

Pyrite utilization in the carboniferous region of Santa Catarina, Brazil - Potentials, challenges, and environmental advantages

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Jéssica Weiler^{1,2}

<https://orcid.org/0000-0002-2808-7502>

Ivo André Homrich Schneider^{1,3}

<https://orcid.org/0000-0001-9056-1925>

¹Universidade Federal do Rio Grande do Sul - UFRGS,
Departamento de Engenharia de Minas,
Porto Alegre - Rio Grande do Sul - Brasil.

E-mails: ²jessica.weiler18@gmail.com,

³ivo.andre@ufrgs.br

Abstract

The main coal seams explored in the coal region of Santa Catarina are Barro Branco (BB) and Bonito (BO). Owing to the association with the mineral matter, the tailings generated in the beneficiation are arranged in disposal areas, and subject to the generation of acid mine drainage (AMD). The objective of this study was to evaluate the use of pyrite present in the coal rejects of the BB and BO seams and the environmental gains with desulfurization. For this purpose, densimetric separability test, ash, sulfur, and AMD generation analyses were performed. In addition, the amount of pyritic concentrates and sulfuric acid were estimated considering the current level of production. Three densimetric fractions were obtained: less than 2.2 (energetic material), between 2.2 and 2.7 (low sulfur material), and greater than 2.7 g cm⁻³ (pyrite concentrate). The results revealed that the two seams could be beneficiated by gravimetric processes, obtaining pyrite concentrates with approximately 60% pyrite. By converting pyrite to sulfuric acid would represent an increase of 500,000 tons per year in the Brazilian production of this material and, in environmental terms, a reduction of up to 90% of the acidity potential to be disposed in the environment in the case of the BB seam and 65% for the BO seam. It was also observed that the “desulfurized” fraction of the BO had higher levels of pyrite and a higher potential for acidity generation than the BB seam, implying greater risks of environmental contamination and higher acid water treatment costs.

keywords: pyrite, coal waste, desulfurization, waste recovery, environment.

1. Introduction

The coal-producing region of Santa Catarina, Brazil, supplies the Jorge Lacerda thermoelectric complex, the largest thermoelectric plant in South America with an installed capacity of 852 MW (Kalkreuth *et al.*, 2010). In this region, the Barro Branco, Bonito and, to a lesser extent, the Irapuá (I)

seams are commercially exploited. Historically, the Barro Branco (BB) seam was the most exploited. However, with the depletion of many of the mines in recent years, production has also turned to the Bonito (BO) seam.

Thus, it is important that studies should be conducted aiming at the environmental aspects of both seams. Figure 1 contextualizes the Santa Catarina coal region, presenting the stratigraphic profile of the region and details of the BB and BO seams.

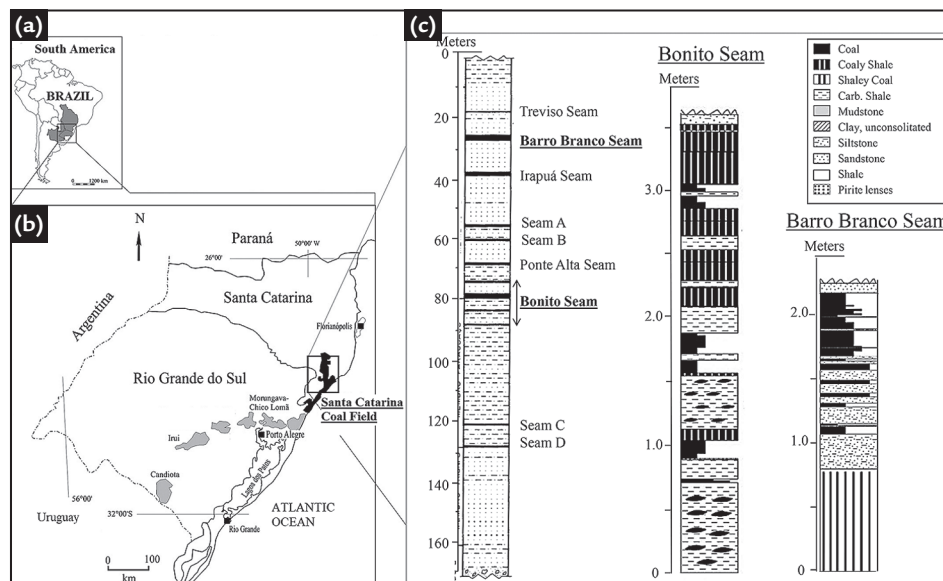


Figure 1
(a) Location of the southern Paran Basin, Brazil;
(b) Distribution of major coalfields in Rio Grande do Sul and Santa Catarina States;
(c) Transect showing lithological profile in Santa Catarina Coalfield and highlight of the Bonito and Barro Branco seams. Modified from Kalkreuth *et al.* (2010).

