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Mixed Layer Sub-Tropical Clayey Soil-AMD Percolation: Analysis of Permeation Limits and Breakthrough Patterns

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Abstract

The increasing population growth, depletion of natural resources from mining and similar activities, and consequent insistent climate change have triggered diverse environmental challenges. It is well known that acid mine drainage (AMD) from mining operations in South Africa constitutes a serious environmental problem which requires heightened and crucial attention. The contamination threats from AMD presents a severe risk requiring prevention and mitigation measures for curbing consequential impacts on environmental and human health. This amassing challenge gave the study the impetus to investigate the permeation limits and breakthrough curves of a mixed layer subtropical clayey soil; kaolinite, halloysite and illite/vermiculite permeated with AMD towards establishing its mineral attenuation effectiveness for use as natural barrier against contaminant species in areas under AMD attacks. As such in this study, successive protracted permeation up to 24 pore volume passage of AMD through the soil medium was conducted using a laboratory scale bespoke column hybrid permeameter device. The results of the study showed that after approximately 2.4 pore volumes of 0.01N CaSO₄, a baseline hydraulic conductivity of 1.3×10^{-11} m/s was obtained for the soil which remained unchanged even after 19 pore volume of AMD seepage through the soil. The value was found to be below the accepted criterion of 1×10^{-9} m/s specified for clay liner construction in South Africa but may as well be considered for use as a natural material in the construction of composite barriers for acid-generating wastes.

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1. Introduction

Acid mine drainage (AMD) is generated in mining areas when ore and waste materials, containing sulphide minerals such as pyrite, become exposed to water and oxygen. Alternatively, when some mine tailings from processed

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sulphide-rich ores are oxidized, AMD may be formed. However, in South Africa, several research investigations on AMD generation, migration, and consequent effects, particularly in the Witwatersrand have been performed by the Council for Geoscience, on behalf of the Department of Mineral Resources [1]. The turbulence of ore bodies and the transport of large volumes of pyritic material to the surface create conditions highly favourable to the formation of AMD, due to both the exposure of ore in the underground environment and the transport of blasted and/or crushed waste rock and tailings to the surface. Hence, for pyrite to become oxidized, both oxygen and water must be present, in which case water serves not only as a reactant but also as a reaction medium and a product transport solvent [2]. The rate of AMD formation in any area where sulphidic material is exposed to oxygen and water will be determined by a number of features. 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 dictate the rate of AMD generation and its consequential environmental effects. Nevertheless, the foremost source of groundwater contamination by mine tailings have been suggested to be AMD [3]. Some of the environmental effects of AMD studied by [4] include but are not bound to; metal toxicity, sedimentation processes and high acidity, which usually have the potential to cause interrelated negative impacts on terrestrial and aquatic life. Considering that AMD holds potential hazardous human and environment health implications, significant efforts are being employed by researchers to either prevent or reduce its formation and migration to points with substantial impacts. As such, several processes and technologies have been made available as recorded by [5-7] for mitigating the environmental effects of AMD. Some of these measures include the design and construction of lining barrier or water covers, co-disposal of waste rock and mine tailings and the construction of wetlands. Soil covers and liners may also be utilized as hydraulic barriers for containing AMD leakage at mine sites. When employed for this purpose, it is usually vital to establish whether the soil liner can attenuate myriad contaminant species or is compatible with AMD. Conversely, a number of studies have been conducted on the compatibility of some soils from temperate regions with AMD [8-10]. Nonetheless, it still remains pertinent that an investigation into the interaction of AMD with tropical/subtropical clayey soils be conducted as there still remains a significant gap in knowledge and insufficient data available in this area of AMDsoil compatibility concerns whose trench requires filling. The history of mining in South Africa reveals enormous economic benefits which still continually plays an important role in preserving the country's position in the global market. In light of these beneficial contributions, the mining sector operations in South Africa similar to the mining activities in other countries around the globe, have ensued in severe environmental consequences, regarding the notably poor environmental and water management, and in the case of the gold mines of the Witwatersrand AMD.

