Using static, kinetic and metal mobility procedures to evaluate possibilities of coal waste land disposal at Moatize Mine, Mozambique

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Abstract

Coal mine waste rocks produced during mining and beneficiation can affect the environment due to land degradation and acid mine drainage generation. However, proper characterization and treatment of mineral processing wastes creates options for safe disposal, enhancing the environmental performance. The aim of this study was to use static and kinetic tests to assess properties of coal waste produced at the Moatize Mine in Mozambique, one of the largest "world-class" coal deposits. The study included sampling and characterization of coarse and fine wastes generated during coal beneficiation. Both materials were analyzed in terms of granular properties, static procedures - immediate composition, sulfur forms, mineralogical composition, elemental composition, acid-base balance – and kinetic behavior in humidity cell tests. The conjugation of these techniques indicates that these wastes have a low water acidification potential and low geochemical mobility of toxic elements. Therefore, it was concluded that fine and coarse wastes could be used in land shaping procedures.

Keywords: coal; acid rock drainage; geochemical mobility; mine waste management.

1. Introduction

The extraction of coal is an activity practiced around the world, as it is used to generate thermal energy and metallurgical coke. However, in most coal mines, the extracted material does not present adequate specifications for use, and must undergo a beneficiation process. Coal preparation plants generate wastes with varying quantities of iron sulfides (such as pyrite) and other chemical elements that may cause damage to the environment.

The pyrite (FeS_2) oxidation is the main cause of Acid Mine Drainage (AMD) generation. Sulfide oxidation occurs whenever water in the presence of oxygen percolates through pyrite bearing rocks, acquiring high acidity and high concentrations of Fe (as Fe^{2+} and/or Fe^{3+}) and sulfates (Kontopoulos, 1998; Simate and Ndlovu, 2014). Due to its low pH, the AMD can solubilize and mobilize other metals present in the waste, compromising the quality of surface and ground water (Akcil & Koldas, 2006; Grande *et al.*, 2018) and soils that it might contact (Chen *et al.*, 2015; Yang *et al.*, 2016).

In this context, coal waste characterization can help assessing the potential of acid generation, predicting possible risks and works as a tool in relation to its management (Komnitsas *et al.*, 2001; Lengke *et al.* 2010; Amaral Filho *et al.*, 2017). Static and kinetic tests have been developed and applied in order to predict the behavior of wastes, especially in terms of acid mine generation (Sapsford *et al.* 2009;

Bouzahzah *et al.*, 2015; Choparda *et al.*, 2017; Pondja *et al.*, 2017). Furthermore, the mining sector should improve its environmental performance by looking for and implementing alternatives related to the management and disposal of mineral processing wastes.

The Moatize Coal Basin is located in Mozambique's Tete Province (16°10'20''S 33°47'00''E), southern Africa. In this region, the coal is associated to the Karoo Supergroup (Johnson *et al.*, 1996). The Moatize formation reaches a thickness of 340 m with six coal seams known - from bottom to top: Sousa Pinto, Chipanga, Bananeiras, Intermédia, Grande Falésia and André – of which Chipanga is the thickest and the main one explored (Vasconcelos, 2000).

The Moatize Coal Mine Complex is one of the largest "world-class" coal deposits, explores the Chipanga seam and is estimated to operate until 2049. The beneficiation plant generates coarse and fine wastes: coarse waste results from processes like dense medium cyclone and jig operation; and fine wastes are derived from a parallel circuit, such as spiral and flotation systems (Leonard, 1991, Riazi and Gupta, 2015). The wastes generated have differences in terms of properties and generally neither one has a future use other than the surface disposal predicted, such as backfill, dumps and/or tailings dams.

Regarding the current practice of coarse and fine waste management and disposal at the mine, coarse wastes are

continuously removed by large trucks to backfill the mineral exploration pit. This is a safe and satisfactory practice and it additionally assists in land mine reshaping. Fine wastes are disposed in tailings dams, without any future use predicted. Tailings dams require large areas and have associated risks of potential failure. Fine material can also be transported by wind or water to nearby areas, compromising soils and water bodies. Additionally, the wastes can become self-compacted over time making, it difficult to vegetate, and can present risks of acid generation (Franks *et al.*, 2011; Kossoff *et al.*, 2014).