The extraction of these layers is performed predominantly by underground mining and, due to mechanization, intercalation with sedimentary rocks (shales, siltstones, and sandstones), and the presence of pyrite nodules (iron sulfide - FeS_2), the run-of-mine (ROM) coal should

be processed to meet the combustion standards. The coal beneficiation plants discard approximately 65% of the mass of the ROM. These discarded materials have a sulfur content close to 6%, corresponding to 11.2% of pyrite (Amaral Filho *et al.*, 2013). It is estimated that

about 320 million tons of coal wastes has already been produced in Santa Catarina (SIECESC, 2017). Figure 2 presents data on the production of ROM coal and the amount of coal rejects generated during the beneficiation phase between 1990 and 2016.

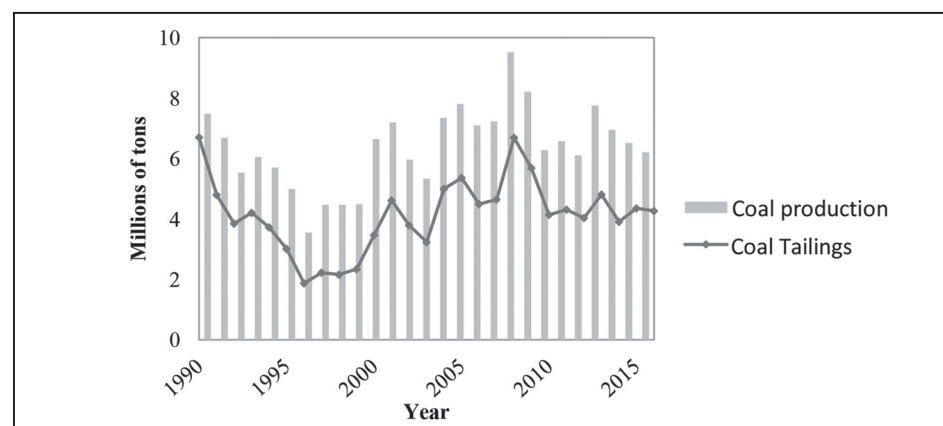


Figure 2
Coal (run of mine) and reject production in the coal region of Santa Catarina between the years 1990 and 2016 (SIECESC, 2017).

The wastes that were disposed improperly and left untreated for many years generate acid rock drainage (ARD), contaminating soils, as well as surface and underground water resources. In this region, it is estimated that 6000 ha were severely degraded, reaching 1200 km of river stretches, corresponding to 6.1% of the Ararangu, Urussanga, and Tubaro basins (Brasil, 2016; Romano Neto *et al.*, 2017). Currently, the recovery of liabilities is

being handled by a Public Civil Action with local mining companies, which is gradually mitigating the ARD effects, but many areas are abandoned and continue to impact the environment (Rocha-Nicoleite *et al.*, 2017).

Even with all the environmental problems, the coal industry does not apply techniques that aim at the recovery of the products contained in the tailings. Separation of pyrite would allow it to be used for various products, such as sulfuric

acid, avoiding considerable environmental impact and adding value to the mineral coal production chain.

The production of sulfuric acid from pyrite is achieved by roasting: heating the material at temperatures from 600 to 1000°C in an oxidizing environment (addition of air) and transforming them into gaseous sulfur dioxide. Subsequently, sulfur dioxide is oxidized to sulfur trioxide and then hydrolyzed to sulfuric acid. This well-

established technology is used worldwide (ESA/EFMA, 2000; Chepushtanova and Lukanov, 2007; Runkel and Sturm, 2009; Ashar and Golwalkar, 2013).

