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Ultrasonic flotation cleaning of high-ash lignite and its mechanism

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ABSTRACT

Lignite, as a low rank coal reserve plays an important role in energy supply. However, fine lignite is difficult to upgrade using the conventional flotation because of its high surface hydrophilicity. This investigation was to enhance the flotation recovery of high-ash lignite combined with wet-screening, scanning electron microscopy (SEM), X-ray photoelectron spectroscopy (XPS), and induction time measurer to reveal the mechanism of simultaneous ultrasonic treatment enhancing the lignite flotation. Flotation results indicate the clean coal yield is greatly increased whereas the clean coal ash content is highly reduced by simultaneous ultrasonic flotation compared with the conventional flotation. SEM and XPS results show coal surface is physically cleaned and hence the clay coating is reduced by simultaneous ultrasonic flotation. The removal of high-ash mineral particles from coal surface benefits the coal-bubble attachment. Furthermore, ultrasonic treatment creates many micro-bubbles that enhance the floatability of lignite through micro-bubble-based coating on coal surface. The micro-bubbles play a bridging role in the coal-bubble attachment. The water film on lignite surface may be also unstable and easy to rupture during bubble-coal attachment under the condition of ultrasonic treatment.

1. Introduction

Lignite is one type of low rank coals which has a large reserve around the world. In China [1], Turkey [2] and Australia [3], lignite plays an important role in the energy supply. However, fine lignite is usually refused during the mining and coal preparation processes in China because it is difficult to upgrade using both gravity separation and flotation [4].

In general, flotation is an effective method to upgrade fine coal particles based on the difference in the surface hydrophobicity between coal and gangue particles. However, lignite surface is highly hydrophilic because there are many oxygen-containing functional groups (C–O, C═O, O–C═O) on its surface [5,6]. These oxygen-containing functional groups are highly hydrophilic groups which are bonded with water through hydrogen bond, resulting a thick water film that prevents the attachment between coal surface and oil droplets/bubbles [7,8]. Additionally, lignite surface usually presents a porous property. The water can fill up the pores in coal surface, which enhances the stability of water film and the attachment of coal-bubble is further prohibited [9]. In other words, a thick water film not only prevents the attachment of bubble-coal but also prevents the adsorption of oily collectors on lignite surface. Additionally, the porosity of lignite surface causes lignite flotation to consume more collectors than middle and high rank coal flotation [10].

In the past decades, pretreatments, surface modification and effective collectors have been widely adopted to improve the flotation of lignite. Ozbayoglu et al. [11] pretreated lignite by microwave radiation in order to increase its flotation performance, and found the major moisture in coal pores were removed as well as a slight oxidation on coal surface, and hence the surface hydrophobicity of lignite was increased. Atesok and Celik [8] pretreated Soma low rank coal by dry grinding with pitch and then the ground coal achieved a greater increase in flotation recovery but a significant reduction of concentrate ash compared with non-ground coal. Cinar [12], Celik and Seyhan [13], and Ye et al. [14] applied a low temperature heating process to pretreat low rank coal and both the moisture of pores and a part of oxygen containing groups were removed [15]. The above-mentioned pretreatments are dry-based treatments. The aim of these pretreatments is to remove the moisture, oxygen-containing functional groups, and the surface coating with hydrophobic materials. However, the dry-based treatments are not well suitable for the coal cleaning industry.

Wet-based pretreatments are usually achieved using kinds of surfactant. Zhang and Tang [16] used sorbitan monooleate to condition lignite samples in the flotation cell for a period in order to introduce the surface coating of sorbitan monooleate on lignite surface and hence the flotation recovery of pretreated lignite was improve. Ni et al. [17] also found that the surfactant (polyoxyethylene sorbitan monostearate) not only coated lignite surface but also improved the froth properties, such

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as maximum height and half-life time of three-phase foam. Polat and Chander [18] found that the addition of surfactant could reduce the required amount of oily collectors and also promote the emulsification of oily collectors in the flotation of low rank coals. Kelebek et al. [19] indicated that using a combination of dodecylamine and kerosene as collector was suitable for Turkish Tuncbilek lignite flotation. Cebeci [20] claimed a combination of kerosene, emulsifier and surfactant was suitable to the cleaning of lignite by flotation. Qu et al. [21] also found that the synergism of non-ionic surfactant and oily collectors could significantly enhance the floatability of lignite. Therefore, the surfactant could improve the floatability of lignite if the usage of oily collector accompanying surfactants is recommended.

The usage of effective collectors is considered as a simply and quick way to obtain high flotation recovery of lignite. Jia et al. [22] used THF esters [$C_4H_7O{-}CH_2{-}OOC{-}R$] to successfully improve the floatability of low rank coal. Xia et al. [23] used a mixture of dodecane and 4-dodecylphenol to greatly increase the flotation recovery of lignite because dodecane can absorb on the hydrophobic sites of lignite surface while 4-dodecylphenol coat the hydrophilic sites. However, these effective collectors are difficult to apply in industrial coal flotation due to the new collectors are not very economy.

