Using Humidity Cells Tests to Evaluate an ARD Minimization Approach in Santa Catarina Coal Field, Brazil

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Abstract The aim of this work was to use humidity cells tests to evaluate the effectiveness of an ARD minimisation approach for coal discard from the Santa Catarina coal field, Brazil. Coal waste sample was obtained from a typical beneficiation plant and separated into three density fractions, using a ferrosilicon medium. Further characterisation of these density fractions indicated that the low (<2.2 g/cm^3) and high (> 2.7 g/cm^3) density fractions are potentially suitable for energy and sulfuric acid production respectively. The waste fraction of intermediate density (2.2-2.7 g/cm^3) had a reduced sulphide sulfur content, and it was postulated that it may be suitable for land disposal with minimum risk to the surrounding environment. This hypothesis was tested using laboratory-scale static and humidity cell tests. The results of these tests indicated that, whilst the intermediate density waste fraction remained acid generating, the release of metals, salts and acidity into solution was considerably lower than that obtained for the discards before density separation. This has important implication in terms of reagent requirements and sludge generation during subsequent ARD treatment.

Keywords humidity cells tests, coal waste, ARD, prevention

Introduction

The grade of coal deposits in Brazil is relatively low, and approximately 65 % of the run-of-mine (ROM) coal extracted from underground mines in the carboniferous regions of Santa Catarina, Brazil, is discarded as waste in dump deposits. These waste discards contain sulfide minerals, particularly pyrite, which oxidize and give rise to acid rock drainage (ARD).

Currently, the Brazilian coal mining operations emphasise an end-of-pipe treatment approach to coal waste and ARD management. Chemical ARD treatment techniques such as lime neutralisation typically consume large amounts of expensive reagents, generate significant quantities of sludge requiring further treatment as well as disposal, and are only effective in reducing ARD risks in the short term. As pointed out by Kontopulos (1998), many of these shortcomings can be overcome by implementing ARD preventative techniques which are aimed at minimising the generation as well as subsequent dispersion of ARD from waste dump deposits over the long-term. One such approach entails the pre-disposal removal of ARD generating sulfide minerals, by means of physical separation techniques such as flotation and density separation (Amaral Filho 2010, Benzaazoua *et al.* 2000, Hesketh *et al.* 2010, Kazadi Mbamba *et al.* 2012). Apart from reducing ARD risk, integration of a sulfide removal step into the beneficiation circuit also offers opportunity for additional value recovery (Amaral Filho 2010; Hesketh *et al.* 2010, Kazadi Mbamba *et al.* 2012).

The ability to accurately predict the ARD generating potential of wastes plays an important and essential role in the development of effective approaches and technologies for mitigating associated impacts and liabilities. Methods for quantifying the ARD potential of sulfide wastes can be classified as either "static" or "kinetic" tests. Static testing methods are short term (hours to days) tests which do not take into account the relative rates of acid forming and neutralising reactions, whilst kinetic testing methods are long term (month to years) tests which allow for the study of the dynamic factors influencing ARD generation (Sapsford *et al.* 2009, Lengke *et al.* 2010). Although a number of kinetic test protocols have been developed, only humidity cell tests follow standard procedures (ASTM 2007).

The objective of this study was to use standard static and humidity cell ARD tests in order to verify the effectiveness of an intervention based on sulphur removal and low-grade coal recovery, in terms of metals leaching and acidity reduction.

Methods

The coal waste was collected from the coal preparation plant of the "Verdinho Mine", Santa Catarina State-Brazil, which extracts the "Barro Branco" seam. The material was subjected to laboratory-scale dense medium (Fe-Si) separation processing to produce three density fractions, namely < 2.2 g/cm^3, 2.2-2.7g/cm^3, and > 2.7 g/cm^3. All size fractions were subjected to standard proximate (ASTM 2007a), ultimate (ASTM 2009), and chemical speciation (ASTM 2002) analysis.