Nomenclature

Acid mine drainage (AMD)	the outflow of acidic water from metal mines or coal mines
Contamination	the action or state of making or being made impure by polluting or poisoning
Percolation	refers to the movement and filtering of fluids through porous media

As recorded by [11-13] pyrite (FeS_2), sphalerite (ZnS), galena (PbS), arsenopyrite (FeAsS) and occasionally chalcopyrite (CuFeS_2) are concomitant with gold mineralization in operating gold mines in South Africa. The continuous mining activities and processing of these sulphidic ores have over the years led to the oxidization of several tonnes of mine waste thereby causing the widespread formation of AMD in and around environments where such operations are prevalent in South Africa [7]. The Witwatersrand Gold Fields, the various Coal Fields and the O'Kiep Copper District are some of the areas where AMD is mostly known to occur in South Africa. Consequently, potable sources of water in mining areas, some of which are heavily populated, are critically at risk due to AMD contamination. Some of the key factors which distinguish 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 zones are located in or close to major urban areas. Most mining sites are also covered by the natural habitats of some rare and endangered species. Bioaccumulation of metals from AMD harmfully affects flora and fauna in South Africa and this has led to the loss of food sources and elimination of some sensitive species [14, 15]. 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 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 [6, 16]. At present, 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 locally available construction materials is therefore essential with a view to cost reduction, resource conservation and environmental sustainability. In previous stages of this study, certain properties and composition of the sampled soil, before and after AMD percolation were determined and the results presented. However, this study examines the migration of AMD through a mixed layer kaolinite, halloysite and illite/vermiculite compacted clayey soil, sampled around the City of Johannesburg (CoJ) where waste disposal activities take place. To evaluate the effect of contaminant migration processes on the attenuation effectiveness and performance of the soil as lining material, a bespoke column hybrid permeameter device was used in the permeation process of the soil with AMD at low flow rates typical of field conditions. The pH, electrical conductivity and solute breakthrough curves from AMD percolation were examined as well as batch sorption and diffusion tests. The experimental data gathered from this investigation, forms part of a broader study to characterize a number of solute transfer processes as AMD interacted with the sampled mixed layer designated as Soil A towards potential utilization as natural and locally available mineral liners for the containment of mine waste and allied contaminants.

2. Experimental process

The contaminant mass transport mechanisms in natural and compacted clayey soils have been extensively studied by several authors from past to recent times [17-24]. Sorption, diffusion and hydraulic conductivity tests were performed to determine retardation factors, effective diffusion coefficients and hydraulic conductivities respectively. Diffusion may be the dominant contaminant transport mechanism in clayey soils and as such, laboratory diffusion testing was carried out as a routine approach in the assessment of the tested clayey samples. The advection-dispersion expression by [25] was used to calculate the one-dimensional solute transport through the homogeneous, isotropic compacted clayey Soil A sample as expatiated in a previous investigation.

3. Results and discussion of finding

3.1. Analysis of effluent breakthrough patterns

The initial AMD composition, soil porewater composition and the cation exchange reactions largely affect the effluent composition. This is due to the interactions between cations in AMD and the cations at the soil exchange sites, and the impact of AMD on the crystal lattice of soil minerals. As such, the effluent pH, electrical conductivity and solute breakthrough patterns observed as a resulting influence of AMD percolation through the Soil A medium is generally used to examine the properties and composition of the effluents.

3.2. pH and electrical conductivity

The relative effluent pH breakthrough pattern showed an increase in pH of the effluent as against the pH of the influent (AMD) prior percolating roughly 4 pore volumes of AMD through the compacted soil as shown in Fig. 1(a). The pH of porefluid of the natural soil and 0.01N CaSO₄ initially percolated through the soil was greater than the pH of the AMD influent of 2.8. This plausibly resulted in the initial increase in effluent pH on percolating the soil with AMD. A sharp decrease in effluent pH after only a few pore volumes of AMD percolation through the Soil A suggested a low acid attenuation effectiveness. A plausible explanation by [26] records that organic matter and carbonate content of soils, are soil constituents known to possess high pH attenuation efficiency. However, the soil used in the study was found to have fairly low organic matter and carbonate content which may have perhaps contributed to its poor behaviour in acid attenuation. Beyond 4 pore volumes of AMD percolation, the acid attenuation effectiveness of the soil had neared depletion, thus, effluent pH relative to influent pH approached unity.

