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Valorization of South African Coal Wastes through Dense Medium Separation

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Abstract: Sustainable management of coal waste is one of the major environmental concerns for coal mining, whether active or legacy, worldwide. Coal dump deposits demand a large physical area or footprint for disposal of solid waste, change the topography, and generate both pyritic dust and acid rock drainage (ARD) where pyritic coal waste is deposited. The beneficiation of dump deposits or, preferably, of coal waste prior to its dumping can reduce or even eliminate the liabilities related to coal waste management. In this work, dense medium separation studies of coal discards, using heavy liquids, resulted in three pooled fractions from typical South African coal waste discards from the Mpumalanga region for future use: (a) a fraction of low density with increased calorific value; (b) a fraction of intermediate density, rich in ash and acid neutralizing minerals and lower in sulfur; and (c) a fraction of high density, rich in sulfidic minerals including pyrite. The fractions were characterized using particle size analysis, sink-float studies, static tests to predict ARD potential, proximateand ultimate analysis, and gross calorific value. The results showed that approximately 70% of this discard coal is composed of a material of sufficient quality for energy generation in conventional power stations. A pyrite-rich concentrate made up 2% of the total discard mass; comprising more than 45% of the sulfidic mineral present in the feed and displaying no acid neutralizing capacity (ANC). The remaining discard fraction, with intermediate density, presented potential to be used for several ends including soil fabrication, co-disposal or as aggregate material in civil engineering; additional testing to ensure applicability for the selected re-purposing option should be chosen based on proposed use.

Keywords: sink-float; ARD prevention; pyrite; coal discards; waste processing; waste minimization

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1. Introduction

Over 70% of South Africa's primary energy needs are still provided from coal [1]. To achieve market requirements, most of the run-of-mine (ROM) coal in South Africa (characterized in Table 1) is washed. As a result of beneficiation, approximately 30% to 40% of the total mass of coal mined in South Africa is, or has been, disposed as coal discards, typically in coal waste storage facilities, with no economic value recovered [2,3]. While these large amounts of material both increase the operational costs and negatively affect the environment in long term basis, they also contain significant amounts of usable coal as well as other minerals of interest such as pyrite and aluminosilicates [3–6], the recovery of which could enhance resource efficiency.

Currently, South African coal mining operations emphasize an end-of-pipe treatment approach to coal waste and acid rock drainage (ARD) management. Chemical techniques for ARD treatment, such as lime neutralization, typically consume large amounts of costly reagents, generate significant quantities of sludge, and are typically only effective in reducing ARD risks in the short term. The South African regulatory framework is comprehensive on environmental management of mining activities, taking a cradle to grave approach and putting the mining industry under increasing pressure to adopt more proactive approaches to mine waste disposal. Many of the long-term environmental liabilities can be overcome by implementing preventive techniques that both minimize the waste disposed of and minimize its potential for generation and subsequent dispersion of ARD from these waste storage facilities. Apart from reducing ARD risks, integration of a sulphide removal step into the coal beneficiation circuit also offers opportunity for additional value recovery [9–12]. For example, the separation and recovery of pyrite from the coal discards not only mitigates environmental risks, it also provides an integrated approach for managing pyrite in collieries to contribute to the sulfur-related industry in South Africa and has potential to provide a benign particulate phase to replace of quarried sands across a range of applications, thereby enhancing resource efficiency [13–15]. According to the South African Department of Mineral Resources [16], commercial deposits of elemental sulfur are unknown in South Africa; however, sulfur in the pyritic form is found in numerous deposits. Most of the known deposits have not been economically exploited for pyrite alone. Many deposits also contain other minerals, with potential for pyrite to be recovered as a by-product; examples include the auriferous conglomerates of the Witwatersrand and coal deposits in Mpumalanga and Waterberg. Amaral Filho et al. [17] illustrated that by processing coal waste discards from a Brazilian colliery with a total sulfur content of ~7% using gravity concentration techniques such as dense medium separation with iron-silicon suspensions, at least three density fractions could be obtained and used to several ends. The coal waste fractions with low relative densities can be used as raw material for power generation [3]. The mineral fractions with medium relative densities could be re-purposed as a particulate sand or aggregate and used in economic sectors such as construction [18,19]. Recent studies have also demonstrated the feasibility of using the low-sulfur mineral fraction of coal waste as parental material for the production of a Technosol to be used as topsoil in mine rehabilitation [20–22]. Finally, the fraction with relative density higher than 2.7 was rich in pyrite. Through thermal and biohydrometallurgical processes, these pyrite-rich fractions could be converted into valuable products including, but not limited to, ferrous sulphate, sulfuric acid, and iron oxides [13,23–25]. Further, by isolating the pyrite from the coal discards, acid generation potential of the discards can be reduced significantly, not only to obtain products with aggregated market value, but also reducing the related treatment costs and the land footprint concomitantly [17,26,27]. Further studies by Weiler and Schneider [11,17], conducted in two samples from different Brazilian coalfields, confirmed the feasibility of applying dense medium separation for valorization of coal waste with multiple benefits.

Table 1. South African ROM coal characteristics and market requirements for washed coal [7,8].

