Fuel Processing Technology 173 (2018) 30–39

Contents lists available at ScienceDirect

Fuel Processing Technology

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

Research article

Ash removal from bitumen using ultrasonic falling film contactor

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A R T I C L E I N F O

Keywords:
Bitumen
Ash removal
De-ashing
Ultrasound
Falling film contactor

A B S T R A C T

In present study, Ultrasonic Falling Film (USFF) contactor has been proposed as a novel technology for enhancing ash removal from natural bitumen. Similar to froth flotation technology, separation of gangue from valuable particles using USFF is based on difference in hydrophilicity and hydrophobicity of particles. The most significant reason for positive effect of USFF is micro cavitation bubbles which play a role of producing bubbles in froth flotation technology and carry hydrophobic particles to the free surface. Micro-scale cavitation bubbles provide high surface area which gives the chance to carry more particles to the surface. Also, ultrasound induces micro-turbulence through the medium that help more dispersion of bubbles. Falling film contactor provided thin fluid layer, which intensify physical effects of ultrasound, with high contact surface area with the air, which provide more possibility to float for valuable particles, so flotation of bitumen particles become quite efficient. SEM images showed that the bubble cavitation breakage and transmutation of the particles that lead to increasing the surface area and in consequence increase in the hydrophilicity of ash-forming minerals and hydrophobicity of bitumen particles. Ash removal of 80.2% and bitumen yield of 78.7% was achieved at optimum conditions which were determined using (Artificial Neural Network) ANN and (Response Surface Methodology) RSM models.

1. Introduction

Many studies in literature have been reported for de-ashing and desulfurization of coal, which is quite close to this study subject. Common methods for this goal are physical method (such as froth flotation, washing and dense medium method) and chemical method (using some chemicals such as acid or alkali). These methods and their effective parameters have been always interested by researchers.

Mukherjee and Borthakur [1] investigated the de-ashing and desulfurization of coal from India using both aqueous sodium hydroxide (at different concentrations) and hydrochloric acid (at concentration of 10%). They concluded that by using both alkali and acid, much more recovery can be achieved compared with using each of them alone. Analyzing the processed samples was done by ASTM methods. Ash and sulfur removal increased with increasing alkali concentration. Ash, inorganic sulfur and organic sulfur removal were obtained respectively 43–50%, 100% and 10% at maximum concentration of sodium hydroxide and 10% hydrochloric acid.

Park et al. [2] tried to find a proper alternative for methyl isobutyl carbinol (MIBC) which is a very used frother in coal flotation, however is not enough safe due to its low flash point. They compared the influence of methyl cyclohexane methanol (MCHM), which is a cyclic alcohol with high flash point, with MIBC which is an aliphatic alcohol. They used two different coals and conducted measurements of frother, foam and frother film stability and also the surface tension for both frother solutions. They concluded that MCHM has more surface activity and produces more stable froth and foam compared with MIBC, so is more efficient for coal flotation.

In another investigation, Ceylan and Kucuk [3] studied the performance of two physical methods (dense medium and froth flotation) to refine three types of lignite. They concluded that flotation method with proper frother could clean the lignite more efficiently due to more pyritic sulfur removal. However, their results showed that none of the two methods are quite useful to reach high recovery.

Akdemir and Sonmez [4] showed that in froth flotation process, the particle size of coal had more effect on ash removal relative to impeller speed and pulp density. They used kerosene and methyl isobutyl carbinol as collector and frother, respectively.

Vasseghian et al. [5] used a laboratory flotation cell and in sequence leaching method by sulfuric acid to de-ashing and desulfurization of bitumen from Iran. All experiments were performed in the presence of gasoline and pine oil as collector and frothing agent, respectively. pH in all experiments was set at 7 by adding alkali and acid. They concluded that the percentage of ash and sulfur removal decreased with increase in percentage of solid weight. At optimum condition of froth flotation process followed by leaching method, the sulfur and ash removal were

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https://doi.org/10.1016/j.fuproc.2018.01.010
Received 26 October 2017; Received in revised form 10 January 2018; Accepted 11 January 2018

47% and 61%, respectively. Obtained results using Artificial Neural Network (ANN) modeling showed a good agreement with experimental results.

Ultrasonic treatment has been employed to clean the coal prior to combustion due to significant positive effects [6]. Ultrasonic waves induce some chemical and mechanical effects which make it influential in applications such as; cleaning, homogenization [7], mixing [8], heat and mass transfer enhancement [9–11] and many other fields. Acoustic cavitation and acoustic streaming are the most significant phenomena which induced during ultrasonication [11]. Pressure oscillations of ultrasonic vibrations cause formation, growth, size fluctuations and implosion of cavitation bubbles which are dominant phenomenon in low frequency ultrasound. The shock wave, produced as a result of bubbles implosion, is the most important factor which can clean the coal particles surface.

