Removal of sulfur element from high-sulfur coal by superconducting HGMS technology

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Abstract

Coal is the most abundant fossil fuel on Earth and is used in a wide range of applications. The direct combustion of high-sulfur coal produces a large amount of sulfur dioxide, which is a toxic and corrosive gas. A new superconducting high gradient magnetic separation (HGMS) technology was studied to remove sulfur from high sulfur coal. The magnetic separation concentrate was obtained under the optimum parameters, such as a particle size of -200 mesh, a magnetic field strength of 2.0 T, a slurry concentration of 15 g/L, and a slurry flow rate of 600 ml/min. The removal rate of sulfur is up to 59.9%. The method uses a magnetic field to remove sulfur-containing magnetic material from a pulverized coal solution. It is simple process with, high efficiency, and is a new way.

Keywords: high-sulfur coal, superconducting HGMS, sulfur removal, magnetic field strength

1. INTRODUCTION

Coal is the most abundant fossil fuel on the planet and is widely used in various fields. It currently accounts for 28 % of the world's primary energy consumption [1]. The mass production and use of coal have promoted economic development and also presented severe challenges to the environment. Direct combustion of high-sulfur coal produces a large amount of sulfur dioxide, which is a toxic and corrosive gas. The direct release of sulfur dioxide is the main cause of acid rain. In the production process, sulfur dioxide will also corrode equipment, underground pipelines and so on. Therefore, it is extremely necessary to remove the sulfur from high-sulfur coal. In China, coal accounts for about 70 % of primary energy, and for a long time, the basic energy status of coal will not change. A considerable part of China's coal reserves are high-sulfur coal, and the average sulfur content of most coal fields is about 4 % [2]. China's industrial SO₂ emissions are 19.91 million tons, accounting for 85.8 % of SO₂ emissions [3]. The large amount of industrial SO₂ emissions has caused many areas to become acid rain areas. Therefore, the effective removal of sulfur from coal is of great significance for the development of the economy, the strengthening of environmental protection, and the management of severe acid rain problems.

At present, the methods of coal desulfurization include physical method, chemical method and microbial method. The physical method is simple to operate, but the treatment effect is poor. The removal effect is good by chemical method, but the process is complex. Microbiological testing conditions are harsh. Magnetic separation is a kind

* Corresponding author: lisuqin@metall.ustb.edu.cn Xingyi@ustb.edu.cn of environmentally clean separation technology [4]. Superconducting high gradient magnetic separation (HGMS) is a new type of magnetic separation technology. It is a physical separation method based on the magnetic difference of different substances [5]. It has been widely used in many fields, but combined with microwave treatment in coal desulfurization [6]. The steel wool is filled in the background magnetic field, so that the magnetic field gradient is significantly increased, and particles with different magnetic susceptibility can be separated [7]. This technique uses a superconducting coil instead of a copper coil to increase the magnetic flux density to more than 10 T, and the separation efficiency is satisfied for a weak magnetic material [8]. The purpose of our research was to remove sulfur from high-sulfur coal using superconducting HGMS technology.

2. EXPERIMENTAL

2.1. The raw materials

The coal samples used in the experiment came from two places, Jincheng and Datong in Shanxi province. The samples was ground to 200 mesh before test. The samples were analyzed by XRF and XRD. The XRF analysis results are shown in Table 1 and Table 2. According to these two tables, we can see that the coal produced in Jincheng is higher in sulfur than the coal in Datong. Jincheng's coal has sulfur content of 4.021 %, and the sulfur content of another coal is 2.579 %. The XRD results are presented in Fig. 1 (a) and Fig. 1 (c). The SEM results are shown in Fig. 1 (b) and Fig. 1 (d). It can be seen from the figure that sulfur element exists in the form of FeS and FeS₂.

TABLE 1 CHEMICAL COMPOSITION OF COAL FROM JINCHENG.

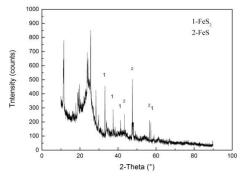
Chemical composition	O	S	Si	Al	Ca	Fe	Others
Content(%)	88.189	4.021	2.712	3.078	1.305	1.601	3.094

 ${\it TABLE~2} \\ {\it CHEMICAL~COMPOSITION~OF~COAL~FROM~DATONG}.$

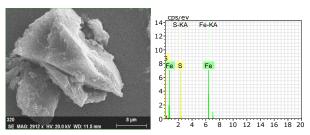
Chemical composition	0	S	Si	Al	Ca	Fe	Mg	Na
Content(%)	84.813	2.579	5.582	4.586	1.035	0.934	0.166	0.305

TABLE 3
THE MAIN PARAMETERS OF MAGNET

T4		
Item	Specification	
Central magnetic field strength	5.5 T	
Room temperature aperture	300 mm	
Length of magnet (not including iron shield)	1110 mm	
Height of magnet (not including iron and service tower)	780 mm	
Working current	150 A	
Magnets inductance	127 H	



(a) XRD pattern of the coal powders from Jincheng.



