1000 psi).

IV CONCLUSIONS

The study depicts an experimental view of and assessment of three subtropical clayey soils interacted with AMD towards determining their mineral buffering efficacy for use as natural containment liners in areas under AMD attack. From sequential prolonged testing on the respective soil systems, pertinent tests leading to more decisive interpretations and analysis in succeeding studies were achieved. The species concentrations obtained from either the effluents or the solution and sections of soil samples obtained from dissolution and diffusion tests suggested that the soils-AMD interaction is a complex process. The results of the study suggest that interactions between AMD and the soils plausibly resulted in dissolution of metals from soil grains, desorption of adsorbed contaminant species at the exchangeable sites of the soils and, alteration and dissolution of soil minerals. Hence, in consideration of the general outcomes of the study with further interpretations and analysis in succeeding studies, the three sampled natural subtropical soils were found to be incompatible with AMD which invariably may not be utilized solely as natural clayey mineral lining materials towards AMD containment disposal. However, may be considered for use in the construction of composite barriers for containing acid-forming waste bodies.

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