



## Physical and physicochemical cleaning of lignite and the effect of cleaning on biodesulfurization



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### HIGHLIGHTS

- Microbial desulfurization by an original strain as a complementary approach was applied.
- Sulfur contents of MGS and flotation concentrates were decreased 54.39% and 47.08%, respectively.
- Ash contents of MGS and flotation concentrates were decreased 67.61% and 60.95%, respectively.

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### ABSTRACT

In this work, physical, physicochemical and biological processes were combined to decrease the sulfur and ash content of finely ground coal. Two separation methods – flotation and a multigravity separator (MGS) – were studied to determine the optimum conditions of separation. The results showed that it is possible to obtain clean lignite that contains 34.36% ash and has a 77.39% combustible recovery by using MGS and 39.22% ash with a 37.38% combustible recovery by using flotation techniques. When the MGS and flotation products were treated by biological methods, the total sulfur contents of lignite were reduced by 54.39% and 47.08%, respectively, whereas the ash contents of MGS and flotation product were decreased by 67.61% and 60.95%, respectively.

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### 1. Introduction

Because most of the lignites in Turkey include high amounts of sulfur and ash, using these lignites results in a high amount of particulate matter and sulfur dioxide emissions. These emissions cause severe environmental pollution and must be reduced to an acceptable level through suitable techniques, such as by cleaning coal by physical, physicochemical, chemical and biological methods prior to combustion, removing SO<sub>2</sub> gasses by post-combustion control methods, synthetic fuel production and removing SO<sub>2</sub> during combustion techniques [1].

Physical processing methods are widely used to clean lignite fines, and gravity concentration techniques are preferred because of their low cost and ease of use. The major limiting factor for gravity concentration is its dependence on the particle size of coal. Pyritic sulfur and other inorganic materials are generally

disseminated finely and may be liberated only by fine grinding; therefore, fine coal cleaning should be applied. However, most of the modern physical processing techniques are unable to recover fine materials, so large amounts of lignites are considered lost reserves.

The recently designed multigravity separator (MGS) is a gravity concentration device that is preferred in fine coal cleaning because of its high performance; it was originally designed and used in fine metallic mineral concentration during the 1990s [2–4] before being successfully applied to fine coal cleaning [5–10]. An MGS is able to increase the gravity force up to 25 g, which improves its ability to process finer materials. Drum speed, shake frequency, shake amplitude, wash water flowrate and tilt angle are the most important variables that affect the separation of minerals in an MGS. As the particle size of the minerals becomes finer, the interaction between these parameters becomes more complicated and reduces the efficiency of the cleaning process. Therefore, studies have been conducted to determine the interactions of these variables [11,12].

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The technique of flotation enables the extraction of low-grade and complex ore bodies that would have otherwise been regarded as economically unfeasible [13], and its application range was subsequently widened in most areas. Coal flotation is an important technique because coal is a naturally hydrophobic material. Other important applications of flotation are in the recovery of fine coal particles from coal washing plant tailings [14] and recycling of power plant waste [15].

Physical methods may not be effective for separating finely dispersed minerals and minerals bound to the coal structure. Some sulfur compounds, such as its organic form, remain chemically bound in the coal and cannot be physically or physico-chemically removed [16]; in such cases, biodesulfurization may be considered an alternative separation method. Microbial desulfurization, the process of using microorganisms or their enzymes to remove sulfur compounds [18,19], is an ecologically sustainable coal-cleaning method that consumes low amounts of energy and produces non-toxic by-products [17]. The biodesulfurization of coal was first investigated with pure cultures of *Acidithiobacillus ferrooxidans* [20,21], *Acidithiobacillus thiooxidans* [22] and *Sulfolobus acidocaldarius* [23,24] and produced widely differing results. There are reports of microorganisms isolated from coal itself. Acharya and coworkers studied the desulfurization of three different types of coal using an isolate of *At. ferrooxidans* [25], and they also tried to remove sulfur from the coal from the North Eastern coalfields of Assam in India by utilizing an indigenous fungal culture (*Aspergillus* sp.) isolated from coal. Gomez et al. studied the microbial ecology of different Spanish coal and used it in a biodesulfurization process [26,27]. Aller et al. conducted desulfurization processes for various types of coal by using enriched cultures from the bacteria adhering to different the coal types as inoculate [28]. In addition, Martinez and coworkers combined physical and biological processes to reduce the sulfur and ash content of finely ground semianthracite coal and produced reductions in ash and total sulfur contents of 22% and 21%, respectively, when hydrocyclone was utilized alone. The reduction in ash content increased to 41% when hydrocyclone and flotation were both used; however, no change in sulfur elimination was found. The application of biodesulfurization treatment along with the hydrocyclone and flotation processes increased the respective reductions to 59% and 42% [29]. The differences in the values may be a result of the different coal samples used.