In this context, the characterization of the materials in terms of AMD generation, along with the relationship with the mining procedures and weather conditions, are important in terms of environment safety in the short and long term. The aim of this study was to assess properties of coarse and fine coal wastes while addressing possible environmental risks of the surface disposal. This study included the sampling, quantification and characterization of the coarse and fine coal waste in terms of static procedures – immediate composition, sulfur forms, mineralogical composition, elemental composition, acid-base balance – and kinetic behavior in humidity cell tests. After which, the mobility of metals in these materials and their appropriate disposal were evaluated. Finally, possibilities for appropriate management are presented and discussed.

2. Materials and methods

The Moatize Coal Mine Complex has the capacity to produce between 18 and 22 million tons per year, 65% coking coal and 35% thermal coal. In order to do so, the beneficiation of coarse ROM particles is performed by dense medium cyclones (1 mm < d < 50 mm) while the fine ROM particles, by spirals or elutriation (fraction 0.25 < d < 1 mm) and by flotation (fractions d < 0.25 mm). During a sampling campaign, the total waste production in relation to the coal "run of mine" (ROM) was about 63% (or 34 million tons per year) divided into coarse waste (90%) and fine waste (10%). Coarse and fine waste samples were collected

and studied separately due to the different generation and disposal process.

Granular properties of samples were evaluated by particle size distribution, particle density, bulk density, and porosity. Also, the incorporation of the fine material inside the empty spaces of coarse particles was performed to reduce the volume of disposal and to assess the reduction of porosity of the mixed materials. Particle size distribution was performed using a Tyler standard screen series. Particle density was determined gravimetrically by pycnometry, bulk density (or apparent density) and porosity, which were carried out following ASTM D167

(ASTM, 2012a).

Waste samples were ground, when necessary, via sequential comminution (jaw crushers, roller crushers and a planetary ball mill) to reach the particle size specification of each analysis. Characterization of the materials was performed by immediate analysis (ash, volatile matter and fixed carbon), sulfur forms, mineralogical composition, elemental analysis (environmentally available metals) and acid-base accounting to predict water acidification via static and kinetic methods. For most of the analyses, the material was milled to particle size under 0.25 mm (60# Tyler), with the

exception of kinetic tests that the specification required be under 6.3 mm. In this case, coarse waste was ground to below 6.3 mm.

The ash, volatile matter and fixed carbon content (elemental analysis) were determined using the ASTM D3174 (ASTM, 2012b) and ASTM D3175 (ASTM, 2011a) methods. The total sulfur content was determined using a LECO SC 457. The sulfate and pyritic sulfur forms were determined using ISO 157 standard procedures (ISO, 1996) and organic sulfur was calculated by subtracting total sulfur from other forms. The results of the immediate analysis and sulfur analysis were converted to a dry basis by the moisture analysis that was performed using ASTM D 3173 (ASTM, 2011b).

Mineralogical analysis was carried out by X-ray diffraction (XRD) in a Siemens (Bruker AXS, United States) X-ray diffractometer, model D-5000 (θ–θ), equipped with a curved graphite monochromator in the secondary beam and a fixed Cu anode tube, operating 40 kV and 40 mA, with an incident radiation of 1.5406 Å. The angular range analyzed was from 5° to 75°, 2θ with a step size of 0.05° s^{-1} using divergence and anti-scattering slits of 1° and 0.2 mm in the detector. This method enables the qualitative identification of the minerals present in the samples. If sulfide minerals are present, there is potential for acid generation; if caustic minerals are present, then there is potential for acid neutralization.