ARD prediction tests were carried out on the bulk (pre-separation) discard and intermediate density fraction (2.2-2.7 g/cm^3) samples in order to evaluate and compare their acid generating potentials. Traditional and modified acid base accounting (ABA) procedures were carried out according to the method prescribed by Sobek (Sobek 1978, EPA 1994). These tests measure the net neutralising potential (NNP), which represents the balance between the sample's maximum acid production potential (AP) and its neutralization potential (NP). The net neutralization potential (NNP) was calculated from the difference between NP and AP. A sample is classified as acid forming when it has NNP values less than −20 kg $CaCO_3$/t and as non-acid forming when it has NNP values greater than +20 kg $CaCO_3$/t. Samples are classified as uncertain when their values range from −20 to +20 kg $CaCO_3$/t. These samples were also subjected to long-term humidity cell tests in accordance with the ASTM "Standard Test Method for Accelerated Weathering of Solid Materials Using a Modified Humidity Cell" procedure (ASTM 2007b). Humidity cell leach solutions were analysed over 52 weeks in terms of pH, redox potential (Eh), acidity, and concentrations of sulphate and metals (Al, Mn, Zn, Fe).

Results and Discussion

Results of the density separation tests (Fig. 1) indicate that 17 % of the coal discards has a density of < 2.2 g/cm^3, 69 % a density of between 2.2 and 2.7 g/cm^3, and 14 % a density of >2.7 g/cm^3.

The characterization results for the bulk discards and discard fractions after density separation are shown in Table 1.

Whilst all samples had a relatively high total ash content, the low density fraction (< 2.2 g/cm^3) was enriched in carbonaceous matter (16.4 % carbon) and depleted in sulfur (1.8 % total sulphur).Previous studies (Li *et al.* 2006, Li *et al.* 2011, Muthuraman *et al.* 2010) have demonstrated the feasibility of co-combusting high ash coal with carbonaceous wastes to produce energy. The majority of the pyritic sulfur (>70 %) was reported to the high density frac-

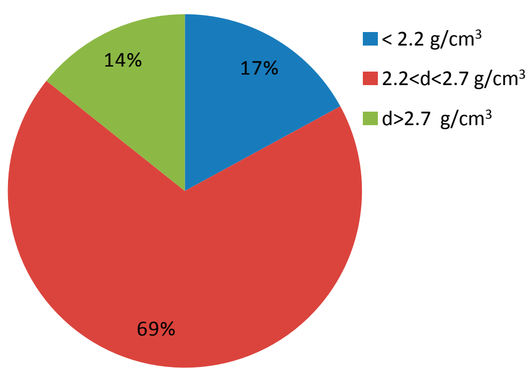

Fig. 1 *Mass deportment of coal discards to density fractions.*

	Bulk discards	Low density fraction[1] 17	Intermediate densityfraction[2] 69	High density fraction[3] 14
a) Proximate analysis %				
Ash (%)	82.7	75.8	88.3	64.8
Volatile Matter (%)	12.1	11.6	8.4	27.6
Fixed Carbon (%)	3.4	10.5	1.0	6.9
Moisture (%)	1.8	2.1	2.3	0.7
b) Ultimate analysis %*				
C (%)	6.7	16.4	3.9	8.9
H (%)	1.2	1.8	1.1	0.9
N (%)	0.3	0.5	0.3	0.3
S (%)	6.2	1.8	1.1	37.7
c) Forms of sulfur %				
Pyritic sulfur	6.0	1.3	0.7	32.5
Sulfate sulfur	0.1	0.2	0.1	0.4
Organic sulfur	0.9	0.3	0.3	4.8

Table 1 *Characteristics of the downstream discards samples pre-intervention and after gravity separation by ferrosilicon medium dense.*

tion. This fraction had a pyritic sulfur content of 32.5 %, equivalent to 60.4 % pyrite. Pyrite roasting has been used in Brazil and it is a well-known technology to produce sulfuric acid worldwide (Runkel and Sturm 2009). Although comprising the bulk of the discard material (69 % by mass), the pyritic sulfur content in the fraction of intermediate density (2.2–2.7 g/cm^3) was relatively low (0.7 %), amounting to less than 10 % of the pyritic sulfur in the feed discards.