The objective of this study was to

2. Material and methods

Coal tailings samples were obtained during the jiggling stage, coarse circuit (with grain size between 2.0 and 50.8 mm), from the coal beneficiation processes for the BB and BO seams, located in the coal region of Santa Catarina. In both samples, the coarse fraction was selected because it contributes to more than 70% of the discarded mass. Sampling was conducted by the mining companies, and the procedure consisted in taking samples of approximately 10 kg twice a day during 1 month, numbering approximately 300 kg, which was sent to the experimental site. At the laboratory, after sun drying and homogenization, the samples were properly divided with a Jones Riffle splitter for the experimental work.

The washability test (float/sink) was carried out according to ASTM D4371-06 (ASTM, 2012a). The densities used were 1.7, 2.0, 2.1, 2.2, 2.3, 2.4, 2.5, 2.6, and 2.7 g cm⁻³. The preparation of dense medium was performed by mixing dense organic tribromomethane (CHBr₃), having a density of 2.81 g cm⁻³, and tetrachlorethylene (C₂Cl₄), having a density of 1.62 g cm⁻³. The liquids were mixed to the

measure the production of pyrite concentrates and their potential in terms of sulfuric acid conversion from the current coal exploration scale of the BB and BO seams. In addition, we evaluated the reduction of the

desired density that was measured using a densimeter. Each fraction obtained in the densimetric tests was submitted to ash and total sulfur analyses.

From the washability test, the densimetric separability curves were constructed for ash and sulfur contents, as was the Near Gravity Material (NGM) curve (Leonard, 1979; Tavares and Sampaio, 2005). The analysis of the densimetric separability curves allowed dividing the two samples (BB and BO) into three distinct densimetric fractions: fraction with a density less than 2.2 g cm⁻³ (energetic material, d < 2.2), fraction with an average density between 2.2 and 2.7 g cm⁻³ (material with reduced sulfur, 2.2 < d < 2.7), and fraction with a density greater than 2.7 g cm⁻³ (pyritic concentrate, d > 2.7).

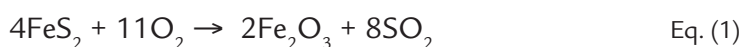
The ash analysis was performed by a gravimetric procedure according to ASTM D3174-12 (ASTM, 2012b). The total sulfur analysis (Stotal) was conducted by using instrumentation in a Leco SC 457 device. Sulfur forms (Spyritic and Ssulfate) were analyzed by titration procedures according to ASTM D2492-02 (ASTM, 2012c). The

acidity potential of the remaining material, with lower pyrite content. The results of the two seams were compared, considering the generation of coal wastes and environmental risks in the region of study.

value of the organic sulfur was calculated by the difference of the total sulfur concentration and the sum of the sulfate and pyritic sulfur. The values of ash and sulfur analyses, on a wet basis, were corrected for a dry basis according to ASTM D3173-11 (ASTM, 2011).

The information obtained from the pyritic concentrate was used to estimate the production of sulfuric acid in the region. For this purpose, the following were used: data of coal production and coal wastes generation in Santa Catarina provided by SIECESC (2017), production data/consumption of S in Brazil published in the Mineral Summary 2014 (DNPM, 2016), and molar ratios of the pyrite roasting reactions for the formation of sulfuric acid.

The thermal treatment of pyrite (FeS₂) in the presence of air current produces ferric oxide (Fe₂O₃) and sulfur dioxide (SO₂) (Eq. (1)). Sulfur dioxide is oxidized to sulfuric anhydride (SO₃) (Eq. (2)), which, by hydrolysis, is converted to sulfuric acid (H₂SO₄) (Eq. (3)). To calculate the mass ratio of pyrite required to produce sulfuric acid, the following reactions were used:



When adding the reactions, the result is the following global equation (Eq. (4)):



Thus, knowing the molar masses of pyrite (FeS₂ = 120 g mol⁻¹) and sulfuric acid (H₂SO₄ = 98 g mol⁻¹) and that for each 4 mol of pyrite (480 g FeS₂), produce 8 mol of acid (784 g of H₂SO₄), it is estimated that, for every 1 kg of pyrite, 1.6 kg of sulfuric acid is produced.