	ROM Coal	Thermal Coal
Sulfur (%)	0.5–15	0.7–1
Calorific Value (MJ/kg)	16–21	19–27
Ash (%)	20–40	20–30

Re-mining coal dump deposits to recover a thermal product is already a reality in South Africa, using old mineral processing facilities. Potential exists to combine this with further approaches for waste minimization, enhanced resource efficiency, and value recovery. Considering that proper and integrated management of downstream emissions is critical to minimize environmental legacy, understanding the technological and environmental characteristics of the material is fundamental to decision-making for responsible disposal of coal discards or, preferably, their re-purposing to move "towards zero mine waste" strategies. Sink-float tests offer a potential method to characterize coal wastes for both their environmental risks and their re-purposing potential and are already widely used for run-of-mine coal analysis, since they estimate the mineral accessibility when using gravity concentration. The data from such tests are used to analyze the content of ash, volatile matter, moisture and sulfur. Washability curves, such as the density curve, cumulative float-ash and sink-ash curves, cumulative float-sulfur and sink-sulfur curves, elementary curve and tolerance curve, also known as Near Gravity Material or simply NGM [28,29], can also be plotted. Additional analyses can be readily conducted on the different density fractions to determine other characteristics, such as acid generating potential and geochemical deportment.

The aim of this study was to use gravimetric separation to obtain coal discard fractions of varying density with potential for further application. Through their characterization and subsequent evaluation of techno-environmental aspects, future applications are suggested based on emerging and established technologies.

2. Materials and Methods

The bulk coal discards (feed) used in this study were collected from a dense medium separation unit from a colliery within the eMalahleni region in Mpumalanga. The mineralogy was determined by X-ray diffraction (XRD). Particle size distribution was determined by dry sieving. Further, potential for acid generation was explored by acid base accounting according to Miller and Stewart et al. [30,31].

Sink and float studies were performed in a single repetition to determine the coal amenability to gravity concentration and to prepare samples of varying density for further characterization, so as to inform potential for both waste minimization and waste repurposing. In order to determine the distribution of the discards as a function of the density, the samples were sieved to obtain particle diameters in between 2.0 and 50.8 mm, and subject to float and sink studies according to ASTM D4371-2012. Based on the results obtained by Amaral Filho et al. [5,17], different mixtures using several organic liquids, such as bromoform, perchloroethylene, and xylene at densities of 1.8; 2.0; 2.2 and 2.7 g/cm^3 were used in this study.

The five fractions obtained from sink and float studies were homogenized and prepared for techno-environmental characterization. Each fraction was analyzed in terms of proximate and ultimate analysis, sulfur forms and gross calorific value. The total sulfur content was determined according to ASTM D4239:1997. The sulfur speciation was conducted according to ASTM D4239. The ash analysis of the fractions obtained was carried out using ASTM D3174-2012. Proximate analysis was performed according to ASTM D5373-16. Semi-quantitative XRD analysis was conducted for assessment of mineralogical composition on each separated material. To determine the potential of each fraction to form acid rock drainage, static acid-base accounting (ABA) tests were carried out in all separated fractions according to Miller and Stewart et al. [30,31].

3. Results and Discussion

3.1. Characterization of the Head Sample

Figure 1 and Table 2 present the mineralogical composition and particle size distribution of the bulk sample respectively. The primary minerals present are quartz and kaolinite with the primary acid-forming mineral being pyrite. Some 40% of the discards lie in the size range 5.6 to 22.5 mm while 20% is larger than 31.5 mm and 14% smaller than 4.5 mm (Table 3).

The findings from the ARD static tests conducted on the bulk samples are presented in Table 4, showing that the samples are potentially acid forming, with very low acid neutralizing capacity. The presence of pyrite (Table 2) indicates that, after the consumption of the buffering capacity provided by the non-acid forming material, the samples generate ARD with its well-known risks and liabilities.

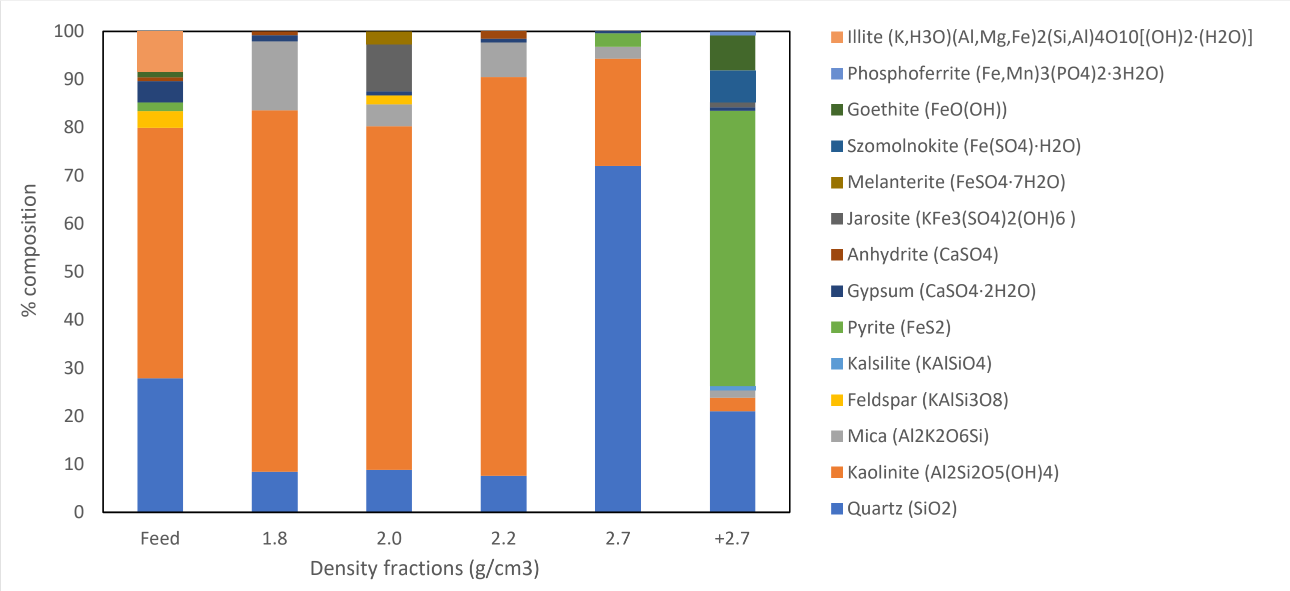

Figure 1. Semi-quantitative mineralogical compositions of the mineral matter in the density fractions obtained from washability studies of coal waste.

