

A Step-by-step Classification Method of Coal and Miscellaneous Materials by Laser-induced Breakdown Spectroscopy

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ABSTRACT: The miscellaneous materials, including gangue, plastic, and wood commonly present in coal. These miscellaneous materials affect the reliability of coal analysis using laser-induced breakdown spectroscopy in power plants, but have significantly distinct spectral characteristics from coal. Hence, this paper proposes a step-by-step classification method to screening the false spectra of miscellaneous materials. The first step aims to identify the plastic and wood spectra by determining the existence of specific characteristic spectral lines using the standard deviation (SD) values. The spectral lines Si I 288.16 nm with the SD value of more than 850 counts and Li I 670.78nm with SD value of more than 1750 counts were used as the distinguishing markers. The classification accuracy of first step was 100%. Due to the high similarity between gangue and coal, the second step utilized the random forest (RF) classification model to identify the gangue spectra. The number of trees and random variables in the RF model was optimized. The accuracy of classification model without and with the proposed step-by-step method was 98.30 and 99.96%, respectively. To assess the necessity of spectra classification, a set of calorific value analysis was performed by adding false spectra of different proportions, which were compared with analysis after removing the false spectra. The root mean square error of prediction (*RMSEP*) was 0.42 MJ kg⁻¹ (after removing), compared with 0.50 MJ kg⁻¹ (mixing with 10% gangue spectra), 0.56 MJ kg⁻¹ (mixing with 20% gangue spectra) and 0.57 MJ kg⁻¹ (mixing with 30% gangue spectra). The results demonstrated that the proposed step-by-step classification method could effectively identify the spectra of coal and miscellaneous materials and improve the accuracy of coal analysis.

INTRODUCTION

Recently, China has set the goal of reaching carbon peaking and carbon neutrality, making the transition to a new energy structure imperative. In this context, the flexibility of thermal power generation and its ability to regulate peak shaving will become more prominent. The ultimate and proximate analysis of coal is crucial to support the data needs of power plants in optimizing combustion. Knowing the coal property prior to utilization is helpful for flexible peak shaving. Commercial analytical devices based on prompt gamma neutron activation analysis (PGNAA)

and X-ray fluorescence (XRF) technology already exist. However, PGNAA requires bulky equipment and must be operated under strict safety regulations. XRF cannot measure low atomic number elements which are essential measurements.¹ Laser-induced breakdown spectroscopy (LIBS) is a type of frontier atomic emission spectroscopy that has been extensively applied in the ultimate and proximate analysis of coal. LIBS has advantages of quick, simultaneous multi-element, and online detection. Measurement equipment based on LIBS can be installed on the coal conveyor belt to provide fast measurement and timely data,²⁻⁸ supporting the need for flexible peak shaving of power plants.⁹

However, LIBS spectra are highly sensitive to the analyzed sample, and the spectra of different samples can vary significantly. During coal mining, transportation, and storage, various miscellaneous materials, such as gangue, plastic, and wood can mix with the coal, which can affect the accuracy of coal analysis using LIBS. It is essential to screen the miscellaneous materials on the conveyor belts in the power plants. Visual recognition combined with machine learning has been successfully used to classify coal and miscellaneous materials, with effective outcomes reported in previous studies.¹⁰⁻¹³ However, this method requires the introduction of various devices, which can adversely affect the convenience and efficiency of LIBS for coal analysis in power plants.

LIBS combining various classification methods has also been applied to screen the substances.¹⁴ Especially the machine learning algorithms show better performance for samples with similar compositions, such as coal and gangue. Lin *et al.*¹⁵ tested four classification models, including Naïve Bayes, K-nearest neighbor (KNN), decision tree (DT), and random forest (RF) combined with LIBS to differentiate metal additive manufacturing (AM) parts, and results showed that the RF model performed best. Sheng *et al.*¹⁶ compared the performance of support vector machines (SVM) and RF in classifying iron ores and concluded that RF exhibited a better level of classification prediction. Janovszky *et al.*¹⁷ tested three statistical methods, including classification tree (CT), linear discriminant analysis (LDA), and RF, for classifying mineral particles and found that the accuracy of both RF and LDA was better than 92%. Rzecki *et al.*¹⁸ used seven classifiers, including DT, RF, KNN, SVM, probabilistic neural network (PNN), multilayer perceptron (MLP), and generalized regression neural network (GRNN), to classify paper and ink samples. The results showed that the RF classifier had the highest recognition accuracy. The above studies suggest that LIBS combined with RF is a highly effective classification method. Additionally, our group developed a method to check whether the characteristic spectral lines were reliably present that used its standard deviation (SD) values. Bai *et al.*¹⁹ chose Si 288.16 nm as the analysis line and used its SD value to identify invalid breakdown spectra from all the particle stream spectra. Similarly, Yao *et al.*²⁰ calculated the SD value of C 247.86 nm to judge whether the coal is effectively excited. This method can be effectively applied as a basis for classification when the elemental composition of different samples differs significantly.