Vasseghian et al. [12] studied simultaneous ash and sulfur removal from bitumen using a flotation column equipped with 24 kHz ultrasound transducer. Effects of four experimental variables, which were the concentration of collector and frothing agent, and also power and time of ultrasonication were studied by a series of experiments. In another study carried out by Ozkan and Kuyumcu [13, 14], a flotation cell from stainless-steel, which was equipped with different configurations of 25 and 40 kHz ultrasonic transducers. The results of 25 kHz ultrasonic transducers are slightly more than those of 40 kHz. The observations showed that ultrasonic waves could distribute the froth and air bubbles in cell more uniformly and make the bubbles size smaller. They found that using ultrasonic irradiation would lead to doubling percentage of combustible recovery and ash content in refuse while concentration of consumed reagent decreased to one quarter. It is due to full contact between reagent and coal particles.

Kang et al. [15] did a series of diagnostic analyses such as XRD, XRF and SEM to find the influence of ultrasonication on the coal and pyrite composition. The obtained results showed that ultrasonicated coal samples had higher purity due to decrease in iron and sulfur and increase in carbon content of them. Also, their observations suggested that surface cleaning developed by ultrasonication caused higher hydrophobicity of coal surface and hydrophilicity of pyrite surface, so that separation performance increased. Saikia et al. [16] also illustrated the effect of irradiating ultrasound for 3 h to a coal-water-alkali solution on coal cleaning. They could drastically enhance the ash and sulfur removal from coal.

A summary of some results of the highest ash removal obtained in previous work using different methods are presented in Table 1.

It seems that reducing thickness of liquid on the ultrasonic transducer surface could lead to better results due to intensifying its induced phenomena. Also, it is clear that these cavitation bubbles have short lifetimes, so reducing the liquid height provide this opportunity for some of them to take the valuable particles to the froth zone.

In the present study, a falling film contactor, which was equipped with low frequency ultrasonic transducers, was employed to remove ash from bitumen. It is notable that choosing falling film system has advantage of creating thin fluid layer with high contact surface with air. This is the novel issue in the present study. Ultrasound energy could lead to more collisions and increasing the surface area of the particles [19, 20]. Separation using USFF technology similar to froth flotation is a method for physically separating based on difference of hydrophilicity and hydrophobicity of particles. In USFF technology, the most effective factor is the numerous produced micro-scale cavitation bubbles which could improve ash removal from bitumen. Many previous researchers proved enhancement of coal cleaning with decreasing the bubble size due to increase in the contact angle and probability of particles collision [21–23]. Based on previous work on froth flotation, it is better to produce bubbles whose diameter is comparable with particle size [24]. A sequence of experiments was designed using Central Composite Design (CCD) of Response Surface Methodology (RSM) considering five variables. For process optimization and finding the best model for ash removal prediction, RSM and Artificial Neural Network (ANN) modeling techniques were employed. The optimum operational conditions have been reported.

Table 1
The results of ash removal in previous work using different methods.

Author	Method	% ash removal
Ceylan and Kucuk [3]	Dense medium	60%
	Froth floatation	42%
Vasseghian et al. [5]	Combination of floatation and leaching with sulfuric acid	61%
Vasseghian et al. [12]	Froth floatation with ultrasound pretreatment	83.29%
Saikia et al. [16]	Ultrasound in aqueous and mixed alkali media	87.52%
Saikia et al. [17]	H_2O_2 with ultrasound	32%
Araya et al. [18]	NaOH solution	29%

2. Materials and methods

2.1. Materials and analytical tests

Bitumen samples with the particle size of 200 mesh with about 96% cumulative passing size and about 85% liberation degree were provided from mine of Kermanshah, Iran. Degree of liberation defined as mass of valuable mineral which is in the form of free particles divided by mass of total amount of that in the coal [25]. Ash composition of raw bitumen sample with $<$ 2% error was determined using XRF analysis as presented in Table 2. Gasoline and pine oil were used as collector and frothing agents, respectively.

2.2. Experimental setup

In order to investigate the effect of USFF technology on de-ashing of bitumen, some experiments was performed using an experimental apparatus schematically showed in Fig. 1. The experimental setup consisted of an inclined surface as the test section, overflow container and collection container.

Using a circulating pump, which was placed in the collection container, a flow loop was created to have a constant liquid flow as a falling film with the thickness between 0.9 and 2.5 mm depends on fluid flow rates. For measuring the mass flow rate, a flow meter (with ± 0.5% uncertainty) was located at the entrance of the overflow container. Also, a mixer was located in the collection tank to have a uniform mixture.

The inclination of the test section, which is a 50 cm × 8 cm rectangular stainless steel plate, was set at 15° toward the horizon. In order

Table 2
Ash composition of the raw bitumen.

Component	% content
So_3	64.528 ± 0.029
Cao	12.626 ± 0.012
SiO_2	10.348 ± 0.02
MgO	4.995 ± 0.089
Al_2O_3	2.638 ± 0.029
Fe_2O_3	2.155 ± 0.029
Cl	0.965 ± 0.001
K_2O	0.601 ± 0.001
SrO	0.565 ± 0.0001
V_2O_5	0.526 ± 0.001
TiO_2	0.264 ± 0.001
NiO	0.099 ± 0.0003
Mo	0.055 ± 0.0001
ZnO	0.041 ± 0.0001