Table 2. Mineralogical semi-quantitative XRD phases obtained for the ash component of bulk coal discards samples (feed).

Mineral Phase	Chemical Formula	(%)
Quartz	SiO_2	27.9
Kaolinite	$Al_4Si_4O_{10}(OH)_2{\cdot}H_2O$	52.1
Gypsum	$CaSO_4{\cdot}2H_2O$	4.4
Illite	$(K,H_3O)(Al,Mg,Fe)_2(Si,Al)_4O_{10}[(OH)_2,(H_2O)]$	8.5
K-feldspar	$KAlSi_3O_8$	3.5
Pyrite	FeS_2	1.8
Anhydrite	$CaSO_4$	0.8
Magnetite	Fe_2O_4	1.1

Table 3. Particle size distribution of the bulk sample used in the sink-float studies.

Sieve Aperture (mm)	Retained Mass (%)	Cum. Mass (%)
31.5	20.8	20.8
22.5	4.5	25.3
16.0	10.2	35.5
11.2	15.8	51.3
8.0	6.2	57.5
5.6	21.5	79.0
4.5	7.0	86.0
2.0	14.0	100.0

Table 4. Acid base accounting test (ABA) results showing the total sulfur (S), maximum potential acidity (MPA), acid neutralizing capacity (ANC), and net acid producing potential (NAPP) for coal discards. PAF = potential acid forming.

Sample Name	Total S (%)	MPA	ANC kg H_2SO_4/t	NAPP	ARD Classification
Bulk sample (feed)	1.6	49.9	3.9	45.9	PAF

3.2. Valorization Opportunities

3.2.1. Float Sink Separations

The sink and float studies resulted in five fractions which were weighted and characterized for ultimate and proximate as well as gross calorific value analysis. The results are summarized in Table 5. Calculated feed presented 41.7% of ash, and 1.6% of total sulfur. Sulfur speciation is presented in Table 6. The experimental results of the ANC static tests are summarized in Table 7. In Figure 1, the results for the mineralogical studies are presented. The distinct characteristics of the density fractions demonstrate the potential for density separation to enable the re-purposing for the obtained fractions for discrete uses.

Table 5. Sink and float results in terms of mass recovery, calorific value, and ultimate and proximate analysis. Where: FC = fixed carbon; VM = volatile matter; C = carbon; H = hydrogen; N = nitrogen; S = sulfur; and CV = calorific value.

Density Fraction (g/cm)	Mass (%)	Proximate Analysis (%)				Ultimate Analysis (%)				CV (MJ/kg)
		Ash	FC	Moisture	VM	C	H	N	S	
1.8	58.5	19.6	53.2	2.7	24.5	60.5	3.3	1.6	0.5	24.6
2.0	3.5	49.4	25.3	1.8	23.5	42.0	2.7	1.2	0.4	13.1
2.2	7.9	59.1	18.4	2.2	20.3	35.0	2.4	1.0	2.8	9.4
2.7	28.1	80.4	1.4	1.6	16.6	12.7	1.6	0.5	1.5	1.2
>2.7	2.0	60.6	11.2	1.7	26.5	11.4	0.9	0.4	34.7	8.8

Table 6. Sulfur speciation of five fractions obtained from sink-float studies.

Density Fraction (g/cm^3)	Sulfur Forms (%)		
	Organic S	Sulfate S	Sulfide S
1.8	0.01	0.09	0.40
2.0	0.01	0.08	0.30
2.2	0.01	0.56	2.07
2.7	0.01	0.20	1.40
>2.7	0.01	0.70	33.30

Table 7. Acid base accounting test results showing the total sulfur (S), maximum potential acidity (MPA), acid neutralizing capacity (ANC), and net acid producing potential (NAPP) of five fractions obtained from densimetric separation.

Density Fraction (g/cm^3)	Total S (%)	MPA	ANC kg H_2SO_4/t	NAPP	ARD Classification
1.8	0.5	15.3	17.8	−3.0	NAF
2.0	0.4	12.2	0	12.2	PAF
2.2	2.8	85.7	13.1	98.8	PAF
2.7	1.5	45.9	4.4	50.3	PAF
>2.7	34.7	1061.8	0	1061.8	AF