In summary, the presence of false spectra generated by miscellaneous materials can affect the accuracy of coal analysis using LIBS. It is necessary to screen the false spectra, which has not yet been investigated. This work proposes a step-by-step classification method based on the characteristic spectral lines and the RF model for coal and false spectra. The SD values of specific spectral lines were used to screen the spectra of plastic and wood. Then, the RF model was adopted to screen the spectra of gangue. To assess the efficacy and necessity of this classification method,

Fig. 1 LIBS spectra comparison of coal, gangue, plastic and wood.

Fig. 2 Step-by-step classification method for coal and miscellaneous materials spectra.

a set of calorific value analysis was performed by retaining false spectra of different proportions.

METHODS

Step-by-step classification method. Fig. 1 compares typical

Fig. 3 Characteristic spectral lines comparison of (a) Si I 288.16 nm and (b) Li I 670.78nm, the dashed line is the fitted line when the SD value of the specific spectral line is the set threshold value.

LIBS spectra of coal, gangue, plastic, and wood samples obtained by averaging 300 spectra collected from each sample. The figure highlights several characteristic spectral lines of major elements that may be used to distinguish the spectra. The elemental composition of plastic and wood differs significantly from that of coal, making their spectra, especially for some characteristic spectral lines, entirely different. Therefore, the presence of several specific spectral lines can be used to screen the plastic and wood spectra. In contrast, the element composition in coal and gangue is highly similar. The differences in elemental content are reflected in the intensity of spectral lines, such as the 350~450 nm band in Fig. 1. However, the fluctuation of the LIBS spectrum makes the spectral intensity highly uncertain, and it is difficult to classify the coal and gangue spectra by comparing the SD values of specific spectral lines.

Therefore, a step-by-step classification method for coal and miscellaneous materials is proposed, which includes three steps. First, the classification model based on the characteristic spectral lines is applied to screen the plastic and wood spectra. Second, the spectra of coal and gangue are preprocessed by reducing the data dimensionality. Finally, the random forest model is used to identify the spectra of coal and gangue. Fig. 2 illustrates the scheme, and the following sections will discuss the basic principles and descriptions of these three steps in detail.

Characteristic spectral lines classification model. In this study, plastic and wood sample spectra were screened by checking the existence of specific spectral lines using their SD values. SD can be calculated as:

$$SD = \sqrt{\sum_{i=1}^N (X_i - \bar{X})^2 / (N - 1)} \quad (1)$$

where SD is the standard deviation of the spectral line intensity, X_i is the intensity of the i th pixel in the peak of spectral line, \bar{X} is the average intensity of all the pixels in the peak of spectral line.

N is the pixel number in the peak of spectral line. The value of N is set to 5 in this work.

After screening, all spectra would be classified into two kinds: coal and gangue, and plastic and wood. The selection of the analyzed spectral lines was based on the principle that their intensity and corresponding the content of elements were significantly different in two kinds of samples. Two ways were used to choose the analyzed spectral line: selecting the main elements of each sample or selecting trace elements with significant differences in the samples. While the main elements for all samples were evident, including C for coal, Al and Ca for gangue, C for plastic, and O for wood. They were also found in other samples, often in substantial amounts. As shown in Fig. 1, the high intensities of characteristic spectral line of these primary elements were common in all spectra with minor differences, rendering them unsuitable for the analyzed spectral lines. Thus, trace elements that only existed in coal and gangue, such as Si and Li, were selected for analysis.²¹