The accurate balance between the potential for acid generation and its neutralization was determined using an acid-base accounting method and by kinetic tests in humidity cells. The Modified Acid-Base Accounting (ABA) Method (Sobek *et al.*, 1978) was used for the static tests. The test calculates the sample's acid potential (AP) generation and also measures its neutralization potential (NP). The acid potential (AP) was determined from the pyritic sulfur content (S_{pyr}); stoichiometrically, one mole of $CaCO_3$ is required for the neutralization of a mole of S_{pyr} (Eq. 1).

$$AP \ (kg \ CaCO_3 \ t^{-1}) = (1000/32) \times \%S_{pyr} \qquad [Eq. \ 01]$$

The neutralization potential (NP) was determined by acid digestion at room temperature for 24 hours to consume the neutralizing minerals, followed by titration with NaOH until pH 8.3 is reached (Eq. 2).

$$NP \ (kg \ CaCO_3 \ t^{-1}) = (HClcons, \ g \ g^{-1} \ of \ sample) \times (50/36.5) \times 1000 \qquad [Eq. \ 02]$$

These data are used to calculate the net neutralization potential (NNP) (Eq. 03):

$$NNP \ (kg \ CaCO_3 \ t^{-1}) = NP - AP \qquad [Eq. \ 03]$$

The analysis criteria for the potential of acid generation of the materials were as follows:

NNP < -20 – indicates acid generation,

NNP > +20 – indicates no acid generation,

-20 < NNP < +20 – there is no clear indication whether acid generation will or will not occur.

The kinetic test was performed in humidity cells according to the D 5744 guideline (ASTM, 1996). The test consisted of adding 1000 grams of a given sample with particles smaller than 6.3 mm into a closed cylindrical vessel where cycles of dry air, moist air and water were applied in sequence for a given time. Initially, the sample was leached with 500 mL of distilled water, and the leachate was collected for analysis. Dry air was then injected into the samples for three days, followed by three days with moist air, and on the seventh day, the sample was leached with 500 mL of distilled water which was collected for chemical analysis. This cycle was repeated for 20 weeks. The test was performed on two cells: one containing coarse samples and the other with fine waste samples. The parameters analyzed in the leachate were pH, Eh, acidity, alkalinity, sulfate concentration and the concentrations of the following chemical elements: Al, As, Cd, Pb, Co, Cu, Cr, Fe, Mn, Mo, Ni, Va, Zn, Ba, Na, Ca and Mg. Analyses of sulfur speciation and neutralization potential were carried out with the samples before and after leaching.

The same chemical elements analyzed in leachate in the humidity-cell tests were determined for both coarse and fine waste solid samples following US EPA 3050 (US EPA, 1996). The concentrations of these elements in solid samples were compared to the accumulated results from the humidity-cell tests and the leached percentage was determined to assess the geochemical mobility of the elements.

All results attained at granular studies, immediate analysis, sulfur forms, mineralogical composition, elemental analysis and acid-base accounting were expressed in terms of the mean and the standard deviation of four independent samples.

3. Results and discussion

Table 1 presents some granular properties of coarse and fine waste from the Moatize Mine, including the incorporation of fine waste in the empty spaces among the coarse particles, which was found to reach up to the mass ratio coarse:fine of 1:0.29. The *top size* of coarse and fine particles are 50 mm and 1 mm,

respectively. Mean density was determined as 2.21 g cm⁻³ for coarse waste and 1.79 g cm⁻³ for fine waste. Considering the apparent density, 1.18 g cm⁻³ with an empty space among particles (porosity) of 39% was found for the coarse material. For the fine particles, the mean values found were 1.09 g cm⁻³ and 46%. The incorporation of the fine material in empty spaces of coarse particles could reduce the porosity to 30%, which could be a good strategy to reduce the volume for waste discharge and minimize water and oxygen transfer. These findings are in agreement with the work of Shokouhi and Williams (2017), which emphasis that co-disposal of fine-grained wastes and coarse-grained wastes generated in the processing of the run-of-mine coal, offers potential economic, geotechnical and environmental advantages over conventional separate disposal practices.

Table 1 - Particle size distribution, density, apparent density, and porosity to fine, coarse, and mixed to saturation of fines inside the coarse particles of Moatize Mine coal waste (mean of n = 4).

	Fine Waste		Coarse Waste		Coarse/Fine Waste 1:0.29 Ratio	
Particles size (mm)	1.0 – 0.1		50.0 – 0.5		50.0 – 0.1	
	Mean	SD	Mean	SD		SD
Density (g cm⁻³)	1.79	0.03	2.21	0.11	-	-
Apparent density (g cm⁻³)	1.09	0.04	1.18	0.07	1.52	0.08
ε (%)	46.5	1.29	39.5	1.91	29.75	4.99

SD – standard deviation.

