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Enhanced desulfurizing flotation of different size fractions of high sulfur coal using sonoelectrochemical method

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ABSTRACT

Enhanced desulfurizing flotation of different size fractions of high sulfur coal was investigated using the sonoelectrochemical method. The supporting electrolyte used in this process was calcium hydroxide and the additive was anhydrous ethanol. The effects of treatment conditions on desulfurization were studied by a single-factor method. The conditions include anhydrous ethanol concentration, sonoelectrolytic time, current density, and ultrasound intensity. For the coal samples with different size fractions, the optimal experimental conditions for anhydrous ethanol concentration, sonoelectrolytic time, current density, and ultrasound intensity, respectively, are achieved. Under optimal conditions, the raw and treated coals were analyzed by infrared spectroscopy and a chemical method. Pyritic sulfur, organic sulfur, and ash are partially removed. Compared with different size fractions coal, desulfurizing flotation of high sulfur coal by sonoelectrochemistry is an effective technology.

1. Introduction

The increasing demand for coal has led to more extensive mining of coal seams, so that the mined coal has high sulfur content. Reducing sulfur content in coal prior to its use in many applications is essential. To this end, researchers have focused on searching for simple, economic, and efficient desulfurization methods as this has become an issue of worldwide concern. The process of enhanced desulfurizing flotation of coal using electrochemistry involves simple technology; this process has been reported in several published articles [1–5]. The method efficiently removes pyritic and organic sulfur, as well as ash. Some studies have reported a form of energy, ultrasound waves, that has been increasingly applied in desulfurizing flotation [6–9]. Ultrasound can enhance clean coal yield and promote the separation of pyrite in high sulfur coal, factors that are favorable for flotation desulfurization. The combination of electrochemical flotation and ultrasonic flotation methods presents promising prospects for effective desulfurization of coal.

In recent years, the integration of electrochemistry and sonochemistry has given rise to an interdisciplinary method called sonoelectrochemistry. Research in this area has attracted considerable attention, and some of the advantages reported in literature include the improvement of electrochemical progress while obtaining results superior to those obtained by electrochemistry alone [10–12]. Studies on a variety of areas including electroanalysis, electroplating, electroorganic synthesis, electropolymerization, and pollutant degradation in emulsion have used sonoelectrochemical methods. However, few reports on enhanced desulfurizing flotation of coal by sonoelectrochemistry have been published. We believe that research on the feasibility of such application is necessary. The combination of ultrasound technology and electrochemistry has also been extensively applied in the industry, leading us to expect that desulfurizing flotation of high sulfur coal via sonoelectrochemistry will find potential industrial applications.

2. Experimental

2.1. Materials and reagents

High sulfur coal from the Tunlan coal plant in Shanxi, China was used for the experiment. By screening, the coal was separated into samples of different size fractions. The date at which proximate analysis was conducted and the sulfur content of the different size fractions of coal are listed in Table 1. In the present study, first of all, coal with size fraction of 0.125–0.2 mm was chosen as the experimental coal sample. According to previous experimental experience, the concentration of slurry and supporting electrolyte were 96 g L^{-1} and 2.0 g L^{-1}, respectively. Other reagents used in this experiment were calcium hydroxide (AR grade), anhydrous ethanol (AR grade), kerosene as the flotation reagent (Chemical Pure, collector), and sec-n-octyl alcohol (CP, frother).

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Table 1
Proximate analysis and sulfur content (W%, ad) of Tunlan coal.

Granulometry	Weight	Proximate analysis			Sulfur content				Calorific
	(%)	Moisture	Ash	Volatile matter	Total	Pyritic	Sulfate	Organic	(value, kJ/kg)
− 0.076 mm	17.69	1.56	22.35	18.16	6.12	3.53	1.08	1.51	25022
0.076-0.125 mm	22.87	1.35	19.93	20.85	5.26	3.19	0.75	1.32	26510
0.125–0.2 mm	16.86	1.46	18.97	19.08	4.14	1.89	0.50	1.75	28269
0.2–0.5 mm	42.58	1.32	18.66	16.87	3.03	1.36	0.33	1.34	28179

2.2. Instruments

Electrolytic power was provided by a DH1722 DC-regulated power supply. Graphite (97.4 cm^2) and stainless steel (180.3 cm^2) were used as the anode and cathode, respectively; the electrolytic trough was set at 300 mL cup. The experimental scheme is presented in Fig. 1, and the flotation process and conditions are shown in Fig. 2.

Other equipment used in the study are an SK250HP ultrasonic cleaner (59 kHz, 250 W, ultrasonic irradiation area: 203.3 cm^2), XFGC-80 flotation cell, HXZ-S3 sulfur determinator, and Shimadzu FTIR8400S (KB_r:coal = 200:1).