The effluent pH value of the Soil A was observed to increase after only 1 pore volume of AMD percolation after which a steep decrease in effluent pH was observed until 4 pore volumes of AMD percolation through the soil. Generally, an increase in the relative electrical conductivity of effluent with increasing pore volumes of AMD percolation through the soil was observed. The increase in relative electrical conductivity of the effluents was consequently ascribed to the increasing concentrations of ions, particularly aluminium, iron, magnesium, calcium, sodium and potassium in the effluents resulting from AMD percolation through the soil. Fig. 1(b) shows the relative effluent electrical conductivity (EC_e/EC_i) breakthrough curve obtained from the percolation of Soil A with AMD. The relative effluent electrical conductivity breakthrough curve is a weighted contribution from the influent ionic composition and the ions originally present at the soil exchange sites. Hence, the electrical conductivity of a fluid depends on the number and type of ions contained in the fluid. The initial increase in Soil A effluent pH was possibly due to mineral alterations caused by the subsequent AMD percolation while approaching equilibrium pH after 20 pore volumes. The considerable increase observed in the effluent pH values after 11 pore volumes of AMD seepage through the Soil A could not be accounted for as this was unclear from the results. However, a similar behaviour was also recorded by several authors [27, 9, 17]. Correspondingly, it has been shown that electrical conductivity is an indicator of the ease with which fluid carries an electrical current [28]. Similarly, it is expected that close to 1 pore volume of permeant percolation, relative electrical conductivity of 0.5 would be reached [29].

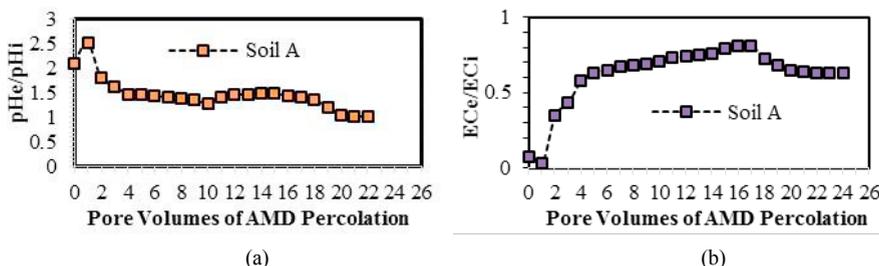
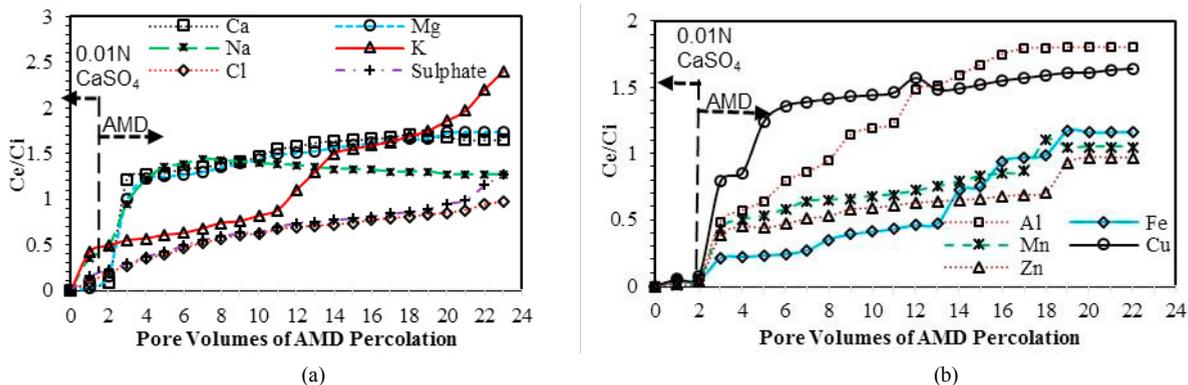