The results of the subsequent static ARD tests, conducted on the bulk discards (before density separation) and the intermediate density fraction (2.2–2.7 g/cm^3), are summarised in Table 2. In accordance with both the traditional and modified ABA test results, all samples were classified as acid forming. This can be attributed to the negligible neutralising capacity (NP=0) of both samples, indicative of the absence of reactive neutralising minerals such as carbonates. Nevertheless, a comparison of the static test results indicate that the fraction of intermediate density has a significantly lower acid production potential (AP) and higher net neutralising potential (NNP) than the bulk discards, which can be attributed largely to the reduced pyritic sulfur content.

The time-related profiles for the humidity cells tests (HCT) conducted on the bulk discard and intermediate density fraction samples are summarized in Fig. 2. Both samples generated slightly acidic leachates from the beginning of the tests, with pH values in the region of 4.5. These pH values continued to decline steadily, stabilising at values of approximately 2–2.5 after 30 weeks. The bulk discard sample presented with slightly lower pH values than the intermediate density fraction sample throughout the experiment. Redox potentials increased from initially low values of around 300 mV to peak values of between 550 and 600mV after 13 and 22 weeks for the bulk discard and intermediate density fraction samples respectively. Redox potentials of >550-600 mV at pH values <3 are generally indicative of rapid oxidation of ferrous iron and sulfide minerals, and are normally associated with microbial activity

	AP (kg $CaCO_3$/t)		NP (kg $CaCO_3$/t)		NNP (kg $CaCO_3$/t)	
	Trad.	Modif.	Trad.	Modif.	Trad.	Modif.
Bulk discards	217.3	196.9	0.0	0.0	-217.3	-196.9
Intermediate density fraction	35.0	22.5	0.0	0.0	-35.0	-22.5

Table 2 *Static ARDprediction tests for the bulk coal discards and intermediate density fraction samples.*

