Research Article

Separation Process of Fine Coals by Ultrasonic Vibration Gas-Solid Fluidized Bed

Shuai Wang,1 Yaqun He,1,2 Hua Wei,1 and Weining Xie1

1Advanced Analysis and Computation Center, China University of Mining and Technology, Xuzhou 221116, China
2School of Chemical Engineering and Technology, China University of Mining and Technology, Xuzhou, Jiangsu 221116, China

Correspondence should be addressed to Yaqun He; yqhe.cumt@126.com

Received 16 March 2017; Revised 7 May 2017; Accepted 25 May 2017; Published 6 August 2017

Academic Editor: Xun Hu

Copyright © 2017 Shuai Wang et al. This is an open access article distributed under the Creative Commons Attribution License, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

Ultrasonic vibration gas-solid fluidized bed was proposed and introduced to separate fine coals (0.5–0.125 mm fraction). Several technological methods such as XRF, XRD, XPS, and EPMA were used to study the composition of heavy products to evaluate the separation effect. Results show that the ultrasonic vibration force field strengthens the particle separation process based on density when the vibration frequency is 35 kHz and the fluidization number is 1.8. The ash difference between the light and heavy products and the recovery of combustible material obtain the maximum values of 47.30% and 89.59%, respectively. The sulfur content of the heavy product reaches the maximum value of 6.78%. Chemical state analysis of sulfur shows that organic sulfur (-C-S-), sulfate-sulfur (-SO\textsubscript{4}), and pyrite-sulfur (-S\textsubscript{2}) are confirmed in the original coal and heavy product. Organic sulfur (-C-S-) is mainly concentrated in the light product, and pyrite-sulfur (-S\textsubscript{2}) is significantly enriched in the heavy product. The element composition, phase composition, backscatter imagery, and surface distribution of elements for heavy product show concentration of high-density minerals including pyrite, quartz, and kaolinite. Some harmful elements such as F, Pb, and As are also concentrated in the heavy product.

1. Introduction

Coal is an important primary energy source worldwide, especially in China. In China, coal-fired power is predominant in the production of the Chinese electric power and accounts for more than 70% [1]. Coal contains appreciable quantity of inorganic minerals and harmful elements, such as sulfur, lead, arsenic, and mercury; these minerals and elements can be transformed into inhalable particles, acid rain, and other pollutants and can be discharged into the atmosphere during coal combustion [2], thereby resulting in serious pollution to the atmosphere and large economic losses [3–6]. Thus, desulfurization and deashing of coal prior to combustion are important to prevent fog haze weather.

Although lump coals are separated prior to milling to remove large pieces of waste rock, fine waste rocks containing harmful elements inlay in the coal are not always removed. If coal is sufficiently crushed to fine particles, then the mineral particles can be fully dissociated from the coal. Such condition is favorable to the separation process. Fine coals are currently separated mainly by flotation, which can effectively separate <0.5 mm fraction coal [7–9]. The development of cyclonic-static microbubble flotation column and new reagent systems has enabled good results of low-rank coal separation [9–13]. However, the flotation process consumes large amounts of water, and its development in arid regions is limited by water resource deficiency. Thus, high-efficiency dry separation technology of fine coals should be investigated.

Dry separation technologies, especially the separation technology of the gas-solid fluidized bed, are currently used to separate coal [14–19]. For example, air dense medium fluidized bed is used to effectively separate coal of 50–6 mm size fraction [20–24]. Xu and Zhu [25] examined the influence of vibration parameter on the fluidization characteristics of fine materials. Luo et al. [26] separated coal of 6–1 mm size fraction by use of air dense medium fluidized bed and analyze the particle force condition. Yang et al. [27, 28] used
vibrated fluidized bed to separate coal of 6−3 and 3−1 mm size fractions without a dense medium. These abovementioned dry separation methods can effectively separate >0.5 mm fraction coal but present difficulty in separating <0.5 mm fraction coal and exhibit many limitations. Thus, new dry separation methods to deal with fine coal of <0.5 mm fraction should be explored.