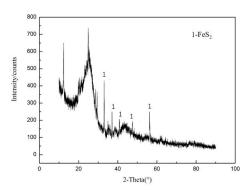
(b) SEM image and energy spectrum of the coal powders from Jincheng.

Fig. 1. Compositions and microstructure of coal.

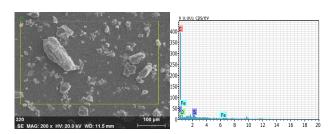
2.2. The equipment

The magnet system is mainly composed of a superconducting coil, vacuum de watts, a chiller, an electric control instrument, and an iron shield [9]. The main parameters of the magnet are shown in Table 3. The material of the separation device is polymethyl methacrylate and it has a cylindrical shape. It has a length of 440 mm and a diameter of 40 mm. The hoses are connected at both ends for slurry transfer. This device is fixed at the center of the superconducting magnetic separator during the test.

The experimental process of the separator is shown in the Fig. 2 [10]. The slurry is fed to the high gradient reactor



(c) XRD pattern of the coal powders from Datong.



(d) SEM image and energy spectrum of the coal powders from Datong.

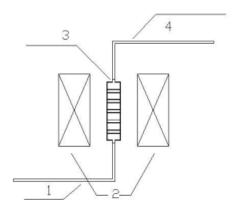


Fig. 2. Structure diagram of separating device for dynamic processing. (1. outlet, 2. Superconducting magnets, 3. separation device, 4 inlet)

through the inlet. The magnetic particles are captured by the steel wools, and the non-magnetic particles flow out through the outlet with the slurry. Magnetic particles can flow out through a magnet. In the following, the content of sulfur in non-magnetic concentrate is expressed as S(c). The content of sulfur in tailings is expressed as S(t). The content of Fe in tailings is expressed as S(t).

The inside of the separator is filled with steel wools, which captures magnetic particles. Steel wools can increase the magnetic field strength, thereby increasing the magnetic force of the magnetic particles. The steel wools were provided by Changsha Yigao Mining and Metallurgy Environmental Protection Materials, Ltd. The steel wools are made of 304 stainless steel and its diameter is 0.02 mm. Its chemical composition includes: $C \le 0.08$, $Si \le 1.0$, Si

In this study, some auxiliary equipment is needed, including ball grinder (PM10L), constant temperature oven (JXX1-277607), peristaltic pump (WT600-1F/KZ25), analytical balance (AUY220), X-ray camera (D/MAX-RB), temperature controlling magnetic stirrer (HJ-3) and so on.

3. RESULTS AND DISCUSSION

3.1. Effect of magnetic field strength

Different magnetic induction from 0.5 to 4.0 T were tried under the conditions of a particle size of -200 mesh, a slurry concentration of 15 g/L, and a slurry flow rate of 500 ml/min. The effluent concentrate and the steel wool adsorbed tailings are then subjected to chemical analysis. The results are shown in Fig. 3. It can be seen from Fig. 3 (a) and Table 1 that the sulfur content of the high-sulfur coal sample concentrate decreased from 4.02 % to 2.01 %. According to Fig. 3 (b) and Table 2, the sulfur content of the low-sulfur coal sample is reduced from 2.58 % to 1.76 %. As the magnetic field intensity increases and F_M is greater than gravity, the sulfur removal rate becomes higher and higher. When the magnetic field is too high, the phenomenon of magnetic flocculation will occur, and other non-magnetic particles will also be adsorbed and the sulfur removal rate will decrease. Therefore, 2.0 T and 2.5 T were the optimum magnetic field strengths for high sulfur coal samples and low sulfur coal samples, respectively.

3.2. Effect of particle size

The effect of particle size on separation efficiency was investigated over 100-325 mesh. The experiments were carried out with a magnetic induction of 2.0 T and 2.5 T, a slurry concentration of 15 g/L, and a slurry flow rate of 500 ml/min. The results are shown in Fig. 4. It can be seen from Fig. 4 that the particle size of the high-sulfur coal sample (produced in Jincheng) has a little influence on the separation efficiency, and the particle size of the low-sulfur coal sample (produced in Datong) has little effect on the separation efficiency. As the particle size decreased, the sulfur content in the concentrate of the high sulfur coal sample decreased by about 0.7 %. Therefore, the follow-up experiment used two samples for -200 mesh.

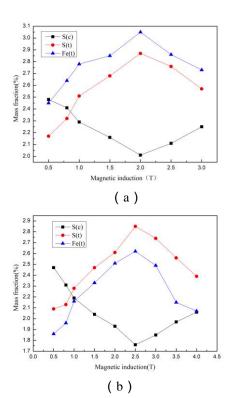


Fig. 3. The effect of magnetic induction on separation efficiency ((a) origin is Jincheng. (b) origin is Datong.)

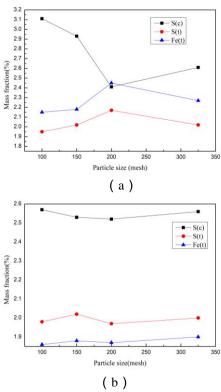


Fig. 4. The effect of particle size on separation efficiency ((a) origin is Jincheng. (b) origin is Datong.)

