Clean Desulphurization of High-Sulfur Coal Based on Synergy Effect between Microwave Pretreatment and Magnetic Separation

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ABSTRACT: Coal is the world’s second largest energy resource and the primary energy resource in China. Coal-fired power generation is the dominant source for obtaining power both in China and the world. This situation is considered to remain unchanged for the coming few decades. The paper is to study the mechanism of microwave absorption capacity of the magnetic medium. A mechanism for the microwave energy absorption of fine coal is proposed by obtaining the parameters of dielectric properties of the magnetic medium. The prediction model of electromagnetic parameters during the process of microwave dielectric enhancement of fine coal is established by using the neural network method. The content of magnetic medium, by increasing the electromagnetic parameters to improve the energy utilization rate and reduce energy loss, can effectively improve the absorption and broaden bandwidth of the coal sample, improving the frequency adaptive design of coal by the microwave pretreatment process. A neural network model for the prediction of the electromagnetic parameters of fine coal is established by using the neural network method. The desulfurization rates were 23.99 and 45.78% for Weinan and Lu’an coals, respectively.

1. INTRODUCTION

Coal is the world’s second largest energy resource and is the primary energy resource in China. Coal-fired power generation is the dominant source for obtaining power both in China and the world. This situation is considered to remain unchanged for the coming few decades. Globally, wet coal preparation technology was in use to prepare coal. However, with the new coal mining areas transferred to cold and arid regions, over two-third of the coal resources is now scattered all over the drought hit region of western China. Therefore, it is urgent to study efficient dry separation technology. The existing dry separation technology mainly includes wind jiggling machines,5−7 compound dry separation technology,8−9 and dry dense medium fluidized bed technology.10−13 At present, desulfurization and separation technology of fine coal has become a hot research topic in dry coal preparation technology. Generally, the medium of dense phase gas–solid fluidized bed is magnetite powder. In the process, the complicated abrasion condition of magnetite particles and formation process of fine particles can be analyzed by studying the abrasion model and particle size distribution of the magnetite particles.13 Chemical and biological desulfurization technologies can remove inorganic sulfur and organic sulfur in coal, but problems such as high cost, long production cycles, and pollution still remain to be addressed. Physical desulfurization methods can only remove inorganic sulfur and are economical and practical in nature.14,15 High-gradient magnetic separation technology is an effective method for physical desulfurization, which can be used to deal with fine particles of paramagnetic minerals and is applied to the desulfurization of fine coal (mainly pyrite).16,17 The magnetic properties of pyrite can be improved by heating treatment.18 Compared with coal, pyrite can be heated more rapidly due to its higher dielectric constant.19 Microwave radiation is an effective technique for heat treatment and has the advantages of rapid heating process, selective heating, and uniform heat distribution.20 The magnetic properties of the pyrite in the coal are enhanced, which is more favorable for the magnetic separation technology.21−23 For low-rank coal (lignite), microwave heat treatment can significantly reduce the water content and increase its calorific value and fixed carbon content, hence improving the quality of lignite energy.24,25 Additionally, microwave heat treatment is also conducive to act as a followup treatment step to the frictional electrical selection operation.26 Microwave heat treatment selectively gets absorbed in the pyrite and improves the absorption ability of the fine coal layers for microwaves on a magnetic medium. Due

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to these characteristics, the magnetic field gradient and magnetic properties of the pyrite have been enhanced and desulfurization effect has been optimized.\textsuperscript{27}

The main research content of this paper is to study the mechanism of microwave absorption capacity of the magnetic medium. A mechanism for the microwave energy absorption of fine coal is proposed by obtaining the parameters of dielectric properties of the magnetic medium. The prediction model of electromagnetic parameters during the process of microwave dielectric enhancement of fine coal is established by using the neural network method.