This study investigated the separation process of the 0.5−0.125 mm fraction coal with ultrasonic vibration gas-solid fluidized bed. The composition of products under different experimental conditions was studied by advanced analysis and test methods to evaluate the separation results.

2. Materials and Methods

2.1. Sampling and Experimental Device. The 0.5−0.125 mm fraction coal was chosen to study the separation process. The ash content of the coal was 36.21%, and its sulfur content reached 2.82%. The coal was obviously of high sulfur content. The schematic of experimental system is shown in Figure 1. The system included air supply and separation systems. The air supply system included a roots blower, an air reservoir, a vortex flow meter, and an air valve. The separation system included a gas-solid fluidized bed and an ultrasonic vibration device. The composition of products under different experimental conditions was studied by advanced analysis and test methods to evaluate the separation results.

2.2. XRF Analysis. X-ray fluorescence spectrometer (XRF, S8 Tiger, Bruker, Germany) was applied to the sulfur content analysis for different products to study the separation effect at different experimental conditions. The XRF worked at 20 kV−60 kV and 10 mA−100 mA; the collimator angle was 0.23°.

2.3. XRD Analysis. The phase composition analysis was run with an X-ray diffractometer (XRD, D8 Advance, Bruker, Germany) for heavy product from separation process with a voltage of 40 kV and a current of 30 mA. XRD data were recorded in a scanning mode from the detective angle of 3°−90° with the step of 0.01945° (step) and the scanning speed of 0.1 s/step.

2.4. XPS Analysis. X-ray photoelectron spectrometer (XPS, ESCALAB 250Xi, Thermo Fisher, America) with AlKa radiation (hv = 1486.6 eV) and a 900 μm light spot size was used to analyze the chemical state of sulfur in the coal.

2.5. EPMA Analysis. Field emission electron probe microanalyzer (EPMA, 8050G, Shimadzu, Japan) was applied to the microstructure, backscatter imaging, and area distribution of element analysis for the heavy products. The beam size was Min, the BC electric current was 10−100 nA, and the testing voltage was 15 kV.

3. Results and Discussion

3.1. Effect of Operating Parameters on Separation. The separation results in Figure 2 present the influence of vibration
Table 1: Sulfur content of each layer of product.

| Layer number | 0 Hz  | 20 kHz | 25 kHz | 30 kHz | 35 kHz | 40 kHz |
|--------------|------|--------|--------|--------|--------|--------|
| First layer  | 1.52 | 1.38   | 1.35   | 1.33   | 1.26   | 1.48   |
| Second layer | 2.11 | 1.96   | 1.93   | 1.86   | 1.82   | 1.98   |
| Third layer  | 2.23 | 2.18   | 2.26   | 2.14   | 2.22   | 2.08   |
| Forth layer  | 2.26 | 2.32   | 2.35   | 2.28   | 2.25   | 2.29   |
| Fifth layer  | 6.56 | 6.75   | 6.73   | 6.68   | 6.89   | 6.77   |

Frequency ranging from 20 kHz to 40 kHz and no vibration at a fluidization number of 1.6 and a separation time of 30 s. The ash difference between the light and heavy products and the recovery of combustible material are all minimal at 29.08% and 83.13%, respectively, under no vibration condition. With the increase in vibration frequency from 20 kHz to 35 kHz, the ash difference between the light and heavy products and the recovery of combustible material increase and reach the maximum values of 47.07% and 89.04%, respectively, at 35 kHz. After 35 kHz, the ash difference and the recovery of combustible material decrease. Thus, the addition of ultrasonic vibration field intensifies the separation process of fine coal under different densities.

Table 1 shows the sulfur content of each layer of product at different vibration frequencies. Sulfur is mainly concentrated in the fifth layer, the contents of which are all above 6%, and the maximum value is 6.89% when the vibration frequency is 35 kHz. On the contrary, the sulfur contents of the first layer at different vibration frequencies are low at below 1.6%. The minimum value is 1.26% when the vibration frequency is 35 kHz.