3.2.2. Low Density Fractions

As expected, with decreasing density, the ash content decreases and caloric value increases (Table 5). The lowest density fraction (1.8 g/cm^3), which represents nearly 60% of the overall mass, presents a calorific value typical of saleable coal (24.6 MJ/kg) and an ash content less than 20%. This indicates that the current beneficiation method dislocates significant amount of material of interest to the rejects when compared with the ideal sink-float separation. The fractions with densities of 2.0 and 2.2 g/cm^3 report calorific values of 9.4 to 13.1 MJ/kg and ash content of 50–60% respectively. The total mass of these fractions combined resulted in 11.4% of the mass of the bulk (feed) sample. To reduce the volume of waste disposed and consequently improve resource efficiency, the three fractions with lower densities and higher carbon contents can be blended, and the combined fractions would present a theoretical density of <1.86 g/cm^3 and represent 70% of the bulk mass with an ash content of 25.5% and sulfur content of 0.7%. In terms of gross calorific value (CV), the three blended densities (1.8; 2.0 and 2.2 g/cm^3) presented an average value of 22.3 MJ/kg, with a combustible recovery higher than 95%. Mineralogical results (Figure 1), despite the sulfur speciation results found in Table 6, showed pyrite was not detected in the 1.8 g/cm^3, 2.0 g/cm^3 and 2.2 g/cm^3 fractions. The 2.0 g/cm^3 fraction showed the presence of iron bearing minerals such as jarosite and melanterite, which explains the null acid neutralizing capacity found in this density fraction due to its stored acidity [32]. The major mineral phase found in the three lowest density fractions was kaolinite, indicating that most of the acid neutralizing components were reported to the lower densities. It also contains approximately 95% of the carbonates and iron oxides, particularly kaolinite and jarosite. This is confirmed by the acid neutralizing capacity (Table 7) of the fraction with density 1.8 g/cm^3, whichwas considerably higher than its acid producing potential, hence NAPP was negative and this sample characterized as non-acid forming (NAF). The low-density fractions are rich in combustible coal as illustrated by a carbon content of roughly 57%, with high calorific value and low sulfur.

Based on the theoretical recovery of the sink and float studies, the combined fraction complies with market standards for thermal coal according to Table 1. The results support the ongoing re-mining activities already taking place in Mpumalanga area.

Beyond the evident presence of thermal coal available for recovery, the findings also showed further re-purposing opportunities where a second stage separation is done using a dense medium to separate the residual material into fractions above and below a density of 2.7 g/cm^3, as observed by previous research [5,11,17]. Ferrosilicon suspensions are widely used for high-density applications due to their capacity to achieve medium density ranges varying from 3.2 to 4.2 g/cm^3 [33].

3.2.3. Intermediary Density Fractions

According to results shown in Table 5, the material recovery using dense media of 2.2 and 2.7 g/cm^3 has an ash content of 80% and a total carbon content of 12.7% while comprising 28% of the mass of the bulk material. Approximately 26% of the total sulfur of the feed is still reported to this fraction. The reported high-ash content (>80%) in the density fraction lying between 2.2 and 2.7 g/cm^3 indicated the presence of substantial mineral matter. Its high quartz content (72% of the ash content) is accompanied by clay minerals indicating a more inert geochemical environment when compared with the bulk discards. Despite the increased amount of neutralizing and inert material reporting to the 2.2 to 2.7 g/cm^3 fraction, the presence of pyrite is still observed (Figure 1). To maximize maximize the environmental benefits, further washability studies might provide information to evaluate the potential for further sulfur recovery in the lower densities e.g., 2.4; 2.5 and 2.6 g/cm^3. More comprehensive washability studies would indicate the accessibility of this pyrite in lower densities. The XRD results confirm that the density fraction of 2.2 to 2.7 g/cm^3 is characterized predominantly by minerals of low reactivity, especially quartz. The authors of [9,26] demonstrate that the removal of the pyritic fraction in coal discards significantly reduces the acid generation potential and consumption of neutralizing reagent in ARD treatment facilities.

This fraction may be used for several ends including soil fabrication, co-disposal, or as an aggregate in the construction industry or for road building [18–20,27,34,35]. Understanding the deportment of the metals and acid from mine waste is crucial in enabling correct decisions on waste disposal and re-purposing studies. Further the balance of acid neutralizing capacity and acid generation of material for disposal, together with the physical approach to disposal, affects the final quality of the seepage from coal waste storage facilities, thereby delaying, preventing, or even removing the acid forming reactions with long term economic and environmental benefits [34,36].

Firpo et al. [20] developed a procedure to fabricate topsoils from coal mine discards comprised of the following steps: gravity processing, comminution and classification, amendments (pH adjustment and addition of active organic matter), soil analysis, and plant growth (macro- and micro-nutrients and metals concentration). Validation was conducted with a coal mine waste amended with steel slag and sewage sludge, on which growth of Sorghum bicolor was successfully achieved. In the same line, [6,21] investigated the feasibility of using the desulfurised fraction after two-stage flotation separation of fine coal waste as the main component for the manufacture of a 'FabSoil' for use as topsoil in the rehabilitation of mine sites. Compost and anaerobic digester sludge were added as organic matter and nutrient source, whilst malt residue was used as physical ameliorant. Native soil from the study area was used as control. Validation of the potential of the fabricated soils was conducted through germination and growth experiments using the grass species Teff (*Eragrotis tef*), an indigenous grass from the Mpumalanga region of South Africa commonly cultivated on degraded mine land.

In terms of civil construction applications, Santos et al. [19] studied the use of coal waste as fine aggregates to produce concrete blocks for paving. The results showed that using dense medium separation, it was possible to process the high-sulfur coal waste and obtain a desulfurised recycled fine aggregate that can be used in civil construction. Concrete blocks for paving produced with 25% and 50% recycled coal waste in substitution of river sand presented satisfactory results in terms of mechanical strength.