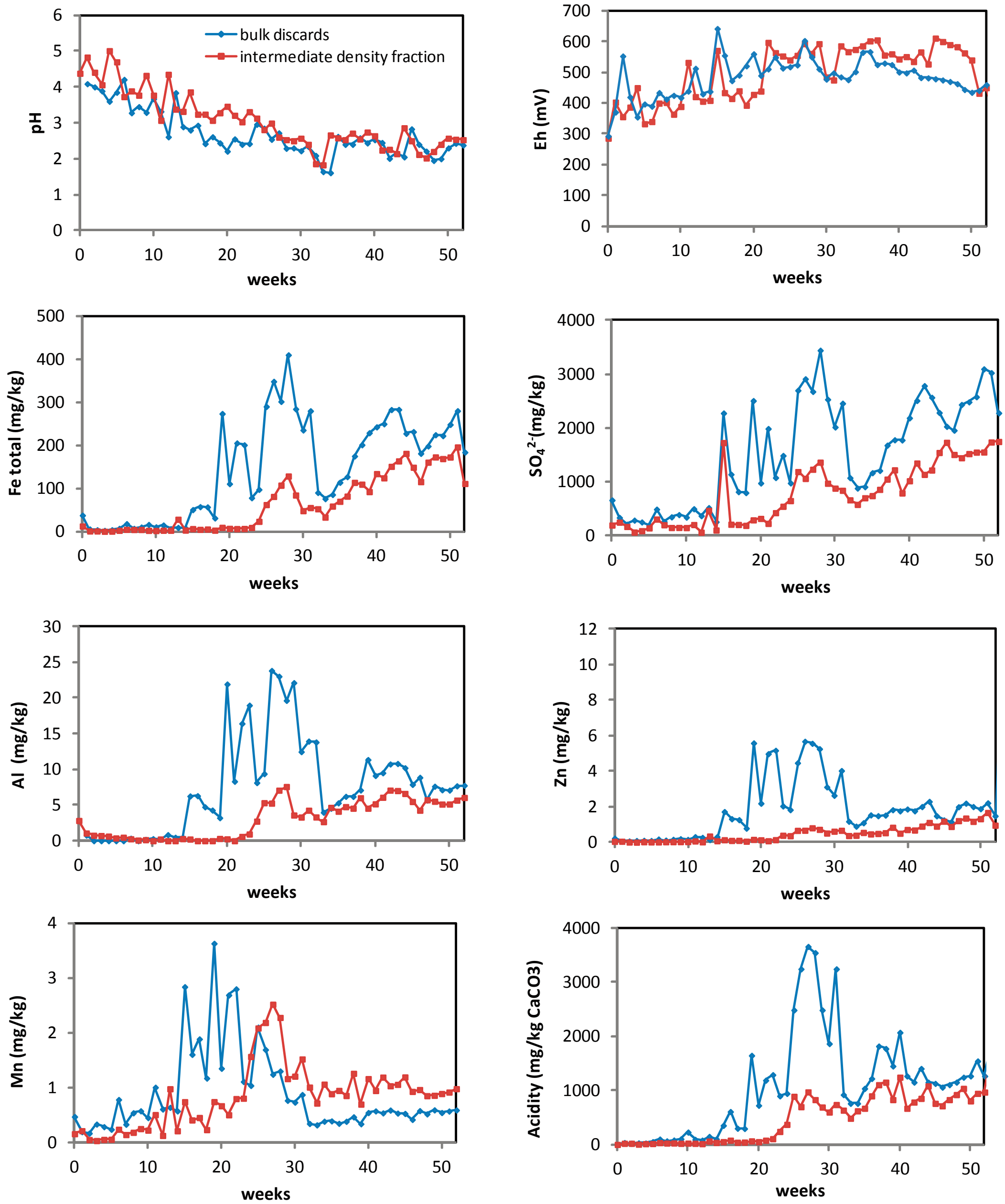

Fig. 2 *Plot of Humidity Cells Tests profiles for the bulk discard and intermediate density fraction (2.2–2.7 g/cm³) samples.*

	Bulk discards	Intermediate density fraction	Reduction (%)
Fe (mg/kg)	7630	3338	56
Al (mg/kg)	403	158	61
Zn (mg/kg)	96	25	74
Mn (mg/kg)	46	43	7
Acidity(mg/kg $CaCO_3$)	56715	24840	56
Sulfate (mg/kg)	80928	40765	50

Table 3 *Comparison of the accumulated release values over the 52 week leach period.*

(Acharya *et al.* 2001, Hesketh *et al.* 2010, Kazadi Mbamba *et al.* 2012). The significant increases in the soluble iron, sulfate and acidity values after 13 and 22 weeks for the bulk discard and intermediate fractions respectively are consistent with the on-set of rapid and extensive pyrite oxidation at these time intervals. Similar leach profiles were obtained for the trace metals, with the on-set of rapid pyrite oxidation being accompanied by a significant increase in the soluble concentrations of trace metals (Al, Zn and Mn). A comparison of the humidity cell leach profiles for the bulk discards and intermediate density fractions indicates that the leaching of concentrations and release of acidity occurs at a faster rate and to a significantly greater extent in the case of the bulk discard sample. A comparison of the accumulated release values over the 52 week leach period (Tab. 3), confirms that density separation results in a decrease in soluble metal concentrations of up to 65 %, as well as a decrease in the release of sulfate salts (50 %) and acidity (56 %). Considering raw waste cells, at 52 weeks 7630 mg/kg of accumulated iron were leached whilst accumulated released of Mn, Zn and Al were: 46 mg/kg, 96 mg/kg and 403 mg/kg respectively. In contrast, considering processed samples, at 52 weeks 3338 mg/L of accumulated iron were leached whilst accumulated released Mn, Zn and Al were 43 mg/kg, 25 mg/kg and 158 mg/kg respectively.

Conclusions

The results of this test work show that pre-separation of coal waste discards into different density fractions has the potential to generate useful by-products for energy (density <2.2 g/cm^3) and sulphuric acid (density > 2.7 g/cm^3) production, whilst reducing the amount of waste requiring disposal by 69 % (*i.e.* with a density of 2.2-2.7 g/cm^3). Static and humidity cell tests indicate, furthermore, that this separated waste fraction also has a significantly lower ARD generating potential than the bulk discard sample, resulting in reduced release of metals, salts and acidity into solution over the long-term. This, in turn, will reduce the amount of neutralising reagents required and sludge produced during subsequent ARD treatment.

Acknowledgements

The authors thank Brazilian Coal Network, CNPq, FINEP and ABCM for the support provided for this research.

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