2.3. Experimental method

Specific amounts of supporting electrolyte, additive, and coal sample were placed in the electrolytic trough, to which distilled water was added to obtain a volume of 250 mL. Under a certain current density, ultrasonic intensity, and agitation rate (300 rpm), the slurry was subjected to sonoelectrolysis for a preset duration at room temperature. Subsequently, the slurry was immediately transferred to the flotation column. The floating coal was washed, dried in vacuum at 80 °C for 4 h, and then subjected to sulfur determination.

The calculations of sulfur and ash reduction are expressed as [13]

$$\text{sulfur reduction (wt.\%)} = 100[x_1 - x_2(m_2/m_1)]/x_1 \quad (1)$$

$$\text{ash reduction (wt.\%)} = 100[y_1 - y_2(m_2/m_1)]/y_1 \quad (2)$$

where

m_1 is the weight of the original dried sample,
m_2 is the weight of the original dried sample after leaching,
x_1 denotes the sulfur percentage in the original sample,
x_2 represents the sulfur percentage in the coal obtained from leaching,
y_1 is the ash percentage in the original sample, and
y_2 denotes the ash percentage in the coal obtained from leaching.

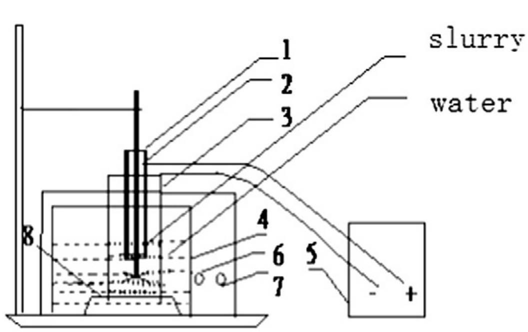

Fig. 1. Test device of sonoelectrochemistry. 1—regular speed stirrer, 2—graphite tube (anode), 3—electrolytic bath(cathode), 4—SK250HP ultrasonic cleaner, 5—DH1722 DC-regulated power supply, 6—ultrasonic output power knob, 7—ultrasonic output time knob, 8—hollow stents.

3. Results and discussion

3.1. Effect of sonoelectrolytic conditions

3.1.1. Effect of ethanol concentration on sulfur reduction

The effects of sulfur reduction, clean coal yield and sulfur content plotted against ethanol concentration are presented in Fig. 3. With increasing ethanol concentration, sulfur content initially exhibits a gradual reduction to a minimum, and then gradually increases; the sulfur reduction rate rapidly increases to a maximum, and then rapidly declines. Before the ethanol concentration reaches 1.0 mol/L, the clean coal yield exhibits a gradual fluctuation. This phenomenon can be attributed to the presence of specific numbers of ethanol molecules, which could have a function of desulfurization, resulted in the reduction of sulfur content in the clean coal using enhanced desulfurizing flotation by sonoelectrochemistry. However, as ethanol concentration reaches 1.0 mol/L, the wettability of the coal surface is enhanced and the clean coal yield begins to rapidly increase. The hydrophilicity of pyrite be weakened and resulted in part of pyrite sulfur transfer to the clean coal by flotation. Therefore, the sulfur content begins to gradually increase; and the sulfur reduction rate begins to rapidly decline. Therefore, the optimal ethanol concentration is 1.0 mol/L. These results indicate that ethanol concentration has a significant effect on sulfur reduction, in which a specific amount of ethanol proves favorable for enhanced desulfurizing flotation by sonoelectrochemistry.

3.1.2. Effect of sonoelectrolytic time on sulfur reduction

Fig. 4 depicts the sulfur reduction, clean coal yield and sulfur content plotted against sonoelectrolytic time. With time progression, the sulfur reduction rate rapidly increases to a maximum, and then slowly declines; the sulfur content initially exhibits a rapid reduction to a minimum, and then gradually increases. Initially, several groups containing sulfur are observed on the coal surface. The sulfur on the coal surface is easy to remove by the electrochemical oxidation of the anode. When a sonoelectrolytic time of about 20 min elapses, part of the inorganic and organic sulfur may have been converted into elemental sulfur because of intense oxidation; the elemental

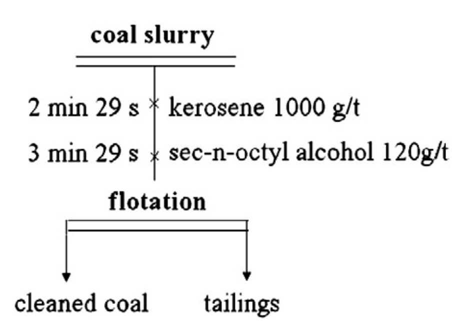

Fig. 2. Flotation experimental processing and conditions.