Figure 3 shows the separation results obtained at different fluidization numbers at a vibration frequency of 35 kHz and at a fluidizing time of 30 s. At low fluidization number, the bed liquidity is poor and the resistance to particle sedimentation is high. Thus, the separation of the coal and high-density minerals becomes difficult. With the increase in the fluidization number, the bed fluidity and the separation effect also increase. The ash difference between the light and heavy products and the recovery of combustible material obtain the maximum values of 47.30% and 89.59%, respectively, when the fluidization number is 1.8. Thereafter, the separation effect decreases with the further increase in the fluidization number. The reason is that the bed stability is destroyed because of the increase in gas velocity, thereby leading to serious back mixing between the light and heavy products.

Table 2 shows the sulfur content of each layer of product at different fluidization numbers. Similarly, sulfur is mainly concentrated in the fifth layer, and the maximum value is 6.78% when the fluidization number is 1.8. The sulfur contents of the first layer are all below 2.0%, and the minimum value is 1.22% when the fluidization number is 2.0.

3.2 Component Analysis of Products. The results of the main element content analysis of the original coal and light and heavy products are shown in Table 3. In the light product, the contents of magnesium, calcium, iron, silicon, aluminum, and sulfur decrease compared with those in the original coal. On the contrary, these contents obviously increase in the heavy product, especially those of sulfur and iron.

The XPS spectra of sulfur in different samples are shown in Figure 4, and the content of sulfur in different chemical
Table 2: Sulfur content of each layer of product.

| Layer number | Sulfur content/% |
|--------------|------------------|
|              | 1.2  | 1.4  | 1.6  | 1.8  | 2.0  | 2.2  | 2.4  |
| First layer  | 2.12 | 1.76 | 1.34 | 1.25 | 1.22 | 1.58 | 2.25 |
| Second layer | 2.23 | 2.03 | 1.92 | 1.78 | 1.80 | 1.89 | 2.28 |
| Third layer  | 2.33 | 2.38 | 2.02 | 1.84 | 1.92 | 2.44 | 2.96 |
| Forth layer  | 2.45 | 2.27 | 2.13 | 2.10 | 2.15 | 2.53 | 3.12 |
| Fifth layer  | 5.36 | 6.15 | 6.33 | 6.78 | 6.59 | 5.62 | 4.98 |

Table 3: Elemental composition of different samples.

| Sample name     | MgO   | CaO   | Fe₂O₃ | Al₂O₃ | SiO₂  | S     | P     |
|-----------------|-------|-------|-------|-------|-------|-------|-------|
| Original coal   | 0.16  | 2.74  | 2.02  | 11.91 | 13.06 | 2.43  | 0.021 |
| Light product   | 0.10  | 0.61  | 0.63  | 9.76  | 12.07 | 1.66  | 0.022 |
| Heavy product   | 0.58  | 2.03  | 3.68  | 20.38 | 39.28 | 6.78  | 0.029 |

Figure 4: X-ray photoelectron spectroscopy spectra of the coal: (a) original coal; (b) light product; (c) heavy product.
Table 4: Content of sulfur in different chemical states.

| Name       | Original coal |         | Heavy product |         |
|------------|---------------|---------|---------------|---------|
|            | Peak BE       | Atomic%| Peak BE       | Atomic%|
| -SO\(_4\)  | 168.91        | 40.75  | 168.88        | 24.03  |
| -C-S       | 164.06        | 45.75  | 163.97        | 23.43  |
| -S\(_2\)   | 162.78        | 13.5   | 162.7         | 52.54  |

The backscatter imagery of different minerals in heavy product by qualitative analysis is shown in Figure 7. Obviously, high-density minerals such as pyrite, quartz, kaolinite, chalcopyrite, gypsum, and iron oxide are effectively separated in the separation process.

