Fuel 192 (2017) 178–186

Contents lists available at ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Full Length Article

Evaluation of combined lignite cleaning processes, flotation and microbial treatment, and its modelling by Box Behnken methodology

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HIGHLIGHTS

- Lignite was desulfurized by the combinations of flotation and biological methods.
- Modelling of flotation parameters were performed by Box-Behnken design method.
- The fungus, *Alternaria sp.* Cf1 has proved effective in desulfurization.
- Ash content of studied sample was reduced by 70.83%.

ARTICLE INFO

Article history:
Received 7 March 2016
Received in revised form 30 November 2016
Accepted 7 December 2016

Keywords:
Coal cleaning
Coal flotation
Box-Behnken design
Biodesulfurization

ABSTRACT

In this study, first, the physicochemical cleaning of Cayirhan region's low-quality lignites was investigated by flotation to reduce ash content. In the flotation experiments, the effects of collector dosage, frother dosage, dispersant dosage, pulp density and air flow rate on ash content and combustible recovery in the concentrate fraction were optimized. Flotation tests were designed with Box Behnken design (BBD) which is a response surface method (RSM). The effective parameters and their interactions in lignite flotation are described with a mathematical model. After physicochemical cleaning, further desulfurization studies were carried out through biological treatment. *Alternaria* sp. Cf1 strain which was isolated from the same region lignites was used in microbial treatment for desulfurization studies. Sulfur emission values (EV) of studied sample, flotation and microbial treatment's concentrates were calculated and compared. After flotation and microbial treatments, sulfur emission values reduced from 12.76 to 1.01 to 0.34 for head sample, flotation and microbial treatment's concentrates, respectively that accounts for 97.37% total reduction in terms of dry ash free basis.

1. Introduction

There are several methods applied in clean coal technologies. In previous studies, physical, chemical, physicochemical and biological methods were utilized to reduce ash and sulfur content of lignites [1–4].

Coarse lignite (>0.5 mm) is generally cleaned by physical methods mainly by heavy medium separation techniques, however a decrease in particle size has a strong negative impact on efficiency of these methods.

Fine lignite (<0.5 mm) is currently recovered by physicochemical methods such as froth flotation in many countries such as China, Australia and USA in industrial scale [5]. The studies to reduce the ash content of oxidized lignites by flotation [6–10] and column flotation or mechanical flotation [11,12] are available in the literature. Desulfurization of lignites by flotation has also been investigated by many investigators with limited success [13–16].

Cleaning of lignites is currently utilized for the coarse size fractions in Turkey by physical methods. On the other hand, generation of fine lignites has been increased along with an increase in the usage of mechanized methods in lignite production [17–19]. In addition, when environmental aspects of lignite usage in energy

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http://dx.doi.org/10.1016/j.fuel.2016.12.015
0016-2361/

production are assessed, it can be said that particulate matter, gas and volatile substances emissions into the atmosphere and solid waste dams can cause significant environmental problems [20]. Therefore fine coal cleaning methods could be utilized to increase the economic value and to reduce the negative environmental impacts of these fine lignites.

Flotation method is one of the complex ore dressing processes and many variables are effective on this method. Some of these variables such as reagent addition, air flow rate, agitation rate, etc. can be controlled by the engineers but the others such as ambient temperature, cell design and type etc. cannot even if they can be measured by the appropriate devices [21]. Statistical methods are widely used to determine the effects of these controllable variables on the process performance. Although many different statistical design methods are available, the response surface method providing much data with fewer experiments has several advantages such as determination of the interaction between parameters, revealing the parabolic effects of the factors, offering simultaneous optimization of the multiple response variables on the full factorial designs [22]. There are several examples in the literature on the usage of the statistical design methods in the optimization studies concerning lignite or coal flotation [16,23–25].

Microbial treatment methods have long been used to remove inorganic sulfur as well as organic sulfur that is difficult to reduce by physical and physicochemical methods [26–30]. Recently, the combination of two or three beneficiation technologies is suggested to clean successfully low rank and high ash and sulfur content lignites [4,16].

In this study, Cayirhan region lignites were utilized to obtain a clean lignite fraction by flotation and biodesulfurization techniques. Cayirhan lignite deposit which is located mid-part of Turkey has over 365 million tons of reserves and 6 million tons of lignite is produced from underground pits by mechanized methods, annually [31]. The produced lignites are mainly sent to the nearby thermal power plant.