Finally, to compare the potential for acidic generation of the coal waste

before and after pyrite removal for both seams, the samples of raw waste (before processing to remove the pyrite) and the desulfurized fraction (2.2 < d < 2.7) were submitted to the static acid and base accounting (ABA) test proposed by Sobek *et al.* (1978). The acidity generation potential (AP) was measured from the total sulfur analysis (Eq. (5)).

$$AP \text{ (kgCaCO}_3 \text{ t}^{-1}) = 31.25 \times \%S \quad \text{Eq. (5)}$$

$$NP \text{ (kgCaCO}_3 \text{ t}^{-1}) = \left(\text{HCl}_{\text{cons.}} \frac{\text{g}}{\text{g}} \text{ sample} \right) \left(\frac{50}{36.5} \right) \times \%S \quad \text{Eq. (6)}$$

Determination of the neutralization potential (NP) was performed by digestion of the sample with hydrochloric acid, being heated at 90°C for 1 h to consume the neutralizing minerals, followed by NaOH titration to pH 7.0 (Eq. (6)). With the AP and NP values, the liquid neutralization potential (NNP) was calculated according to Eq. (7).

$$NNP (kgCaCO_3 t^{-1}) = NP - AP \quad \text{Eq. (7)}$$

If the difference between NP and AP is negative, then there is the potential for the residue to generate acid drainage. If it is positive, the risk is lower. Thus, the ABA criteria for identification of the acidity potential of the materials are as follows (Lapakko, 1993):

$NNP < -20 \text{ kg CaCO}_3 \text{ ton}^{-1}$: indicates the formation of acidity;

$NNP > +20 \text{ kg CaCO}_3 \text{ ton}^{-1}$: indicates that there will be no acidic formation;

$-20 < NNP < +20 \text{ kg CaCO}_3 \text{ ton}^{-1}$: difficult to predict its behavior, and other tests are necessary to predict the occur-

rence of AMD

The data obtained for the BB and BO seams were compared considering the current coal-processing scenario in the Santa Catarina region and the environmental benefits would be if the practice of pyrite concentration was adopted in the region.

3. Results and discussion

Figure 3 presents the washability curves and NGM of the coal waste from the BB and BO seams. From

these curves, it is possible to estimate the mass corresponding to the cut density, as well as the ash and sulfur

contents of the floated and immersed fractions.

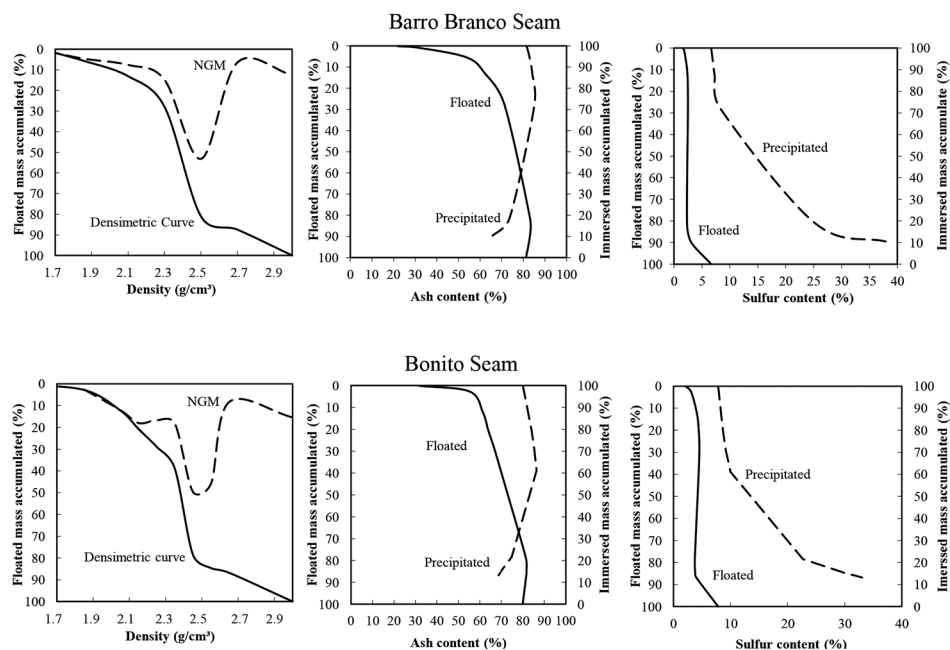


Figure 3 Washability curves of the coal waste from the BB and BO seams: near gravity material (NGM) and densimetric curve, curve of ash content, and curve of total sulfur content.