The aim of this work is to clean Mihalıcık region lignite reserves through the multigravity separator, flotation and biodesulfurization techniques. This work contains the second part of a research project conducted by the same authors; the first part is published elsewhere [30]. In the first part of the project, six different bacteria, five different molds and seven different yeasts were isolated from lignite itself and used to desulfurize the run-of-mine sample. Desulfurization studies have revealed that the most effective isolate is an endophytic type of fungus, and the molecular identification of its 18S rRNA gene showed that it is an *Alternaria* sp. Cf1 isolate with a GenBank accession number of KF564051. The results of the study showed that microbial desulfurization removed approximately 52% of the total sulfur.

In this work, physical and physicochemical cleaning of the same lignite was applied to reduce the ash and sulfur content of the sample before microbial desulfurization by using the same fungus. Therefore, the effect of cleaning of lignite on biodesulfurization was also investigated.

## 2. Material and methods

### 2.1. Materials

Lignite samples from open and underground mines in Koyunagılı, Mihalıcık Province, Eskisehir were used in the MGS,

flotation and biodesulfurization studies. Collected samples were mixed homogeneously in accordance with their production amounts to obtain a composite sample. These samples were crushed to minus 0.500 mm and sieved into  $-0.500 + 0.212$ ,  $-0.212 + 0.106$  and  $-0.106$  mm fractions. In the first part of this project [30], it was shown that the desulfurizing isolate was effective for the size  $-0.106 + 0.038$  mm. Therefore, the  $-0.038$  mm fraction was removed by wet sieving, and the  $-0.106 + 0.038$  mm fraction was used throughout the experimental work.

Proximate and ultimate analyses of the studied samples were performed with a LECO TGA700 (Leco Corporation, St. Joseph, Michigan, USA) to determine the ash and volatile matter amount and a LECO TGA500 (Leco Corporation, St. Joseph, Michigan, USA) to determine the calorific value. A total sulfur analysis was performed with an Eltra CS-530 analyzer (Eltra, Germany) with an infrared absorption detection procedure [31]. The major and minor elements of the samples were determined by the use of inductively coupled plasma-optical emission spectrometry (Perkin Elmer400) and XRF equipment (Thermo-ARL), respectively. The lignite was then incinerated at 550 °C and digested according to TS ISO 11466 for the minor element analyses. The sulfur forms and elements such as C, H and N were determined by following the methodologies of ASTM 2492 and ASTM 5291, respectively [31]. The characteristics of the lignite sample are listed in Table 1. In addition, the sulfur emission value (EV) was calculated by dividing the total sulfur content (wt.%) by the calorific value (MJ) [30].

Tap water and analytical grade reagents were used in the flotation experiments. Fuel oil, Philflo and Philflo + kerosene mixture (2:1 w/w) were used as flotation collectors and pine oil and sodium silicate ( $\text{SiO}_2\text{:Na}_2\text{O}$  ratio 2:1) were used as the frother and dispersant, respectively. Philflo was produced and supplied by the Chevron Philips Chemical Company, LP.

### 2.2. Concentration studies with a multigravity separator (MGS)

A Mozley C 900 laboratory multigravity separator was used in the experiments; its structure and operating conditions were previously reported [2,32]. The adjustable parameters of the MGS and their setting ranges are listed in Table 2. The drum speed, shake amplitude, shake frequency and wash water flowrate were varied and the particle size, solids ratio and feed rate were kept constant throughout the experiments.

The MGS variables were adjusted one-factor-at-a-time at the required levels, and the mixture was agitated mechanically during the experiment. A peristaltic pump was used to feed the slurry to the MGS feed vessel while the MGS was in operation. Samples from the concentrate and tailings streams were collected at steady state

**Table 1**  
Proximate, ultimate and metal analyses of the coal.

Total sulfur (wt.%)	3.42	As (ppm)	163
Calorific value (cal/g)	2152	Ba (ppm)	298
Moisture content (%)	3.85	Cr (ppm)	151
Ash (%)	54.70	Cu (ppm)	73
Volatile matter (%)	24.70	Mo (ppm)	15
Fixed carbon (%)	3.85	Pb (ppm)	21
Na <sub>2</sub> O (%)	8.7	Sr (ppm)	1515
MgO (%)	2.5	Rb (ppm)	55
Al <sub>2</sub> O <sub>3</sub> (%)	14.0	Th (ppm)	32
SiO <sub>2</sub> (%)	43.1	U (ppm)	25
K <sub>2</sub> O (%)	0.6	V (ppm)	260
P <sub>2</sub> O (%)	0.2	Y (ppm)	12
CaO (%)	6.9	Zn (ppm)	148
TiO <sub>2</sub> (%)	0.9	Carbon (%)	26.95
SO <sub>3</sub> (%)	10.1	Hydrogen (%)	2.13
Fe <sub>2</sub> O <sub>3</sub> (%)	7.2	Nitrogen (%)	1.27
MnO (%)	<0.1		

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