3.2.4. High Density Fraction

The highest density fraction (>2.7 g/cm^3) presents a sulfide-rich fraction with a total sulfur content of 34.7% and a pyritic sulfur content of 33.3%, representing a total and pyritic sulfur recovery of 42.4% and 45.3% respectively (Tables 5 and 6). As observed in Table 7 the isolated pyritic fraction was found to be highly acid forming as expected. According to the results from Figure 1, carbonate materials were not identified in the XRD pattern for this material. Hence, the neutralizing capacity was zero for this fraction (Table 7). The XRD results of the high-density fraction showed the dominance of pyrite at a concentration of 57.2% and a low concentration of slow-intermediate acid neutralizing minerals, such as kaolinite. The detected sulphate mineral in the high-density fraction was gypsum. Pyrite is a non-toxic and stable mineral when not exposed to water and air. However, after removal from the ground, mainly in gold, coal and polymetallic mining activities, pyrite is usually treated as gangue with non-market value, and typically placed in mine waste storage facilities generating acid rock drainage that are managed according to the related legal framework. Coal reserves are rich in pyrite and the impact of ARD in the coal fields are extensively studied and documented [4,13,37–39]. Several authors have studied the possibility of applying this waste as an industrial raw material. Pyrite roasting is an established process for sulfuric acid production [23], and the concentration of this mineral by coal mines was a reality until late 80 s in Brazil and considered during a similar time period in South Africa. The Catarinense Carbochemical Industry processed pyritic concentrates from local collieries to produce sulfuric acid for fertilizer processing. However, Brazilian regulations facilitating the importation of elemental sulfur made the process economically unfeasible and the pyrite concentration activities and uses ceased [40], leading to its disposal. Re-assessment of the viability of sulfuric acid generation from pyrite is recommended, taking into account both production costs of the acid and long-term liability costs of pyrite disposal.

In addition, this density fraction with high sulfur content and high acid forming potential has been studied so as to be used as source of iron and sulfur to produce ARD in a controlled environment using biohydrometallurgical techniques. This lab-induced ARD has been applied to manufacture a variety of emerging products of high value including iron oxides and salts such as magnetite and ferrous sulphate [24,41–45]. The reduction in concentration of impurities such as carbonates and other neutralizing minerals in the pyrite concentrate is important given that an alkaline environment inhibits the action of sulfur and iron oxidizing microorganisms [46] and also affects the quality of the pyrite concentrate and potentially the downstream products [13,47].

According to [48], iron oxides can be applied in several industrial sectors and the major applications are: pigments for paints and the construction industry; magnetic pigments and ferrites; and as raw material for the iron and steel industry. Recent studies [24,41] show that the feasibility of the production of materials from coal waste include nanostructured pigments and magnetic material. Application at pilot scale has been successfully tested.

The results presented and discussed above are based on sink-float studies using heavy liquids under controlled conditions. Under industrial conditions, however, the performance is never ideal. As a result, misplaced and undesired materials could be dislocated to the wrong fractions. However, studies conducted by Amaral Filho [49] demonstrated that using iron silicone suspensions as a separation medium, a fraction with similar grades and recovery can be obtained when compared with sink and float studies. Additionally, a small number of Brazilian collieries are already recovering the pyrite fraction using jigging, though a considerable amount of pyrite is still reported to the lower fractions. The presence of pyrite in lower densities and the presence of carbonates in fractions with densities higher than 2.7 g/cm^3 can affect its final application, and preliminary treatments may be required depending on the biogeochemical characteristics of the target waste material [10,13,50].

3.3. An Integrated Approach to Re-Purposing Coal Mine Waste

Re-mining of old mine waste storage facilities (MWSF) is usually driven by the development of new technologies to process and use lower grade minerals; however, the focus is still only on recovering the main minerals of interest. It does not consider any alternative solutions for the total mass of material, with the remaining materials being placed back into the MWSF. Moreover, by considering new processing and waste management routes in the early stage of the mine life cycle is both more cost effective and enables the long-term liabilities of a large amount of waste generated during mineral processing activities to be minimized. Integrated waste management has multiple benefits and is crucial for the establishment of a circular economy strategy in which resource efficiency is centre-stage. For example, the undesired gangue minerals from mine waste can be re-purposed as raw material to be incorporated in other sectors, e.g., the construction, agriculture, and chemical sectors. Using mineral processing techniques, such as flotation and dense medium separation, and with additional (bio)leaching approaches to recover the iron and sulfur from mine waste, better solutions for enabling a circular economy in the mining sector can be developed. Iron and sulfur form raw materials for several industrial processes, and a number of process options have been proposed and investigated for the downstream processing of coal wastes into useful products including rare earth elements [51]. To date these studies have been isolated, and no systematic analysis and assessment of options for integrated coal waste management has been undertaken which takes into account key site-specific variables, such as mineralogy, local markets, etc. Integrated mining waste management which uses desulfurization (pyrite isolation), metal recovery techniques, and re-purposing of the major and minor waste fractions has the potential to reduce economic costs, diversify production, reduce environmental burden, and increase resource efficiency for the mining companies.

Mining companies have been indicating their strong commitment to realizing the potential of green industries to open up new possibilities for development and, through this, to create much needed jobs. Providing R,D&I support, policy direction, and necessary legislation to address integrated waste management is part of the solution. The volume of mining waste, as well as its environmental and economic liabilities, are drivers in the re-evaluation of waste management. Mitigating the long-term effects of these waste deposits is not only legally necessary but also contributes to decreased environmental impact through avoiding pollution and enhancing resource recovery and resource efficiency. Developing sustainable technologies to address mine waste challenges by considering innovative and long-term solutions for the re-purposing of current "waste" materials from mining and mineral processing through an integrated framework is fundamental to the sustainable development framework for the mining sector. Consequently, long-term risk removal and value recovery need to be actively addressed to avoid further contamination of surface and groundwater bodies, to reduce the footprint of chemical reagent transportation, emissions and uses, and landfill disposal. Using this sustainable development approach, the principle of waste minimization is used in conjunction with industrial ecology for the re-purposing of waste materials from conventional treatment and disposal operations.