The cutting densities were defined at 2.2 and 2.7 g cm⁻³, prioritizing (a) the recovery of a carbonaceous fraction still present in the coal waste, (b) obtaining an intermediate fraction with higher mineral content and lower sulfur content, and (c) obtaining a pyrite concentrate, with more than 30% of sulfur (and, consequently, the desulfurization of the remainder of the material). The values of NGM, parameter indicating

the degree of difficulty of separation of the material, were of NGM 18% at the cutting density of 2.2 g cm⁻³ and 10% at cutting density of 2.7 g cm⁻³. In both densities, but especially in 2.7 g cm⁻³, no major separation difficulty is expected in conventional gravimetric processing equipment (Leonard, 1991; Tavares and Sampaio, 2005).

Figure 4 presents images of the material obtained for each densimetric

fraction after cutting at densities 2.2 and 2.7. It is observed that in the densimetric range of less than 2.2, the material presents a darker coloration due to the presence of carbonaceous matter disseminated in the rock. The intermediate fraction, 2.2 < d < 2.7, has a grayish appearance, with a matte appearance due to the presence of clastic materials. The fraction with d > 2.7 has pyrite in the form disseminated and/or nodules.



Figure 4 Images of the energetic fraction (d < 2.2), intermediate fraction (2.2 < d < 2.7), and pyrite rich fraction (d > 2.7) of the Bonito seam obtained after densimetric separation.

Table 1 shows the results for the mass fraction, ash content, and sulfur forms of the raw coal tailings and the three densimetric fractions obtained after cutting at densities 2.2 and 2.7 for both seams. In relation to the mass balance, the two analyzed seams presented a similar distribution in the stipulated cutting densities. The fraction with lower density ($d < 2.2$) represents 16.2% and 20.7% of

the coal waste for the BB and BO seams, respectively. Due to the amount of residual coal and lesser ash content, this fraction has potential for usage in blending with better quality coal or in co-combustion with other industrial waste that have energetic potential (Barbosa *et al.*, 2009; Li *et al.*, 2011; and Muthuraman *et al.*, 2010). The material with intermediate density ($2.2 < d < 2.7$) has the highest

mass ratio (68.8% for BB and 66.1% for BO), which can be considered as a low sulfur reject. Finally, the highest density ($d > 2.7$) represents 15.0% and 13.2% of the waste (seams BB and BO, respectively), with a total S content of 37.6% (seam BB) and 33.1% (seam BO). These values are considered adequate for the production of sulfuric acid (ESA/EFMA, 2000; Runkel and Sturm, 2009).

Table 1
Results of the mass fraction, ash content, and sulfur forms for each relative density obtained after cutting at densities 2.2 and 2.7 ($d < 2.2$, $2.2 < d < 2.7$, and $d > 2.7$) and the raw coal waste of the BB and BO seams.

	Barro Branco Seam				Bonito Seam			
	Coal waste	$d < 2.2$	$2.2 < d < 2.7$	$d > 2.7$	Coal waste	$d < 2.2$	$2.2 < d < 2.7$	$d > 2.7$
Mass (%)	100	16.2	68.8	15.0	100	20.7	66.1	13.2
Ash (%)	84.2	58.5	92.7	73.1	78.6	64.0	84.2	68.8
S_{total} (%)	6.7	1.8	1.1	37.6	7.8	4.4	3.8	33.1
S_{pyrite} (%)	5.6	1.3	0.7	32.5	6.4	3.1	2.8	29.6
$S_{sulfate}$ (%)	0.2	0.2	0.1	0.4	0.2	0.2	0.1	0.6
$S_{organic}$ (%)	1.0	0.3	0.3	4.8	1.2	1.1	0.9	2.9

Although the mass proportions of each fraction are similar for the two seams, the BB seam has advantages over the BO seam due to the following aspects:

- (a) lesser ash and sulfur content in the energy fraction $d < 2.2$
- (b) lesser sulfur content in the intermediate fraction $2.2 < d < 2.7$;
- (c) greater sulfur content in the pyrite fraction $d > 2.7$.