Table 2 presents the results of immediate analysis (ash, volatile matter, and fixed carbon), total sulfur and its speciation (pyritic, sulfate and organic), major and minor crystalline components identified and acid-base accounting balance for the fine and coarse coal wastes. The results of the immediate analysis did not vary greatly between the granulometries of coarse and fine wastes. It is possible to perceive a difference between coarse and fine granulometries in relation to total sulfur and its speciation (pyritic, sulfate and organic). The fine waste showed higher amounts of total sulfur (1.3%) than the coarse waste (0.9%). Differences are also seen in terms of sulfur speciation; pyritic sulfur corresponds to 0.6% in the fine waste and 0.4% in the coarse ones for example.

Table 2 - Immediate analysis (ash, volatile matter and fixed carbon), total sulfur contents and its speciation (pyritic, sulfate and organic), major and minor crystalline components and acid-base accounting balance for the fine and coarse coal wastes produced from coal beneficiation (n=4).

	Fine waste		Coarse waste	
	Mean	SD	Mean	SD
Immediate analysis				
Ash (%)	58.4	5.2	58.9	5.4
Volatile matter (%)	16.6	2.2	16.5	0.9
Fixed carbon (%)	25.0	3.1	24.6	4.5
Sulfur speciation				
S_{total} (%)	1.33	0.13	0.89	0.18
$S_{pyritic}$ (%)	0.61	0.14	0.45	0.13
$S_{sulfate}$ (%)	0.18	0.05	0.09	0.10
$S_{organic}$ (%)	0.55	0.13	0.35	0.21
Crystalline componentes				
Major	Quartz		Quartz	
Minor	Calcite, hematite		Calcite, hematite	
Acid-base accounting				
NP (kg CaCO₃ t⁻¹)	25.3	5.0	23.5	6.7
AP (kg CaCO₃ t⁻¹)	19.0	4.2	14.0	3.5
NNP (kg CaCO₃ t⁻¹)	+ 6.3	4.5	+ 9.5	6.7

SD – standard deviation.

The crystalline mineral analysis showed that quartz was the major component in the samples. Calcite, the crystalline form of calcium carbonate ($CaCO_3$), is present as a minor component in all samples. The presence of pyrite was not identified, possibly due to its low concentration in the materials. In terms of acid-base accounting results, the acid potential was shown somewhat lower than the neutralization potential

for the analyzed samples, resulting in a positive net neutralization potential, although it is in the range of the "no clear indication" of acid generation (-20 < NNP < +20 kg $CaCO_3$ t^{-1}). This balance can be explained by the low concentration of pyritic sulfur and the presence of alkaline components, such as calcite, which was identified as a minor mineral through XRD.