Some harmful elements such as Pb, As, and F are also found in the heavy product by qualitative analysis as shown in Figure 8. These particles contain harmful elements that are generally distributed in large particles with tiny sizes. In addition, S and Fe always appear in the same particle containing the abovementioned harmful elements. This condition means that pyrite is the important medium for those elements. Thus, removal of pyrite is the key to the desulfurization and detoxification of fine coal.

4. Conclusion

The 0.5–0.125 mm fraction fine coal is separated effectively by ultrasonic vibration gas-solid fluidized bed. After adding the ultrasonic vibration force field, the particle separation process based on density is strengthened, and the best result appears when the vibration frequency is 35 kHz. The ash difference between the light and heavy products and the recovery of combustible material reach the maximum values of 47.07% and 89.04%, respectively. Airflow velocity significantly influences the separation. When the fluidization number is low, the bed liquidity is poor and the resistance to particle sedimentation is high. Conversely, the bed stability is destroyed and the back mixing between the light and heavy products occurs, thereby hindering the separation. When the vibration frequency is 35 kHz and the fluidization number is 1.8, the ash difference between the light and heavy products and the recovery of combustible material obtain the maximum values of 47.30% and 89.59%, respectively. Sulfur is mainly concentrated in the heavy product, and the content reaches the maximum value of 6.78%.

The XPS results show that -C-S-, -SO\(_4\), and -S\(_2\) exist in the original coal and heavy product; however, the content of -S\(_2\) in the heavy product is higher than that in the original coal. On the contrary, -C-S- mainly exists in the light product. According to the peak area, pyrite and sulfate are evidently enriched in the heavy product.

The backscatter imagery and surface distribution of elements in heavy product are shown in Figure 6. In the backscatter imagery, high-density particles with high average atomic number are bright, especially the particles that contain iron. The main elements in the heavy product are Si, Al, and Ca. In other words, aluminosilicate minerals are the main minerals in the heavy product. In addition, sulfur and iron exist in the heavy product. The distribution of sulfur is also the same as that of iron, thereby indicating that S exists in the form of pyrite. Therefore, pyrite has been separated during the separation experiment.

Thus, removal of pyrite is the key to the desulfurization and detoxification of fine coal.
Figure 6: Surface distribution of elements in heavy product: (a) backscatter imagery of heavy product; (b) Si; (c) Al; (d) Ca; (e) Fe; (f) S.

Figure 7: Backscatter imagery of different mineral in heavy product: (a) pyrite; (b) quartz; (c) kaolinite; (d) iron oxide; (e) chalcopyrite; (f) gypsum.
Figure 8: Qualitative analysis of particles containing harmful elements: (a) lead particle; (b) fluoride particle; (c) arsenic particle.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

Acknowledgments

This project received funding from the National Natural Science Foundation of China (51404267 and 51574234). The authors would like to acknowledge the Advanced Analysis and Computation Center of China University of Mining and Technology for their technical support.

References

[1] W.-N. Xie, Y.-Q. He, Y.-H. Zhang et al., “Simulation study of the energy-size reduction of MPS vertical spindle pulverizer,” Fuel, vol. 139, pp. 180–189, 2015.
[2] Q. Zhang, Z. Shen, J. Cao et al., “Variations in PM2.5, TSP, BC, and trace gases (NO2, SO2, and O3) between haze and non-haze episodes in winter over Xi’an, China,” Atmospheric Environment, vol. 112, pp. 64–71, 2015.
[3] H. F. Zhang, J. Hu, Y. X. Qi et al., “Emission characterization, environmental impact, and control measure of PM2.5 emitted from agricultural crop residue burning in China,” Journal of Cleaner Production, vol. 149, pp. 629–635, 2017.
[4] Y. L. Zhang and F. Cao, "Is it time to tackle PM2.5 air pollutions in China from biomass-burning emissions?" *Environmental Pollution*, vol. 202, pp. 217–219, 2015.