Thus, it is possible to perceive that the reprocessing of coal tailings in the BB seam was easier. This is due to a greater release of the material components in the BB seam than in the BO seam - carbonaceous matter, silicates, and pyrite. As can be observed in Figure 1, the BB seam is thinner but richer in coal; whereas, the BO seam is thicker,

with the predominance of carbonaceous shales (Kalkreuth *et al.*, 2010). This greater facility to separate the BB seam in relation to the BO is aware by local coal experts and has already been recorded in literature (Feil *et al.*, 2012; Mendonça Filho *et al.*, 2013).

Estimation of sulfuric acid production in the Santa Catarina coal region is shown in Table 2. The pyritic concentrate, regardless of the seam, is in the range of 30%-32% of pyritic S. Considering the average sulfur content of 31% (corresponding to 58.2% pyrite), an annual production of rejects of 4,135,000 tons per year (average between 1990 and 2016) and a 14% mass recovery of pyrite concentrate from the tailings, an annual production of 578,900 tons of pyrite concentrate (or 337,030 tons of pyrite)

is estimated. In terms of elemental sulfur, it represents 179,750 tons per year, a 25% increase in current Brazilian production of 543,000 tons per year. This is significant considering that Brazil is dependent on imports of this raw material, with an apparent sulfur consumption of 2,750,000 tons per year (apparent consumption = production + importation - exportation), largely marketed in the form of sulfuric acid, whose main use is in the production of fertilizers (DNPM, 2016). This is equivalent, in terms of sulfuric acid, to a production of 539,245 tons per year, which currently does not exist in southern Brazil. It should be emphasized that this production could be even greater if the liabilities of the coal deposits were utilized.

Table 2
Estimation of sulfuric acid production in the Santa Catarina coal region based on the pyrites concentrate obtained in the BB and BO seams.

	Santa Catarina coal region
Average coal tailings production (tons year ⁻¹)	4,135,000
Estimated pyrite concentrate production (tons year ⁻¹)	578,900
FeS ₂ content in the pyrite concentrate (%)	58.2
Estimated H ₂ SO ₄ production (tons year ⁻¹)	539,245
Estimated elementary S production (tons year ⁻¹)	179,750

Even though pyrite concentrates can be obtained from both seams, the release difference between pyrite, inert rocks, and coal implies important envi-

ronmental issues with respect to tailings generation. Table 3 presents the results of the AP, NP and NNP of the raw coal tailings samples (before densimetric

separation for pyrite concentration) and intermediate fraction after removal of the pyritic fraction) for the BB and BO seams.

Table 3
Acidity potential (AP), neutralization potential (NP), and net neutralization potential (NNP) of the raw coal waste and intermediate fractions ($2.2 < d < 2.7$) of the Barro Branco and Bonito seams.

	Barro Branco Seam		Bonito Seam	
	Coal waste	$2.2 < d < 2.7$	Coal waste	$2.2 < d < 2.7$
AP (kg CaCO ₃ t ⁻¹)	209.4	34.4	244.0	119.4
NP (kg CaCO ₃ t ⁻¹)	0.0	0.0	24.1	39.1
NNP (kg CaCO ₃ t ⁻¹)	-209.4	-34.4	-219.8	-80.3

The coal tailings of the BB deposit has a lower acidity potential than the BO seam. In addition, considering the mass ratio and AP of the intermediate fraction, the BB seam reduced the acidity by 90% compared to the raw reject and BO reduced it by only 65%. The NP, due to the presence of carbonates in the rock, was zero for the BB seam and 24.1 kg CaCO₃ ton⁻¹ for the BO seam. As the neutralization values were considerably lower than those of acidification, the NNP was negative for both rejects. The intermediate fraction of the BB seam had an NNP of -34.4 kg CaCO₃ ton⁻¹ and the BO seam of -80.3 kg CaCO₃ ton⁻¹, indicating a higher net acidity potential for the latter. When high values of acidity occur, assuming that this material is disposed in waste deposits, the risks of contamination to the environment increase, as well as the costs of the

treatment plants because of the need for a greater amount of neutralizing agents to treat ARD (Weiler *et al.*, 2016).