Since static tests indicated difficulty

in predicting acidification; a kinetic test was performed to examine changes in the leachate with respect to pH, acidity and alkalinity generation, leaching of pyrite oxidation products (Fe and sulfates) and metallic elements present in the medium. Figure 1 shows the results of weekly pH, Eh, acidity, alkalinity and sulfate analyses and Figure 2 shows the results of the weekly analyses of metals in the leachate of the humidity-cells.
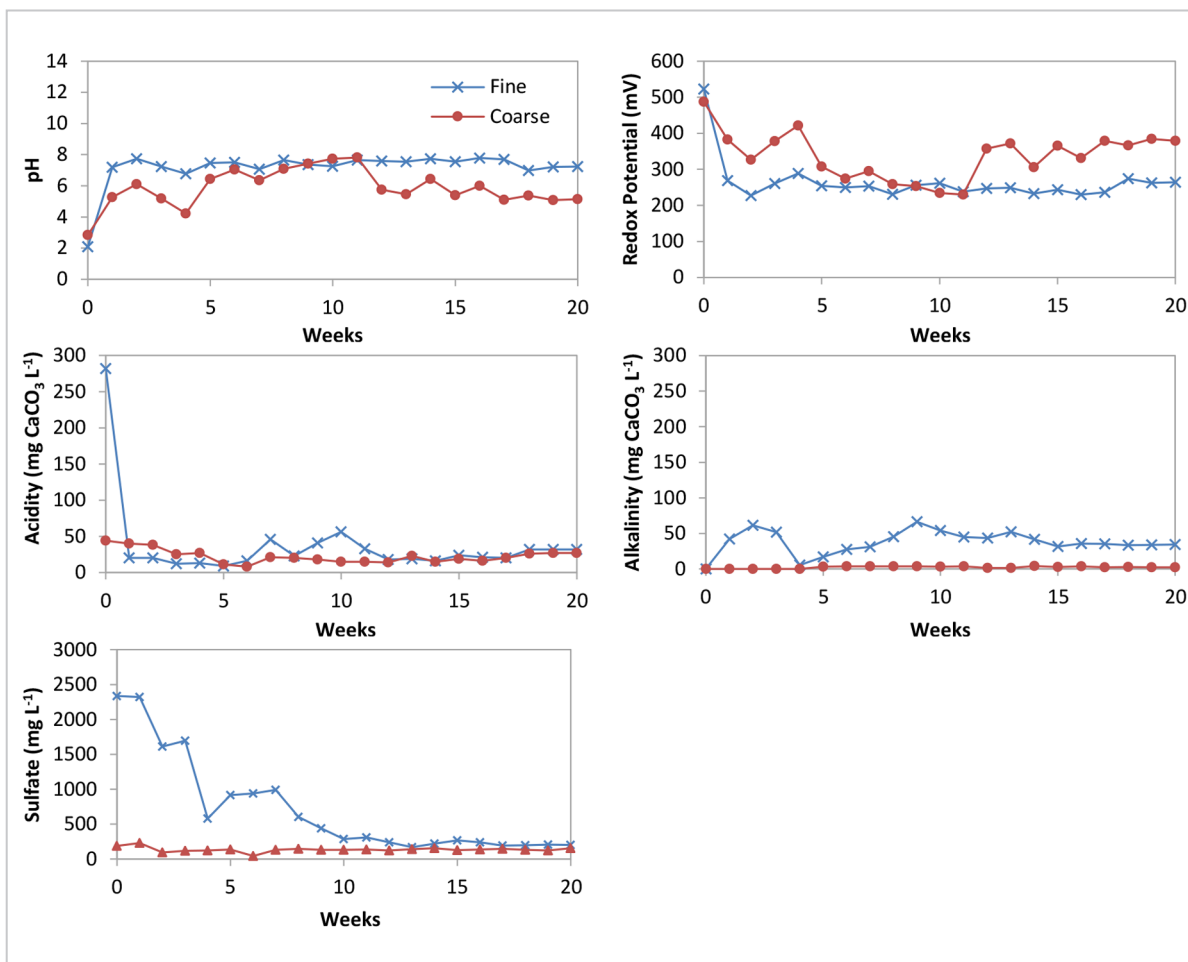

Figure 1 - pH, Eh, acidity, alkalinity, and sulfates of the leachate throughout the humidity cell tests for the coarse and fine wastes.

The pH of the fine waste leachate remained close to neutral (between 6 and 8) during most of the 20-week period. The coarse waste pH behaved differently remaining somewhat lower, with values ranging between 4 and 8. The redox potential varied between +200 mV and +500 mV for all leachates, indicating that oxidizing conditions are dominant in the humidity cells. Higher acidity was found only during the first week for the fine waste. The higher acidity is probably due to the higher amount of pyritic sulfur in this sample and, mostly, a higher degree of oxidation of this mineral.

Anyhow, the alkalinity values overcome the acidity, especially in fine wastes, which showed a higher alkalinity release. Finally, the sulfate values in the leachate of the fine wastes were near 2,300 mg L^{-1} in the first leaching and decreased over the weeks, stabilizing near 200 mg L^{-1}. In the coarse wastes, the sulfate values remained in the 200 mg L^{-1} range throughout the experiment. In summary, the oxidizing environment (high redox potential) allowed the oxidation reactions of pyrite and its transformation into sulfate; the acidity generated by the oxidation was compensated by alkalinity (dissolution

of the alkalinizing minerals present in the wastes), keeping the pH close to neutrality.