[5] X. Fan, G.-F. Liu, Z.-M. Zong et al., "Ni-and Pd/C-catalyzed hydrodenitrogenation of isoquinoline," *Energy Sources, Part A: Recovery, Utilization and Environmental Effects*, vol. 37, no. 1, pp. 55–60, 2015.

[6] X. Fan, G.-F. Liu, Z.-M. Zong et al., "Mechanism for catalytic hydrodenitrogenation of isoquinoline," *Fuel Processing Technology*, vol. 106, pp. 661–665, 2013.

[7] Y. W. Xing, X. H. Xu, X. H. Gui, Y. J. Cao, and M. D. Xu, "Effect of kaolinite and montmorillonite on fine coal flotation," *Fuel*, vol. 195, pp. 284–289, 2017.

[8] K. B. Tankersley, L. A. Owen, N. P. Dunning et al., "Micro-flotation removal of coal contaminants from archaeological radiocarbon samples from Chaco Canyon, New Mexico, USA," *Journal of Archaeological Science: Reports*, vol. 12, pp. 66–73, 2017.

[9] K. Jiang, J. E. Dickinson, and K. P. Galvin, "Two-stage fast flotation of coal tailings using reflux flotation," *Minerals Engineering*, vol. 98, pp. 151–160, 2016.

[10] Y. Xing, X. Gui, Y. Cao, D. Wang, and H. Zhang, "Clean low-rank-coal purification technique combining cyclonic-static microbubble flotation column with collector emulsification," *Journal of Cleaner Production*, 2015.

[11] X. Gui, Y. Xing, G. Rong, Y. Cao, and J. Liu, "Interaction forces between coal and kaolinite particles measured by atomic force microscopy," *Powder Technology*, vol. 301, pp. 349–355, 2016.

[12] W. Xia, J. Yang, and C. Liang, "A short review of improvement in flotation of low rank/oxidized coals by pretreatments," *Powder Technology*, vol. 237, pp. 1–8, 2013.

[13] J.-L. Xia, X. Fan, C.-Y. You, X.-Y. Wei, Y.-P. Zhao, and J.-P. Cao, "Sequential ultrasonic extraction of a Chinese coal and characterization of nitrogen-containing compounds in the extracts using high-performance liquid chromatography with mass spectrometry," *Journal of Separation Science*, vol. 39, no. 13, pp. 2491–2498, 2016.

[14] S. Wang, Y. He, J. He, L. Ge, and Q. Liu, "Experiment and simulation on the pyrite removal from the recirculating load of pulverizer with a dilute phase gas-solid fluidized bed," *International Journal of Mining Science and Technology*, vol. 23, no. 2, pp. 301–305, 2013.

[15] S. A. Macpherson, S. M. Iveson, and K. P. Galvin, "Density based separations in the Reflux Classifier with an air-sand dense-medium and vibration," *Minerals Engineering*, vol. 23, no. 2, pp. 74–82, 2010.

[16] J. Oshitani, T. Kawahito, M. Yoshida, K. Gotoh, and G. V. Franks, "The influence of the density of a gas-solid fluidized bed on the dry dense medium separation of lump iron ore," *Minerals Engineering*, vol. 24, no. 1, pp. 70–76, 2011.

[17] J. He, Y. Zhao, Z. Luo, Y. He, and C. Duan, "Numerical simulation and experimental verification of bubble size distribution in an air dense medium fluidized bed," *International Journal of Mining Science and Technology*, vol. 23, no. 3, pp. 387–393, 2013.

[18] L. Dong, Y. Zhao, C. Duan, Z. Luo, B. Zhang, and X. Yang, "Characteristics of bubble and fine coal separation using active pulsing air dense medium fluidized bed," *Powder Technology*, vol. 257, pp. 40–46, 2014.

[19] Y. Q. He, H. F. Wang, C. L. Duan, and S. L. Song, "Airflow fields simulation on passive pulsing air classifiers," *Journal of the South African Institute of Ming and Metallurgy*, vol. 105, pp. 525–531, 2005.