2. EXPERIMENTAL SYSTEM AND MECHANISM

2.1. Experimental System. E5071C Vector-Network analyzer was used to measure dielectric and electromagnetic properties of an radio frequency network formed by the samples. The analyzer has an adjustable range of test frequency from 300 kHz to 8.5 GHz, with a typical scanning rate of about 9.6 ms/p. It possesses functions for powerful analysis, error correction, and calibration. Microwave pretreatment was performed in a nitrogen/air processing system, as shown in Figure 1. The system consists of a nitrogen protective device, microwave treatment device nitrogen controller, and nitrogen storage device. A microwave frequency of 2.45 GHz was set for the treatment, with a continuously adjustable power, ranging from 0 to 1000 W. The RE Roll adopted a novel punch-out type permanent magnet-based separation and purification equipment. Table 1 shows the numerical analysis for sulfur in four kinds of test samples.

Table 1. Morphological Analysis of the Sulfur of the Coal Samples

| no. | raw coal    | $S_{t.d}$ % | form of sulfur | $S_{p.d}$ % | $S_{s.d}$ % | $S_{o.d}$ % |
|-----|-------------|-------------|----------------|-------------|-------------|-------------|
| 1   | Weinan (WN) | 3.95        |                | 1.01        | 0.14        | 2.80        |
| 2   | Lu’an (LA)  | 3.66        |                | 2.15        | 0.37        | 1.14        |

2.2. Microwave Energy Absorption Process. The ability of fine coal to absorb energy in the microwave heat pretreatment process was enhanced through the magnetic medium, even though the microwave energy was converted into heat energy. The absorption ability of the microwave treatment sample was mainly dependent upon two conditions. First is the microwave energy’s attenuation performance in the interior of the material and is called the attenuation characteristic. Second is how far the microwave energy penetrates the sample. This second condition means that the sample should have better matching characteristics.

The magnetic parameters of fine coal could be improved effectively through magnetic medium, especially the imaginary part of complex permeability and the attenuation performance of the microwave energy in fine coal.

As shown in Figure 2, the model for the microwave energy absorption mechanism is strengthened by the magnetic medium. The incident microwave energy $E$ gets absorbed into the granular layer of fine coal. The microwave energy decays through the dynamic dielectric polarization relaxation and magnetization relaxation, and $E_1$ amount of energy is reflected. The energy loss of dynamic dielectric polarization relaxation and magnetization relaxation increases through more magnetic medium. The reduction of microwave reflection energy is $E_2$. The energy which gets transferred from the magnetic medium particles to fine coal layers through conduction and radiation processes is $E'_1$. Microwave radiation and heat conduction existed simultaneously in the pretreatment process by microwave energy. Thermal conductivity and utilization of microwave energy by pyrite in the coal had both been improved.

3. DISCUSSION AND ANALYSIS

3.1. Optimization Mechanism of Permeability Response Behavior of Fine Coal. The absorption process of microwave energy in fine coal is a kind of electromagnetic response process. The electromagnetic wave energy was
converted into heat energy so that the microwave energy would be absorbed through dielectric and magnetic losses.

Magnetic medium consisted of magnetite particles and absorbent for the microwave energy of the two losing media. As a wave absorbing medium, the characteristic of magnetic loss is the main property of the magnetic medium. According to different contents of the magnetic media, the corresponding response relationship between different fine coal and magnetic properties was established.

The real part of the complex permeability of fine coal samples \( \mu' \) represents the internal storage capacity and the capacity of the coal sample under an external alternating magnetic field. The magnetic medium contents of fine coal were found to be 1, 3, 5, 8, and 10%, respectively. As shown in Figure 3, for Lu’an fine coal, the real part of the complex permeability \( \mu' \) increases with an increase in the medium’s content. But the overall change is smaller, and the trend is similar to that of the fine coal. The real part of the complex permeability \( \mu' \) decreases with the increase of frequency. The range of change appears to be diminishing. Lu’an coal belongs to sulfur coal, and therefore when the microwave frequency is larger than the range of 2 GHz, the real part of complex permeability \( \mu' \) shows small fluctuation in the amplitude. The increase in range of the real part of complex permeability \( \mu' \) is \((0, 0.05)\) through the magnetic medium, and the increase in amplitude is small. The overall trend remains consistent and shows a trend similar to that of the fine raw coal.