For metal analyses, the higher concentrations of leached elements were Ca and Mg, indicating the presence of carbonates, as, for example, $CaCO_3$ and $MgCO_3$. Most metals showed higher values in the first leaching flush, even though the indices remained lower for the rest of the experiment. There was no indication of toxic metal concentrations among the elements analyzed, and the metals Cd, Cr, Mo, Se, Va were not found in the leachate.
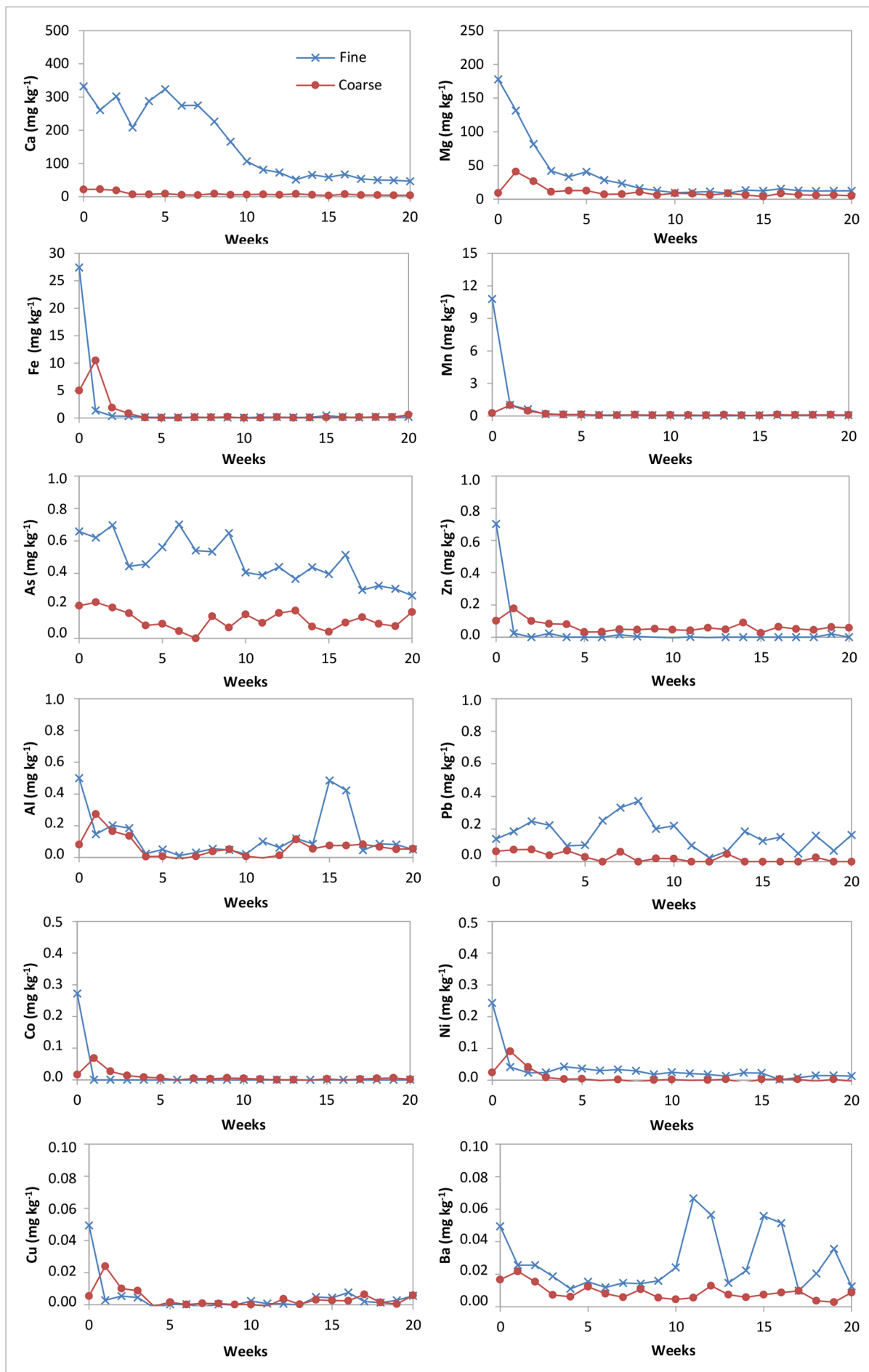
Figure 2 - Metal concentrations (Ca, Mg, Fe, Mn, Al, Zn, As, Pb, Co, Ni, Cu and Ba) in leachate throughout the humidity cell tests for the coarse and fine wastes.