The purpose of this study is to reduce the ash content of lignites by flotation and to reduce the sulfur emission values by biological treatment. Sulfur emission value (EV), one of the most important indicators of environmental pollution, has been used to compare the results in this work [26,30]. Box-Behnken design (BBD), one of the statistical experimental design methods, was applied to design flotation tests.

2. Material and methods

2.1. Lignite

Lignite samples used for experiments were taken from Cayirhan/Ankara in Turkey. Proximate analyses of run of mine ore on dry basis are given in Table 1. According to this data, the samples are low rank lignite with 66.71% ash and 3.98% sulfur content. The proximate and ultimate analyses were done on dry basis. The proximate analysis of the raw and treated coal samples were done in the analyser (TGA 701; Leco Corporation, USA) by following ASTM methods [32] (Ash content: ASTM D7582/ASTM D 3174; volatile matter: ASTM D 7582/ASTM D 3175; sulfur in ash: ASTM D 5016; total sulfur: ASTM D 4239). The forms of sulfur (pyritic sulfur and sulfate sulfur) were determined by following the TS329 ISO 157. The percentage of organic sulfur was calculated by the difference. The gross calorific values were determined in a Bomb calorimeter (Model: Leco, USA) by using ASTM – D5865 standard [32].

Table 1
Characterization of run of mine lignite sample on dry basis.

Proximate analysis of ROM ore	
Ash (%)	66.71
Volatile matter (%)	29.94
Sulfur in ash (%)	2.17
Total sulfur (%)	3.98
Gross Calorific Value (MJ)	7.33
Net Calorific Value (MJ)	6.26

These samples were dried in the open air, passed through the jaw crusher and performed a size classification for enrichment by appropriate methods. The ash and sulfur content of sample according size fractions were given in Table 2.

In this study, flotation tests were carried out by using −0106 + 0.038 mm size fraction. The coarser size fractions are subjected to physical beneficiation methods as part of the scientific research project (Project ID: BAP201-315045).

2.2. Organism

The microorganism was isolated from Mihaliccik region lignites that is neighbor to the studied region lignites and identified as *Alternaria* sp. Cf1 strain (accession number KF564051) in our previous study [33].

2.3. Flotation experiments

Experimental studies were carried out using 1.5 L laboratory type Denver flotation cell. Pulp temperature was kept constant at 21 °C. Collector conditioning time, flotation time and agitation speed were also kept constant as 5 min, 70 s and 1250 rpm, respectively.

Emulsifier reactants are used to increase the performance of flotation collectors [34–36]. Toluene was used in this study as an emulsifier to increase the efficiency of fuel oil used as collector. Fuel oil and toluene mixture (1:2 by volume) were added to pulp. Methyl Isobutyl Carbinol (MIBC) was used as frother. Experiments have been carried out at natural pH and measured as 8–8.5. The obtained concentrate and tail products after each experiments were dried and weighed. Ash and sulfur contents of the products were analyzed, and combustible recovery (CR) and sulfur emission values (EV) were calculated according to the formulas (Eqs. (1) and (2)) [26,30].

$$CR\ (\%) = \frac{\%wt\ concentrate\ \times\ (100 - Ash\ Content\ of\ concentrate)}{(100 - Ash\ content\ of\ feed)} \tag{1}$$

$$EV = \frac{\%wt\ total\ sulfur\ content}{calorific\ value\ (\mathrm{MJ})} \tag{2}$$

Box-Behnken design method (BBD), a response surface method was utilized to determine the effects of the collector, frother and dispersant dosages, pulp density and air flow rate on the ash content and the combustible recovery of clean lignite. BBD is derived from 3-level factorial designs [5,37]. There must be at least three parameters to implement BBD. Graphical representation of BBD design matrix for three parameters was given in Fig. 1 [38].

Eq. (3) has been used to calculate the number of test required for BBD [38]:

$$N = 2k(k-1) + n_0 \tag{3}$$

where N is the number of experiments; k is the number of parameters, n_o is the number of experiments performed at midpoint. As seen in Fig. 1, experiments are usually performed at center levels of parameters. Design Expert 7.0.0 trial version was used for achieving design matrix and obtaining mathematical models of the response variables.