In laboratory, reverse flotation of lignite has also been widely investigated [24]. During reverse flotation, minerals are floated into the foam and discharged as the tailings while organic materials (coal) stay in the flotation pulp as the concentrate. The collectors improve the surface hydrophobicity of minerals and depressant depresses the flotation of organic materials [25]. Öztürk and Temel [26] found that the combustible matter recovery of Muş-Elmakaya lignite using the reverse flotation was higher than that using the conventional flotation. Temel [27] pointed out that the quartz caused significant contamination in the reverse flotation of Karlıova–Derinçay lignite and hence reduced the quality of clean coal. Zhang et al. [28,29] investigated the reverse flotation of lignite in the presence of sodium chloride, and found the particle size had significant effects in both flotation recovery and kinetics. Due to the low surface hydrophobicity of lignite, the reverse flotation becomes an alternative way to upgrade low grade lignite. Furthermore, the combination of gravity separation and flotation has been also proved to be an effective for the cleaning of lignite in many publications. Aksoy et al. [30] combined physical (a multi-gravity separator), physicochemical (flotation) and biological (bio-desulfurization using *Alterneria* sp.) processes to decrease both the sulfur and ash content of Koyunagili lignite. Recently, Xing et al. [10] combined cyclonic-static micro-bubble flotation column and collector emulsification to improve the flotation cleaning of Inner Mongolia lignite.

Back to the aim of this investigation, this paper is to find out the mechanism of ultrasonic flotation cleaning of high-ash lignite. As is known, ultrasonic treatment is proved to be effective in mineral surface cleaning, surface cracking, reagent emulsification. The oily collectors can be emulsified with the ultrasonic treatment and the emulsified oil droplets benefit the coal dosage and the adsorption of oily collector on mineral surface [31,32]. Therefore, less amounts of reagent are needed using the ultrasonic flotation compared with that using the conventional flotation [33,34]. Kursun [35] proved the ultrasonic pretreatment improved both the grade and recovery in zinc flotation because many cavitation bubbles are generated during the ultrasonic treatment. Similar results also found by Ozkan [36]. The cavitation bubbles could replace clay particles on coal surface and make the oil collector more effectively absorb on coal surface. In addition, the ultrasonic treatment not only reduced the froth dimensions but also increased the froth stability [37]. Cilek and Ozgen [38] applied the ultrasound in the froth phase during the flotation of a complex sulphide ore and found the ultrasound had positive effects on flotation performance under intermediate and high level airflow rates. The ultrasound was also claimed to have positive effects in the flotation de-sulphurization of high-sulfur coal [39,40]. The ultrasonic treatment was also found to be effective in ash rejection during the flotation of oil shale [41]. Slaczka [42] found the ultrasonic pretreatment increased the flotation rate of barite while decreased the flotation rate of fluorite, and hence the flotation selectivity of barite from a barite-fluorite-quartz ore was improved. Cilek and Ozgen [43] also found the selectivity of chalcopyrite was improved using ultrasonic flotation.

In the aspects of surface cleaning and cracking, Feng and Aldrich [44] used the ultrasonic preconditioning to remove both the clay layer and the oxidized layer from oxidized coal surface and the floatability of oxidized coal was improved. The oxidized layer of arsenopyrite can be also removed by the ultrasonic treatment, resulting in the increase of floatability [45]. Qi and Aldrich [46] found that the ultrasonic treatment also leaded an effective mechanical removal of zinc hydroxide from gypsum particle surface. Kang et al. [47] found that both the hydrophobicity of coal and the hydrophilicity of pyrite were increased by ultrasonic treatment. Aldrich and Feng [48] found the ultrasonic preconditioning increased the hydrophobicity of sulphides and the hydrophilicity of silicates. As the ultrasonic treatment has both oscillation and cavitation effects, surface attrition and cleaning are hence achieved. Recently, Toraman [49] used an ultrasonic generator to reduce the particle size of calcite powder, which also indicated the breakage function of ultrasound.

In this investigation, simultaneous ultrasonic treatment in the pulp zone of flotation cell was used in the flotation dashing of high-ash lignite. The mechanism of simultaneous ultrasonic treatment in lignite flotation will be fully discussed by the combination of advanced characterization technology and the given evidences in published reports.

2. Experimental

2.1. Coal samples

Lignite sample were collected from Inner Mongolia of China. The proximate analysis is listed in Table 1. It is noted that the volatile matter content is very high while the fixed carbon content is very low, which indicates the coal sample is low rank coal. In addition, the ash content of coal sample is very high, and hence this lignite belongs to high-ash coal.

Table 2 is the size composition of coal sample. This coal sample mainly consists of fine coal particles (−0.045 mm size fraction) while the ash content of −0.045 mm size fraction is 59.90%. The weight of coal particle larger than 0.045 mm is only 23.59% with ash content of 12.33%. It means the coal sample contains amounts of high-ash fines which may cause significant contamination during the flotation cleaning processes.

Table 3 is the density composition of coal sample. This coal sample mainly consists of high-ash coal particles (+1.8 kg/L density fraction). The weight of coal particle with the density less than 1.8 kg/L is only 27.13% with ash content of 16.39%. It means the coal sample contains a small part of low ash materials and is difficult to separate the low-ash components from high-ash gangue particles. In addition, this coal sample is lignite, which is low rank coal, causing an additional challenge during the flotation cleaning processes.

2.2. Flotation procedure

All flotation tests were conducted in a 1.5 L RK/FD flotation cell with the impeller speed of 1900 r/min. The pulp density was 60 g/L and

Table 1
Proximate analysis of lignite sample.

Moisture content, % (air-dried basis)	Ash content, % (dry basis)	Volatile matter content (dry ash free basis)	Fixed carbon content, % (air-dried basis)
2.68	48.68	48.95	24.83