Table 3 shows the results for sulfur speciation and acid-base accounting for coarse and fine wastes before and after the humidity cell tests. Both wastes showed a reduction in total sulfur content after 20 weeks of testing, partly due to a decrease in the pyritic sulfur content, which decreased by 50% in the fine and 20% in the coarse wastes. These results are in accordance with the work of Erguler and Kalyoncu (2015), which demonstrated a higher kinetic of AMD generation for fine particles rather than coarse particles in leaching columns, as a consequence of a higher surface area of exposure. As a consequence, the acid potential (AP) of the material decreased. The neutralization potential (NP) after the test was very similar to the value at its beginning, resulting in a higher net neutralization potential (NNP) value, but still in the range correspondent to "no clear indication" of acid generation.

Table 3 - Total sulfur contents and its speciation (pyritic, sulfate and organic) and acid-base accounting before and after the humidity cell test for fine and coarse wastes.

	Fine waste		Coarse waste	
	Before	After	Before	After
Sulfur speciation				
S_{total} (%)	1.33	0.94	0.89	0.77
$S_{pyritic}$ (%)	0.61	0.32	0.45	0.36
$S_{sulfate}$ (%)	0.18	0.14	0.09	0.28
$S_{organic}$ (%)	0.55	0.53	0.35	0.13
Acid-base accounting				
NP (kg $CaCO_3$ t^{-1})	25.3	29.8	23.5	33.6
AP (kg $CaCO_3$ t^{-1})	19.0	10.0	14.0	11.3
NNP (kg $CaCO_3$ t^{-1})	+6.3	+19.8	+9.5	+22.2

The conjugation of static and kinetic procedures allows predicting that in the short term the coal waste from Moatize Mine is not expected to produce acid drainage. Similar results were found by Pondja *et al.* (2017) in another study about the AMD with coal wastes from de Moatize site. However, results must be carefully addressed, since in the long term, there could be a consumption of alkaline minerals (carbonates) and a production of acid drainage can start due to the oxidation of pyrite without its neutralization counterpart.

Table 4 shows the total available metal content along with the respective accumulated metal content found in the leachate of fine and coarse wastes during the kinetic test (humidity cell test).

Table 4 - Content of total available metal (EPA 3050), accumulated metal from leachate in the humidity cell during the kinetic tests and percentage released (leachate metals in relation to total available metals) for the fine and coarse waste.

Element		Fine waste			Coarse waste		
		Total available metal	Leachate metal in the humidity cell	% leachate	Total available metal	Leachate metal in the humidity cell	% leachate
Al	% m/m	3.8	0.0006	0.015	4.4	0.0003	0.006
As	mg kg^{-1}	< 2.0	10.0	-	< 2	2.4	-
Ba	mg kg^{-1}	226.0	0.57	0.3	225.0	0.19	0.1
Ca	% m/m	1.4	0.67	49.8	0.94	0.04	3.7
Cd	mg kg^{-1}	< 0.2	nd	-	< 0.2	nd	-
Co	mg kg^{-1}	10.0	0.27	2.7	7.7	0.18	2.3
Cr	mg kg^{-1}	36.6	nd	-	37.9	nd	-
Cu	mg kg^{-1}	30.0	0.09	0.3	38.1	0.08	0.2
Fe	% m/m	2.3	0.01	0.3	2.1	0.004	0.2
K	% m/m	0.50	0.005	0.9	0.50	0.002	0.5
Mg	% m/m	0.37	0.14	38.9	0.42	0.04	10.4
Mn	mg kg^{-1}	327.4	13.8	4.2	295.1	3.5	1.2
Mo	mg kg^{-1}	< 0.2	nd	-	< 0.2	nd	-
Na	mg kg^{-1}	334.1	127.4	38.1	394.3	127.2	32.3
Ni	mg kg^{-1}	16.3	0.69	4.3	14.7	0.17	1.1
Pb	mg kg^{-1}	22.6	3.5	15.3	24.7	0.51	2.1
Va	mg kg^{-1}	40.1	0.46	1.1	35.9	nd	-
Zn	mg kg^{-1}	61.9	1.4	2.2	64.0	0.79	1.2

nd = not detected.