The imaginary part of complex permeability \( \mu'' \) represents the energy loss of the coal sample under the action of an external alternating magnetic field. During the process of dynamic magnetization, the imaginary part \( \mu'' \) and the real part \( \mu' \) change with the frequency of the alternating magnetic field. At the same time, the magnetic dispersion is caused by the magnetization relaxation. As shown in Figure 4, when the magnetic medium is not added to the raw coal, the range of change for the complex permittivity’s imaginary part is \((0, 0.20)\). With the gradual increase of magnetic medium, the imaginary part of the complex dielectric constant also increases.

The imaginary part of complex permeability \( \mu'' \) represents the energy loss of the coal sample under the action of an external alternating magnetic field. The magnetic medium is helpful in improving the whole absorption capacity of the fine coal. As shown in Figure 5, when the magnetic media content is 10%, the imaginary part of the complex dielectric constant is increased by 2.5 times. The resonance peak appears when the added amount is 10%.

3.2. Absorption Characteristics of Fine Coal in Medium Strengthening Process. Figures 6 and 7 show the curves of microwave reflection coefficient against the frequency for the magnetic medium content of 0 and 2%,
respectively. In the test frequency range, only one microwave absorption peak appeared at the thickness values of 0.005, 0.010, and 0.015 m. Two peaks appeared at the thickness values of 0.020, 0.025, and 0.030 m. Three peaks appeared at the thickness values of 0.035 and 0.040 m. According to the odd number principle of microwave wavelength $l(1/4)$, the incident microwave gets divided into two parts. One is the reflection, whereas the other is the one that gets absorbed in the coal sample. If the microwave energy, which has entered into the coal, was not absorbed, then the microwave would get reflected. When the phase difference between the first and second reflection waves is an odd multiple of $1/4$, then the two microwaves cancel each other’s effect and produce an absorption peak at the interface. With the increase in the thickness ($d$) of the Lu’an coal, the maximum attenuation frequency shifts to low frequency and the value of the absorption peak increases. This is mainly due to the interference of fine coal’s thickness to the incident and reflection waves. As shown in Figure 7, the first peak (compared with the low-frequency peak) gradually increases with an increase in the thickness of Lu’an coal. The second peak is much larger than that for the peak value of the low frequency and is consistent with the pattern of change for the relative low-frequency peak value; their values gradually increase with the increase in the thickness of the Lu’an coal;
whereas the third absorption peak (compared with the peak frequency) decreases gradually with the increase in thickness of the Lu’an coal samples. If the magnetic content increases, the number of absorption peaks will remain unchanged. As the coal thickness continues to increase, more absorption peaks will appear.

The absorption of microwave energy is determined by the electromagnetic parameters and thickness of the coal samples. Meanwhile, the central frequency position of the absorption peak is determined by the magnetic content and thickness of coal samples. Thickness is the main influencing factor. Under the same coal thickness, with an increase of magnetic medium content, the peak value is greatly improved. When the thickness was 0.040 m, the peak values of relatively low frequency were −1.79, −2.02, −2.64, and −2.53 dB at the magnetic medium content of 0, 2, 5, and 10%, respectively. The middle peak values of Lu’an coal were −3.94, −4.67, −5.28, and −7.69 dB, respectively. The peak values of relatively high frequency were −4.75, −6.20, −6.88, and −12.28 dB. Overall, with an increase in the magnetic medium’s content, the corresponding peak value increased but at the relatively low-frequency peak area, especially for the magnetic medium content larger than 5%, the peak value decreased and the relatively low frequency was near the commonly used microwave frequency of 2.45 Hz. What deserved more attention was that, when the added amount was 10%, the best peak value appeared in the coal sample having a thickness of 0.035 m and the corresponding value was −16.55 dB in the test range of microwaves.