Fig. 1. (a) Effluent breakthrough pattern for pH of tested Soil A; (b) Effluent breakthrough pattern for electrical conductivity of tested Soil A

This was not the case in Soil A as the relative effluent electrical conductivity value of 0.5 was reached after passing around 4 pore volumes of AMD through the soil. Considering the low percolation rates ensured throughout the study, a decrease in the relative electrical conductivity of Soils A was observed after 19 pore volume of AMD passage. This could not reasonably be accounted for from the composition of effluents as well as from related literature, however, an exception was found in a study by [27] who recorded a similar observation on passage of 15 and 16 pore volumes of AMD through a media of tropical like soils.

3.3. Soil A Solute-solvent analysis

Soil A appeared to have been severely impacted by AMD permeation on termination of the tests. The concentrations of almost all the monitored heavy metals and nominal contaminant species in the effluents from the soil were higher than the influent concentrations as shown in Figs. 2a-c.



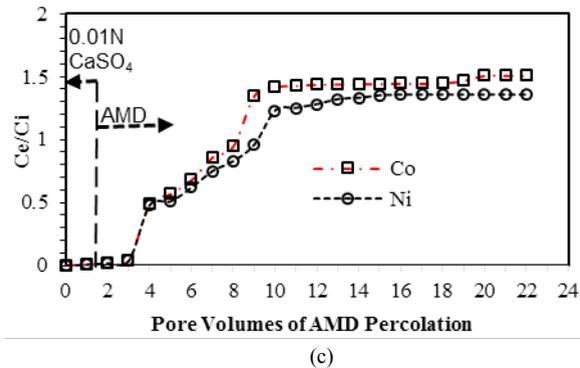


Fig. 2. (a); (b) and (c) Effluent breakthrough patterns from Soil A solute-solvent analysis

This indicated the release of species like iron, aluminium, calcium and magnesium due to the dissolution of gibbsite, diaspore ($\text{Al}_2\text{O}_3\cdot\text{H}_2\text{O}$), magnesioferrite and hydroxy apatite ($\text{Ca}_5\text{OH}(\text{PO}_4)_3$) by AMD in Soil A. Furthermore, the concentration of magnesium in the effluent was found to increase and since magnesium occurs chiefly in the octahedral sheets of clay minerals, it suggested the preferential dissolution of the octahedral sheets of the clay minerals existent in Soil A. Also, previous studies on mineralogical and equilibrium speciation studies indicated partial alteration of the illite/vermiculite mixed layer mineral to discrete illite and vermiculite phases after solute-solvent interaction (Soil A-AMD percolation). Furthermore, calcium, magnesium and sodium were the dominant cations in the exchange complex of Soil A as earlier observed. Notwithstanding, the observed slight desorption peaks of calcium and sodium shown in Fig. 3a indicated that these were the two species dominantly involved in ion exchange reactions with chemical species existent in the AMD permeant. Hence, from the effluent solute breakthrough patterns there was clear indication that the attenuation effectiveness of Soil A was low. Therefore, in a case where AMD of similar composition as used in this study interacts with a barrier built with Soil A, the integrity of the barrier would be compromised.

3.4. Contaminant species relative attenuation and elusion

The procedures outlined by [30] was initiated in this study in determining the relative attenuation number (ATN) for each effluent contaminant species representing the relative mobility of the species through the respective compacted soils. Table 1 shows the calculated ATN of each effluent species monitored during soil-AMD percolation tests using the bespoke constant-head rigid column hybrid permeameter. The approach entailed calculating the area under the breakthrough pattern of each monitored contaminant specie and expressing the difference between this area and the total area (rectangular area bounded by the initial and final pore volumes of AMD passed and the relative solute concentrations of zero and one) as a percentage of the calculated area under the breakthrough pattern of the species.