Thus, the gravimetric concentration of the pyrite present in the mineral coal tailings of the Santa Catarina coal region makes it possible to concentrate the pyrite and reduce the acidity generation potential of the tailings generated during the mineral processing. The production of sulfuric acid is important in the economic context of Brazil, as the country consumes large quantities for the production of fertilizers used in agriculture (Vale, 2017). It should be noted that, in the past, the company Carboquímica Catarinense (ICC), has already conducted this process, when there was a planning for pyrite's use in the region (1970-1990). However, its activities were closed for political and economic reasons (Souza, 2007). There is

an important amount of sulfur associated with Brazilian coal (DNPM, 2016) and the country fails with the breakdown of the production chain, already established in the past, not making full use of its resources. In the present day, when we talk about the substitution of linear economy for the circular economy (Blomsma and Brennan, 2017; Lèbre *et al.*, 2017), the fact is even more evident.

Even more, noteworthy are the growing exploration of the BO seam in recent years and the production decline of the BB seam (Figure 5). In the year 2000, approximately 70% of the coal production in the region was from the BB seam and 30% from the BO seam. Currently, the scenario has reversed, so that the BB seam contributes to approximately 35% of the production and the BO seam to 65%.

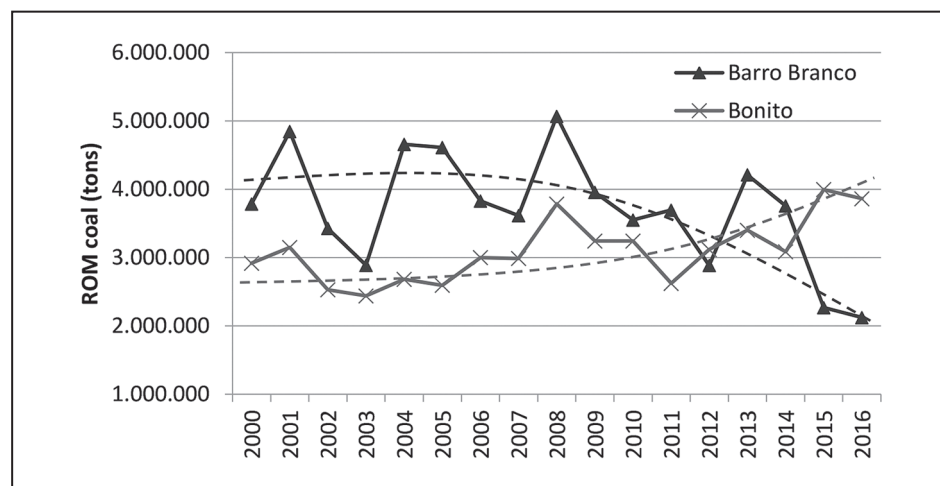


Figure 5 Comparison of the run-of-mine (ROM) coal exploitation increase of the Bonito seam and decline of its exploitation in the Barro Branco seam in the Santa Catarina coal region. Dashed lines indicate the data trend.

This change also opens challenges related to the knowledge, management, and destination of the products of the mining of the BO seam, demanding research and development actions that contribute to the regional (environmental aspects) and national scenario (minor

importation of sulfur). Among the main challenges are the suitability of the coal beneficiation plants for pyrite concentration and the environmentally correct management storage, transport, and destination of the material, avoiding for example the generation of acidic waters

and spontaneous pyritic combustion events. The effective implementation should overcome viability barriers, necessarily through technological, managerial, structural, and cultural changes of all stakeholders - society, companies, and public authorities.

4. Conclusion

The coal tailings from the Barro Branco seam and the Bonito seam can be processed by gravimetric processes to obtain concentrates with 30%-32% pyritic sulfur. When comparing the remaining coal tailings, after removal of the pyrite, it is possible to observe a considerable reduction of the acid-

ity generation potential. Owing to the greater difficulty in the concentration of pyrite, the intermediate fraction of the Bonito seam presents a higher sulfur content and a higher potential for acidity generation. Nevertheless, this study showed that the use of pyrite from coal tailings could bring economic and envi-

ronmental benefits if appropriate procedures are followed. From planning for the use of pyrite in the Santa Catarina coal region, the tailings could be used to produce sulfuric acid, contributing to the Brazilian supply of this input and reducing the environmental impacts caused by rejects disposal.

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