4. Conclusions

The presented approach demonstrates the potential for re-purposing coal discards through density separation to provide fractions of differing properties tailored to differing uses, with associated environmental and economic benefits. Firstly, the removal of the sulfidic component from the bulk coal discards using gravity concentration techniques prior to disposal presents significant reduction in the amount of pyrite in the mid-density coal discards, while also recovering a low-sulfur, low-ash clean coal of acceptable calorific value for use as a thermal coal. The separated low volume sulfur-rich concentrate can be further re-purposed, both enhancing resource productivity and preventing environmental risks associated with its disposal. Economic benefits can be achieved by converting the iron-sulfur components in this high-sulfur fraction into valuable products or into raw materials for utilization in other industrial sectors. The mid-density coal waste fraction can be further analyzed in terms of metal deportment and acid generation for suitability for use as a benign particulate or aggregate material in a number of sectors: agriculture, mine rehabilitation, and construction. Where not suitable, it can be returned to the waste disposal facility at a reduced footprint and with reduced potential for acid generation.

This study presents an opportunity for the handling of downstream South African coal discards through the removal of the pyritic component using gravity concentration enabling recovery of multiple products, thereby minimizing waste for final disposal and improving resource efficiency. The results support the feasibility of the two-stage concentration process for concomitant long-term removal of ARD risk, reduction of long-term liabilities associated with its management and treatment, and potential for value recovery in terms of combustible coal and additional products.

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References

1. Bahrami, A.; Ghorbani, Y.; Mirmohammadi, M.; Sheykhi, B.; Kazemi, F. The Beneficiation of Tailing of Coal Preparation Plant by Heavy-Medium Cyclone. *Int. J. Coal Sci. Technol.* **2018**, *5*, 374–384. [CrossRef]
2. Chugh, Y.P.; Behum, P.T. Coal Waste Management Practices in the USA: An Overview. *Int. J. Coal Sci. Technol.* **2014**, *1*, 163–176. [CrossRef]
3. North, B.; Engelbrecht, A.; Oboirien, B. Feasibility Study of Electricity Generation from Discard Coal. *J. S. Afr. Inst. Min. Metall.* **2015**, *115*, 573–580. [CrossRef]
4. Lloyd, P.J. The Potential of Coal Wastes in South Africa. *J. S. Afr. Inst. Min. Metall.* **2000**, *100*, 69–72.