Table 1. Relative attenuation (ATN) of effluent species from soil-AMD percolation tests.

Soil Designation	Contaminant Species	ATN*	QAC**
	Cl	35.7	M
	SO ₄	20.4	L
	Fe	10.3	L
	Zn	4.1	L
	Mn	-0.42	N
	Ni	-0.79	N
	Co	-13.3	N
Soil A	Na	-20.2	N
	Al	-27.5	N

Cu	-28.3	N
Mg	-37.9	N
Ca	-51.1	N
K	-57.4	N

*ATN: Relative Attenuation Number; **QAC: Qualitative Attenuation Classification- H = High attenuation; M = Moderate attenuation; L = Negative attenuation or Elution

The higher the ATN obtained for a given species present in the effluent the less relatively mobile the species is through the compacted soil while a negative ATN value indicated elution (desorption) of species from the soil. A qualitative description of the attenuation of species was done using the following classification: $0 < \text{ATN} < 30\%$ \equiv low attenuation; $30\% < \text{ATN} < 75\%$ \equiv moderate attenuation; while $\text{ATN} > 75\%$ \equiv high attenuation. From the series of tests conducted, Soil A was observed to moderately attenuate chloride and sulphate from AMD. The ATNs showed that the exchangeable cations; sodium, calcium and magnesium eluted the soil in various levels into AMD.

3.5. Analysis of effluent breakthrough patterns

The results from the dissolution tests indicated that over the soil-AMD test period, sodium, nickel, cobalt and sulphate were dissolved from Soil A by AMD as shown in Table 2. This was however, found to have similar trends with studies by several authors [31, 27] who conducted batch sorption tests using soils from tropical regions under similar conditions with the amounts of cobalt dissolved from the soils by AMD being in ranges of 2.7 to 4.1 mg/L.

Table 2. Concentration of contaminant species over the dissolution/batch sorption tests.

Sample	Test period (days)	Concentration of contaminant species (mg/L) ^{a, b}						
		K	Co	Na	Zn	Ni	Cl	SO ₄
AMD		4.7	7.5	27.9	161.7	181.3	200.6	5075.4
Soil A	5	2.4	7.8	42.3	192.1	235.5	179.6	8856.6
	10	2.6	9.6	42.7	199.3	239.7	174.4	8859.8
	15	2.7	10.9	43.5	201.7	243.3	173.9	8872.3
		Distribution coefficient/dissolution degree (mL/g) ^{a, b}						
Soil A	5	2.9	1.4	1.7	0.7	1.4	0.4	2.5
	10	2.6	1.7	1.9	0.7	1.5	0.6	2.6
	15	2.8	2.1	2.0	0.9	1.6	0.6	2.7

^a Temperature: $22 \pm 2^\circ\text{C}$; ^b Soil: Solution ratio (1:4)

It was further observed that AMD dissolved zinc from Soils A yet was found to have adsorbed potassium. Generally, small amounts of chloride were either adsorbed or dissolved from the soil over the percolation successions of the tests. Consequently, the most substantially extracted metals by AMD from the soil were cobalt and nickel with the soil observed to adsorb potassium and chloride. Generally, the impact of AMD on Soil A during the batch tests over various interaction times was observed to be closely identical. During the batch tests, equilibrium conditions were attained which was evident from the data in Table 2 showing the effect of various interaction times on the insignificant amount of contaminant species dissolved from the soil by AMD.

3.6. Diffusion Tests

The outcomes for Soil A obtained from the diffusion tests on the time-dependent concentrations of chemical species in the chamber of the AMD solution during the diffusion tests and the depth-dependent species soil porewater concentrations at the end of the tests is shown in Figs. 3a-d.