3.3. Magnetic Medium’s Absorption Enhancement Mechanism. When the thickness of Luan fine coal is 0.010 m, varying the frequency changes the reflection coefficients of different magnetic media contents. Figure 8 shows the analysis of absorption characteristics (d = 0.010 m) of different magnetic media contents of Lu’an coal. The results show that the absorption characteristics of fine coal have greatly been improved for the absorption ability of microwave energy by improving the overall content of magnetic media. As shown in Figure 8, when Luan coal additive contents were 5 and 10%, the absorption peak was increased 2 and 2.5 times of the value when no additive was used. The absorption peak bandwidth has been greatly improved with the increase of the content of magnetic medium in fine coal. It can effectively improve the adaptability of the process design of microwave enhancement and provide effective theoretical support for guiding the selection of pretreatment parameters.

The position of absorption peak center frequency was determined by the magnetic content and thickness of Lu’an coal. Coal thickness effect is the main factor. The center frequency of the absorption peak increases with an increase of the magnetic medium, and there is a certain amount of deviation in the direction of low frequency. The magnetic medium is helpful for the coal sample to lower frequency shift of the absorption peak of the microwave energy and provides support for the design of the strengthening process of the microwave pretreatment.

3.4. Prediction Model for Microwave Electromagnetic Parameter Response. Due to the consideration of operating conditions and coal quality characteristics, the prediction model of influencing factors includes a number of variables, such as microwave frequency, magnetic media content, and coal characteristics, including moisture, ash content, fixed carbon content, volatile, organic sulfur, and pyritic sulfur content. Due to the influence of many factors, the characteristics were nonlinear. It is difficult to establish the relationship between electromagnetic parameters and influencing factors. The forecasting method of the traditional model cannot reach a better forecast analysis. In the electromagnetic parameters of the neural network prediction model of the fine coal bearing layer and the output layer, the transfer function is tanh and the number of neurons in the output layer node prediction model is 1. The maximum number of iterations is 50 000. The prediction accuracy of target is 0.00001, whereas the setting rate of learning and training is 0.01. The forecasting model of the neural network will get the output value of the relative error, the comparison diagram of experimental and predictive values, the variation curves of learning training errors, and the simulation results.

Electromagnetic parameters of the dielectric constant of the real and imaginary parts of fine coal were predicted and analyzed by using back propagation (BP) neural network prediction model. Figure 9 is the neural network model to predict the complex permittivity of the real and imaginary parts of the output of the relative error. The relative error of the real part of the complex dielectric constant is controlled at (−0.01, 0.01) in the training quantity of 400. The training error is in the acceptable range, which indicates that the model is effective (Figure 9a). When the training quantity of the imaginary part of the complex dielectric constant is 1500, the relative error is controlled at (−0.02, 0.02). The training error is in the acceptable range, which indicates that the model is effective. The training effect of the prediction model is better than that of the real part learning and training. The goal of the study is to reach 0.00001, which can meet the requirements of learning and training. The BP neural network model of the real part of the complex permittivity appears after 681 training. An error of 0.00012 is reached, which is different from the desired value of 0.00001. However, the obtained errors are small enough. The forecast error of the network model is relatively small, which can meet the requirements of learning and training. The relative error appeared to have a larger value due to the number of samples of learning training.

After the neural network is well trained, the feasibility of neural network prediction and analysis system is needed. The predicted values obtained from test samples and neural...
network were compared and analyzed. In Figure 10, the symbol “o” and the symbol “+” coincide together, which shows the feasibility of the neural network system. The prediction effect of the real part of the complex dielectric constant is better than that of the imaginary part of the complex permittivity.

Figure 11a shows that the real part of the complex dielectric constant having a value of $R = 1$ can explain the fact that the training process has a high significance and that the training results are consistent with the true value. For test and analysis, as shown in Figure 11b, the analysis results show that the predicted target value is consistent with the output value, which shows that the prediction effect of the model can meet the requirements. Figure 11b shows that the virtual part of the complex dielectric constant learning and training has a value of $R = 0.9981$, which shows that the training process has a high significance. This also shows that the training results are consistent with the true value and can meet the requirements of the model. The results of the comprehensive analysis at $R = 0.9982$ show that the predicted target value and the output value are basically the same.