5. Do Amaral Filho, J.R.; Schneider, I.A.H.; de Brum, I.A.S.; Sampaio, C.H.; Miltzarek, G.; Schneider, C. Caracterização de Um Depósito de Rejeitos Para o Gerenciamento Integrado Dos Resíduos de Mineração Na Região Carbonífera de Santa Catarina, Brasil. *REM Rev. Esc. Minas* **2013**, *66*, 347–353. [CrossRef]
6. Harrison, S.T.L.; Broadhurst, J.L.; Opitz, A.; Fundikwa, B.; Stander, H.-M.; Mostert, L.; Amaral Filho, J.; Kotsiopoulos, A. *An Industrial Ecology Approach to Sulphide Containing Mineral Wastes to Minimise ARD Formation PART 2: Design for Disposal and Extraction of Products of Value*; WRC in Pretoria: Pretoria, South Africa, 2020.
7. Eberhard, A. Market, Investment, and Policy Challenges for South African Coal. In *The Global Coal Market: Supplying the Major Fuel for Emerging Economies*; Thurber, M.C., Morse, R.K., Eds.; Cambridge University Press: Cambridge, UK, 2015; pp. 164–203.
8. Jeffrey, L.S. Characterization of the Coal Resources of South Africa. *J. S. Afr. Inst. Min. Metall.* **2005**, *105*, 95–102.
9. Kazadi Mbamba, C.; Harrison, S.T.L.; Franzidis, J.; Broadhurst, J.L. Mitigating Acid Rock Drainage Risks While Recovering Low-Sulfur Coal from Ultrafine Colliery Wastes Using Froth Flotation. *Miner. Eng.* **2012**, *29*, 13–21. [CrossRef]
10. Oliveira, C.M.; Machado, C.M.; Duarte, G.W.; Peterson, M. Beneficiation of Pyrite from Coal Mining. *J. Clean Prod.* **2016**, *139*, 821–827. [CrossRef]
11. Weiler, J.; Schneider, I.A.H. Pyrite Utilization in the Carboniferous Region of Santa Catarina, Brazil-Potentials, Challenges, and Environmental Advantages. *REM Int. Eng. J.* **2019**, *72*, 515–522. [CrossRef]
12. Taha, Y.; Elghali, A.; Derhy, M.; Amrani, M.; Hakkou, R.; Benzaazoua, M. Towards an Integrated Approach for Zero Coal Mine Waste Storage: Solutions Based on Materials Circularity and Sustainable Resource Governance. *Miner. Processing Extr. Metall. Rev.* **2022**, 1–14. [CrossRef]
13. Patricio Ferreira, L.; Müller, T.G.; Cargnin, M.; de Oliveira, C.M.; Peterson, M. Valorization of Waste from Coal Mining Pyrite Beneficiation. *J. Environ. Chem. Eng.* **2021**, *9*, 105759. [CrossRef]
14. Amar, H.; Benzaazoua, M.; Edahbi, M.; Villeneuve, M. Reprocessing Feasibility of Polymetallic Waste Rock for Cleaner and Sustainable Mining. *J. Geochem. Explor.* **2020**, *220*, 106683. [CrossRef]
15. Amar, H.; Benzaazoua, M.; Elghali, A.; Bussière, B.; Duclos, M. Upstream Environmental Desulphurisation and Valorisation of Waste Rocks as a Sustainable AMD Management Approach. *J. Geochem. Explor.* **2020**, *215*, 106555. [CrossRef]
16. Department of Mineral Resources. *Review of the Sulphur Industry in the Republic of South Africa*; Report R101/2013; Arcadia: Pretoria, South Africa, 2012.
17. Do Amaral Filho, J.R.; Weiler, J.; Broadhurst, J.L.; Schneider, I.A.H. The Use of Static and Humidity Cell Tests to Assess the Effectiveness of Coal Waste Desulfurization on Acid Rock Drainage Risk. *Mine Water Environ.* **2017**, *36*, 429–435. [CrossRef]
18. Argane, R.; Benzaazoua, M.; Bouamrane, A.; Hakkou, R. Cement Hydration and Durability of Low Sulfide Tailings-Based Renders: A Case Study in Moroccan Constructions. *Miner. Eng.* **2015**, *76*, 97–108. [CrossRef]
19. Dos Santos, C.R.; do Filho, J.R.A.; Tubino, R.M.C.; Schneider, I.A.H. Use of Coal Waste as Fine Aggregates in Concrete Paving Blocks. *Geomaterials* **2013**, *3*, 54–59. [CrossRef]
20. Firpo, B.A.; Amaral Filho, J.R.D.; Schneider, I.A.H. A Brief Procedure to Fabricate Soils from Coal Mine Wastes Based on Mineral Processing, Agricultural, and Environmental Concepts. *Miner. Eng.* **2015**, *76*, 81–86. [CrossRef]
21. Amaral Filho, J.R.; Firpo, B.A.; Broadhurst, J.L.; Harrison, S.T.L. On the Feasibility of South African Coal Waste for Production of 'FabSoil', a Technosol. *Miner. Eng.* **2020**, *146*, 106059. [CrossRef]
22. Weiler, J.; Firpo, B.A.; Schneider, I.A.H. Technosol as an Integrated Management Tool for Turning Urban and Coal Mining Waste into a Resource. *Miner. Eng.* **2020**, *147*, 106179. [CrossRef]
23. Runkel, M.; Sturm, P. Pyrite Roasting, an Alternative to Sulphur Burning. *J. S. Afr. Inst. Min. Metall.* **2009**, *109*, 491–496.
24. Lopes, F.A. *Producao Hidrometalurgica de Oxidos Magneticos a Partir de Concentrado de Pirita Proveniente de Rejeitos Da Mineracao de Carvao*; UFRGS: Porto Alegre, Brazil, 2017.
25. Vigânico, E.M. Protótipo Em Escala Piloto Para Produção de Sulfato Ferroso a Partir de Concentrado de Pirita Da Mineração de Carvão. Ph.D. Thesis, Federal University of Rio Grande Sul, Porto Alegre, Brazil, 2014.
26. Weiler, J.; do Amaral Filho, J.R.; Schneider, I.A.H. Processamento de Rejeito de Carvão Visando a Redução de Custos No Tratamento Da Drenagem Ácida de Minas—Estudo de Caso Na Região Carbonífera de Santa Catarina. *Eng. Sanit. Ambient.* **2016**, *21*, 337–345. [CrossRef]
27. Harrison, S.T.L.H.; Opitz, A.K.B.; Jera, M.K.; Mostert, L.D.; Kotsiopoulos, A. *An Industrial Ecology Approach to Sulphide Containing Mineral Wastes to Minimise ARD Formation: Characterising Potential for ARD, Design for Disposal and Extraction of Products with Value*; Report in Preparation; Arcadia: Pretoria, South Africa, 2013.
28. Cebeci, Y.; Ulusoy, U.; Sönmez, I. Determination of Optimum Washing Conditions for a Lignite Coal Based on Ash and Sulfur Content. *Fuel* **2014**, *123*, 52–58. [CrossRef]
29. De Souza, K.F.; Sampaio, C.H.; Kussler, J.A.T. Washability Curves for the Lower Coal Seams in Candiota Mine—Brazil. *Fuel Process. Technol.* **2012**, *96*, 140–149. [CrossRef]
30. Stewart, W.A.; Miller, S.D.; Smart, R. Advances in Acid Rock Drainage (ARD) Characterisation of Mine Wastes. In Proceedings of the 7th International Conference on Acid Rock Drainage 2006, ICARD—Also Serves as the 23rd Annual Meetings of the American Society of Mining and Reclamation, St. Louis, MO, USA, 26–30 March 2006; Volume 3, pp. 2098–2119.
31. Miller, S. *ACARP Project C15034: Development of ARD Assessment for Coal Process Wastes*; Environmental Geochemistry International Pty Ltd.: Balmain, Australia, 2008.