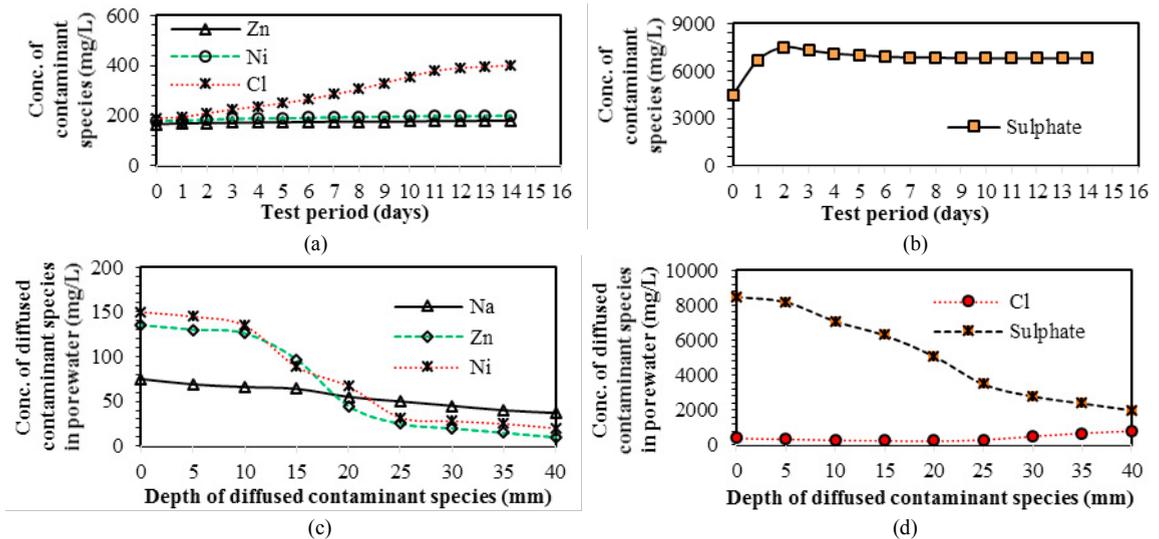


Fig. 3(a); (b); (c) and (d) Depth-dependent chemical species conc. in porewater of Soil A during diffusion test

Out of the seven contaminant species monitored during the diffusion tests, results essentially revealed the concentrations of sodium, zinc, nickel, sulphate and chloride in the permeant chamber (containing AMD) to increase with respect to time and inclined to reaching equilibrium values towards the end of the diffusion tests.

4. Conclusions

This investigation examined the attenuation effectiveness of Soil A- a mixed layered subtropical soil of kaolinite, halloysite and illite/vermiculite sampled from around the CoJ towards its efficient and effective use as a natural mineral liner for AMD containment and mitigation in areas affected by mining pollution. Beyond at least 19 pore volumes of AMD percolation through the soil, the measured soil hydraulic conductivity value was 1.3×10^{-11} m/s. This final hydraulic conductivity value fall short of the maximum soil hydraulic conductivity acceptance criterion of 1×10^{-9} m/s specified for clay liner construction in South Africa. The pH breakthrough curve revealed that the soil has low attenuation efficiency to acids which in turn failed to sufficiently attenuate potential contaminant species present in AMD. Furthermore, the solute breakthrough patterns clearly showed that the soil was not effective in attenuating the heavy metals and the other contaminant species present in AMD. The species concentrations obtained from either the effluents from the hydraulic conductivity tests or the solution and sections of soil samples obtained from dissolution and diffusion tests suggested that the soil-AMD interaction is a complex process. The results of the study suggest that interactions between AMD and the soil conceivably resulted in dissolution of metals from soil grains, desorption of adsorbed contaminant species at the exchangeable sites of the soil and, alteration and dissolution of soil minerals. Consequently, considering the general outcomes of the investigation, the Soil A- mixed layered natural subtropical soil of kaolinite, halloysite and illite/vermiculite was found to be incompatible with AMD and thus, may not be suitable alone as natural clayey mineral liner towards AMD containment or disposal.

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