3.5. Magnetic Separation Desulfurization of Fine Coal. Methods to strengthen the magnetic desulfurization by microwave medium can be effective for desulfurization of high-sulfur coal. In the present study, the effect of magnetic medium’s content on the microwave medium is to strengthen the desulfurization effect. There are more preferred forms of sulfur iron sulfide coal.

Values of the factors considered in the analysis were found as follows: coal sample size of $-0.50$ mm, magnetic particle size of $0.074-0.150$ mm, and microwave pretreatment atmosphere is in the air. Lu’an coal (LA) and Weinan coal (WN) were used in the magnetic separator having microwave exposure time differing from each other by 180 s. Figure 12 shows the results for rare earth magnetic roller separator used with different contents of medium high-sulfur fine coal. Two kinds of high-sulfur coal were used. However, the reduced amplitude decreases with an increase of the media content, especially, for more than 5% content, a slight increase is observed. The yield of clean coal content in magnetic medium of 0–10% increases with the increase of media content. However, in a smaller range of 2–5%, the yield reduced. When the content was more than 2%, the coal yield declined sharply.
As shown in Table 2, rare earth magnetic roller separator used with different contents of medium high sulfur fine coal shows a good desulfurization index when magnetic media content is 5%. The yields of clean coal from Weinan and Lu’an coals were 41 and 38.50%, respectively. The sulfur content in clean coal was found to have values of 3.56 and 2.41% for Weinan and Lu’an coals, respectively. The desulfurization rate was found to be 61.70 and 38.50% for Weinan and Lu’an coals, respectively. Clean coal yield is very low. When the magnetic medium content is 2%, Weinan and Luan coal yielded 92.11 and 88.2% clean coal, respectively. The corresponding values for sulfur content in the clean coal were 3.26 and 2.25%, respectively. The desulfurization rate was found to have values of 23.99 and 45.78%, respectively. Comprehensive analysis of the desulfurization indicates that for both Weinan and Lu’an coals, the overall separation index is the best for a medium content of 2%.

4. CONCLUSIONS

1. The strengthening mechanism of dielectric storage energy, dielectric energy consumption, magnetization energy storage, and consumption of fine coal layers with different medium contents were investigated. The content of magnetic medium was varied by increasing the electromagnetic parameters to improve the micro-wave absorption properties and effectively improve the energy utilization rate and reduce the energy losses. The microwave pretreatment process can effectively improve the absorption and broaden the bandwidth of the coal sample. The process can also improve the frequency adaptive design of coal. The content of magnetic medium is conducive to the absorption of the microwave energy in low-frequency region.

2. On the basis of the theoretical analysis, a physical model for the absorption of microwave energy is proposed. The microwave energy absorption law of fine coal during the process of strengthening of the microwave energy absorption is revealed. Magnetic medium has been used to improve the dynamic dielectric polarization...
relaxation and the energy losses of the magnetic relaxation, whereas the absorption of microwave energy by the magnetic particles through heat conduction and radiation to the fine coal layer transfer energy has also been investigated.

3. Microwave processing of fine coal absorptive capacity mainly depends on two conditions: microwave energy penetrating the material which depends upon the attenuation performance or the attenuation characteristics, followed by microwave energy getting absorbed into the sample surface to maximize access to the sample, that is, the absorption of samples with better matching characteristics. A neural network model for the prediction of the electromagnetic parameters of fine coal is established by using the neural network method. The real and imaginary parts of complex permittivity and complex permeability are trained and predicted. The neural network model has shown satisfactory results.

4. When the magnetic medium content was 2% for Weinan and Lu’an coals, values for the clean coal yield were 92.11 and 88.2%, respectively. The corresponding values for sulfur content in clean coal were 3.26 and 2.25% for Weinan and Lu’an coals, respectively. The desulfurization rates were 23.99 and 45.78% for Weinan and Lu’an coals, respectively. A comprehensive analysis shows that for separation desulfurization of Weinan and Lu’an coals, the separation index appears to have the best values for a medium content of 2%.

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