32. Schumann, R.; Stewart, W.; Miller, S.; Kawashima, N.; Li, J.; Smart, R. Acid-Base Accounting Assessment of Mine Wastes Using the Chromium Reducible Sulfur Method. *Sci. Total Environ.* **2012**, *424*, 289–296. [CrossRef]
33. Grobler, J.D.; Sandenbergh, R.F.; Pistorius, P.C. The Stability of Ferrosilicon Dense Medium Suspensions. *J. S. Afr. Inst. Min. Metall.* **2002**, *102*, 83–86.
34. Kotsiopoulos, A.; Harrison, S.T.L. Co-Disposal of Benign Desulfurised Tailings with Sulfidic Waste Rock to Mitigate ARD Generation: Influence of Flow and Contact Surface. *Miner. Eng.* **2018**, *116*, 62–71. [CrossRef]
35. Taha, Y.; Benzaazoua, M.; Hakkou, R.; Mansori, M. Coal Mine Wastes Recycling for Coal Recovery and Eco-Friendly Bricks Production. *Miner. Eng.* **2017**, *107*, 123–138. [CrossRef]
36. Mjonono, D.; Harrison, S.T.L.; Kotsiopoulos, A. Supplementing Structural Integrity of Waste Rock Piles through Improved Packing Protocols to Aid Acid Rock Drainage Prevention Strategies. *Miner. Eng.* **2019**, *135*, 13–20. [CrossRef]
37. Weiler, J.; Amaral Filho, J.R.; Schneider, I.A. Processamento de Rejeitos de Carvão e Redução Do Impacto Ambiental. *Augmdomus* **2014**, *6*, 80–94.
38. Falcon, R.; Ham, A.J. Characteristics of Southern African Coals. *J. S. Afr. Inst. Min. Metall.* **1988**, *88*, 145–161.
39. McCarthy, T.S. The Impact of Acid Mine Drainage in South Africa. *S. Afr. J. Sci.* **2011**, *107*, 1–7. [CrossRef]
40. Oliveira, M.L.S.; Ward, C.R.; Izquierdo, M.; Sampaio, C.H.; De Brum, I.A.S.; Kautzmann, R.M.; Sabedot, S.; Querol, X.; Silva, L.F.O. Chemical Composition and Minerals in Pyrite Ash of an Abandoned Sulphuric Acid Production Plant. *Sci. Total Environ.* **2012**, *430*, 34–47. [CrossRef] [PubMed]
41. Akinwekomi, V.; Maree, J.P.; Masindi, V.; Zvinowanda, C.; Osman, M.S.; Foteinis, S.; Mpenyana-Monyatsi, L.; Chatzisymeon, E. Beneficiation of Acid Mine Drainage (AMD): A Viable Option for the Synthesis of Goethite, Hematite, Magnetite, and Gypsum–Gearing towards a Circular Economy Concept. *Miner. Eng.* **2020**, *148*, 106204. [CrossRef]
42. Villetti, P.I.C. Produção de Coagulante Férrico a Partir Da Lixiviação de Concen—Trado de Pirita Da Mineração de Carvão via Cristalização/Solubilização de Sulfato Ferroso: Estudo Comparativo Entre Rejeitos de Duas Jazidas. Master's Thesis, Federal University of Rio Grande do Sul—UFRGS, Porto Alegre, Brazil, 2017.
43. Vigânico, E.M.; Colling, A.V.; Silva, R.D.A.; Schneider, I.A.H. Biohydrometallurgical/UV Production of Ferrous Sulphate Heptahydrate Crystals from Pyrite Present in Coal Tailings. *Miner. Eng.* **2011**, *24*, 1146–1148. [CrossRef]
44. Menezes, J.C.C.C.; Colling, A.V.; Silva, R.A.S.; dos Santos, R.H.; Scheneider, I.A.H. Ferric Sulphate Coagulant Obtained by Leaching from Coal Tailings. *Mine Water Environ.* **2017**, *36*, 457–460. [CrossRef]
45. De Silva, R.A.; Secco, M.P.; Lermen, R.T.; Schneider, I.A.H.; Hidalgo, G.E.N.; Sampaio, C.H. Optimizing the Selective Precipitation of Iron to Produce Yellow Pigment from Acid Mine Drainage. *Miner. Eng.* **2019**, *135*, 111–117. [CrossRef]
46. Tambwe, O.; Kotsiopoulos, A.; Harrison, S.T.L. Desulphurising High Sulphur Coal Discards Using an Accelerated Heap Leach Approach. *Hydrometallurgy* **2020**, *197*, 105472. [CrossRef]
47. Menezes, J.C.C.C.; Colling, A.V.; Silva, R.A.S.; Scheneider, I.A.H. Effect of Pyrite Concentration on the Quality of Ferric Sulfate Coagulants Obtained by Leaching from Coal Tailings. *Miner. Metall. Process.* **2016**, *33*, 77–81. [CrossRef]
48. Laurent, S.; Forge, D.; Port, M.; Roch, A.; Robic, C.; Vander Elst, L.; Muller, R.N. Magnetic Iron Oxide Nanoparticles: Synthesis, Stabilization, Vectorization, Physicochemical Characterizations, and Biological Applications. *Chem. Rev.* **2008**, *108*, 2064–2110. [CrossRef] [PubMed]
49. Amaral Filho, J. Aproveitamento de Rejeitos de Carvão em Santa Catarina. Ph.D. Thesis, UFRGS, Porto Alegre, Brazil, 2014.
50. Machado De Oliveira, C.; Gesser Müller, T.; Patricio Ferreira, L.; Prado Cechinel, M.A.; Peterson, M.; Raupp-Pereira, F. Valorization of Iron Pyrite from Coal Mining in Southern Brazil. *J. Environ. Chem. Eng.* **2019**, *7*, 102931. [CrossRef]
51. Akdogan, G.; Bradshaw, S.; Dorfling, C.; Bergmann, C.; Ghosh, T.; Campbell, Q. Characterization of Rare Earth Elements by XRT Sorting Products of a South African Coal Seam. *Int. J. Coal Prep. Util.* **2022**, *42*, 1071–1087. [CrossRef]