The "total available metal" corresponds to the maximum amount of metals that a sample can release to the environment, as defined by the acid digestion procedure (EPA 3050). Percentage-wise, the metals leached in greater quantities were Na, Ca and Mg, for both the coarse and fine wastes. Other metals were leached in minor concentrations, which is consistent with the geochemical mobility of elements of soils and rocks (Sposito, 2008).

Metal concentrations found in this study were compared to agricultural guideline limits for soils of different countries (Table 5), it is found that all comply with the legislation except for chromium in the United States. However, it is important to emphasize that in the humidity cells, chromium has not been leached and should not generate risks to the environment according to this test.

Table 5 - Guideline value for some metals in agricultural soils in several countries (mg kg^{-1}).

	As	Cd	Cr	Cu	Ni	Pb	Zn
Guideline							
Australia	20	3	50	50	60	300	200
Brazil	35	3	150	200	70	180	450
Canada	20	3	250	150	100	200	500
China	20-40	0.3-0.6	150-300	50-200	40-60	80	200-300
United States	0.11	0.48	11	270	72	200	1100
Finland	5	1	100	100	50	60	200
Germany	50	5	500	200	200	1000	600
Tanzania	1	1	100	200	100	200	150
New Zealand	17	3	290	> 104	N/A	160	N/A
Great Britain	43	1.8	N/A	N/A	230	N/A	N/A
Coal wastes							
Fine waste	<2.0	<0.2	36.6	30.0	16.3	22.6	61.9
Coarse waste	<2.0	<0.2	37.9	38.1	14.7	24.7	64.0

Source: US EPA, 2002; EEA, 2007; TMS, 2007; CME, 2009; EPAA, 2012; NZME, 2012; EPMC, 2015 (apud He *et al.*, 2015) and BRASIL, 2009.

According to the results obtained in this study, samples showed a low potential for acid generation and low potential for toxic metal release into the environment. Considering the huge volumes of Moatize Coal Mine, management practices for land shaping are indispensable. Fine wastes can be lodged inside coarse particles reducing the porosity and minimizing water and oxygen transfer. Furthermore, it should be stated that new waste management practices are emerging. Worldwide, there is a strong trend towards the recovery and use of coal waste as raw material in construction (dos Santos *et al.*, 2013; García Giménez *et al.*, 2016; Taha *et al.*, 2017), obtaining value-added chemical elements to be used in industries (Menezes *et al.*, 2016; Silva *et al.*, 2017), and the production of soil additives and technosols (Amaral Filho *et al.*, 2020, Wijesekara *et al.*, 2016; Weiler *et al.*, 2018; Weiler *et al.*, 2020).

Thus, some of these possibilities could be considered for these coal wastes. For example, Weiler *et al.*, 2020 presented technosol produced with fine coal waste from Moatize Mine amended with organic matter and nutrient sources to allow plant growth to assist in mine land reclamation. Aside from the erosion control and nutrient cycling already discussed, this practice reduces the downside environmental costs linked to bringing soils from borrow areas designated for mining restoration purposes. This joint provision can be performed in accordance with the logistics of the truck fleet, allowing the dynamic allocation of loading and unloading points while giving a proper use for urban wastes as well.

4. Conclusion

The conjugation of static and kinetic procedures allowed an understanding of water acidification, and geochemical mobility of elements. The acid-base accounting of the coal waste from the Chipanga seam of the Moatize Mine, both coarse and fine waste, have a slightly alkaline behavior, which is explained by the low concentration of pyrite and the presence of carbonates. The results of the static and kinetic tests showed that the material tends to form a neutral to slightly alkaline leachate. Consequently, the formation of acid drainage by the coal waste of the Moatize Mine is unlikely in the short term. Elemental analysis and leaching studies did not find significant concentrations of toxic metals that could be solubilized into the environment. The study showed that the materials would be geochemically adequate for surface disposal, opening up possibilities, such as technosols production to assist in mine land reclamation.

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