3.3. Effect of the slurry flow rate

According to the above experiment results, the optimal magnetic field strength and the optimal particle size were fixed at the optimum values in subsequent experiments. The effects of slurry flow rate on separation efficiency were

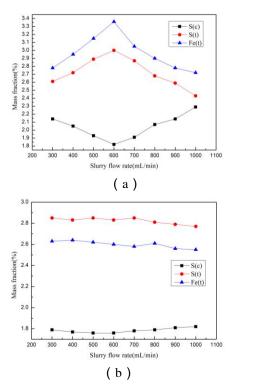


Fig. 5. The effect of slurry flow rate on separation efficiency ((a) origin is Jincheng. (b) origin is Datong.)

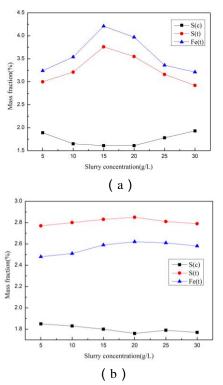


Fig. 6. The effect of slurry concentration on separation efficiency ((a) origin is Jincheng. (b) origin is Datong.)

studied under the conditions of $300 \sim 1000$ ml/min and 15 g/L. The results are shown in Fig 8. It can be seen from Fig. 5 that the slurry flow rate of low-sulfur coal has little effect on the separation efficiency in the teste range. For high sulfur coal, the experimental results are best at a flow rate of 600 ml/min. In actual production, slurry flow rate is an

important parameter affecting production efficiency. Therefore, both samples were selected to be 600 ml/min as the optimum value for the slurry flow rate.

3.4. Effect of the slurry concentration

The slurry concentration was $5 \sim 30$ g/L, and other parameters were fixed at the optimal value. The results are shown in Fig. 6. It can be seen that the concentration has a little effect on the separation efficiency, and the optimum values of the two samples were 15 g/L and 20 g/L, respectively.

Through a large number of comparative experiments, the sulfur content of coal from Jincheng was down from 4.02 % to 1.61 %. The optimum conditions were particle size of 200 mesh, magnetic intensity of 2.0 T, slurry flow velocity of 600 mL/min and slurry density of 15 g/L.

After magnetic separation, the sulfur content of coal from Datong was down from 2.58 % to 1.76 %. The best process parameters were obtained: sample size 200 mesh, magnetic flux density 2.5 T, slurry concentration 20 g/L, and slurry flow rate 600 ml/min.

3.5. Discussion

Both forms of sulfur are magnetic and have different magnetic susceptibility, (Coal: -0.59×10^{-9} m³/kg, FeS: 4321.95×10^{-9} m³/kg, FeS₂: 26.98×10^{-9} m³/kg [5]), so they can be separated by superconducting HGMS technology.

In a magnetic field, particles are subjected to three forces: magnetic force, gravity, and fluid drag. Magnetic force plays an important role in the separation process. The magnetic force (F_M) of the particle is calculated by the following equation [11].

$$F_{M} = KVH \frac{dH}{dx} \tag{1}$$

Where K is magnetic susceptibility; V is particle volume; H is magnetic intensity; dH/dx is magnetic flux density gradient.

The drag force (F_D) which is another important force can be calculated by the formula [12].

$$F_D = 6\pi \eta r_p (v_f - v_p) \tag{2}$$

Where η is the rate of viscosity, r_p is the radius of particle, v_f is the slurry flow velocity and v_p is the particle velocity.

According to the formula (1), F_M is mainly affected by particle size, magnetic susceptibility, magnetic flux density, and flux density gradient. According to the formula (2), F_D is mainly affected by particle size, slurry concentration and slurry flow rate. In summary, the factors affecting the efficiency of particle separation mainly include magnetic susceptibility, particle size, magnetic field strength, slurry flow rate, and slurry concentration. When the magnetic field is too high, the phenomenon of magnetic flocculation will occur, and other non-magnetic particles will also be adsorbed and the sulfur removal rate will decrease. When the particles are fine, agglomeration will occur and it is difficult to separate.

Only when the magnetic field force is greater than the fluid drag, magnetic particles can be separated from non-magnetic particles. Therefore, in order to obtain better separation efficiency, it is necessary to control these two forces by adjusting four parameters.

4. CONCLUSIONS

Superconducting HGMS has proven to be an effective method for the desulfurization of high sulfur coal, especially sulfur exists in the form of FeS. The results show that under appropriate parameters, particle size of -200 mesh, magnetic field strength of 2.0 T, slurry concentration of 15 g/L, slurry flow rate of 600 ml/min, the sulfur content was reduced from 4.02 % to 1.61 %. On a laboratory scale, sulfur content decreased by 59.9 %. When sulfur exists in the form of FeS₂, the sulfur content of coal was from 2.58 % down to 1.76 %, and the removal rate of sulfur was 31.78 %.

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