As shown in Fig. 3, the characteristic spectral lines of Si I 288.16 nm and Li I 670.78 nm were significantly different between the spectra of the two kinds of samples, and they would be used as analyzed spectral lines for the classification model.²² The SD values of these spectral lines were calculated one by one and used to identify plastic and wood spectra. The minimum SD values of coal and gangue were larger than the maximum SD values of plastic and wood. Thus, the SD thresholds of these spectral lines were determined and used to completely distinguish the plastic and wood from coal and gangue. The SD thresholds of the intensities of -Si I 288.16 nm and Li I 670.78 nm -are 850 and 1750 counts, respectively. Fitted lines with SD values set as the thresholds were included in Fig. 3 to demonstrate how the classification model works. As mentioned before, the peak of spectral line consisted of five pixels in this work. It was assumed that the spectral intensities of the first- and fifth-pixel points were equal to the intensities of wood spectra's corresponding points. The shape of the characteristic peaks was composed of two axisymmetric diagonal lines, and the SD values of the characteristic peaks were used as thresholds to draw the fitted lines. The specific spectral lines in coal and gangue spectra were more prominent and distinct than the fitted line, resulting in their SD values being greater than the thresholds. Conversely, the plastic and wood spectra exhibited opposite results.

Data dimensionality reduction. The high-dimensional nature of LIBS spectra results in a large amount of redundant information, which translates to a long computation time when input into the model. To address this issue, feature selection and principal component analysis (PCA) were used to reduce the dimensionality of LIBS spectra and extract valid information when establishing the RF model for coal and gangue classification. Since coal and gangue constituent elements are remarkably similar, this work concentrated on several elements with notable differences in

Table 1. Selected feature for the RF model

Selected Feature	Wavelength/nm
C I	247.86
Si I	251.61 and 288.16
Fe I	275.61 and 373.49
Al I	308.22, 309.28, 394.40 and 396.15
O I	777.19

Fig. 4 The scatter plot of the SD values of (a) C I 247.86 nm and Ca II 393.37 nm, (b) Al 394.40 nm and O 777.19 nm.

content. Several characteristic spectral lines whose intensities varied considerably in coal and gangue were selected for analysis through the manual line selection (MLS) method based on the NIST database,²² as shown in Table 1. The cumulative contribution of eigenvalues after PCA was greater than 99%.

Random forest classification model. Random forest is an integrated learning method composed of a series of classifiers, or ‘trees’, which work together to make decisions and achieve an optimal result. Randomness is observed in the selection of each tree’s training set using the Bagging method, and a certain number of features are randomly chosen from the attributes. The random forest model is popular in LIBS spectrum classification modeling due to its ability to prevent overfitting, resistance to noise, and capacity for parallel processing.¹⁶

In this study, the RF model was applied to classify the spectra of coal and gangue after data preprocessing, with 5100 spectra in the calibration set and 2100 spectra in the validation set. The number of trees and random variables were optimized to maximize the classification accuracy.

The classification model performance was assessed by the accuracy, which can be defined as follows:

$$accuracy = N_c / N_t \quad (2)$$

where N_c and N_t are the number of correct classified spectra and the total number of spectra in the validation set, respectively.

EXPERIMENTAL

Samples. To simulate the proportion of raw coal blended with miscellaneous materials in the power plant, 27 samples were

collected from the power plant site. Following the procedure of ASTM standard methods,²³ the collected coal and miscellaneous materials were ground into powders at 0.2 mm particle size and dried at 45 °C for 4 h to minimize the influence of moisture on the spectral measurements. Then the powder of 3 g was compressed into compact and flat pellets with a diameter of 24 mm using a tablet machine (Pike Technologies Crushir) to reduce the impacts of physical matrix effects. In particular, 19 coal samples were numbered C1 to C19, 5 gangue samples were numbered G1 to G5, 2 plastic samples were numbered N1 and N2, and 1 wood sample were numbered N3. 14 coal samples, 3 gangue samples, and 1 plastic sample were randomly selected to form the calibration set, while the remaining 5 coal samples, 2 gangue samples, 1 plastic sample, and 1 wood sample formed the validation set. Table 2 provides calorific values ranging from 18.23 to 25.92 MJ kg⁻¹ of 19 coal samples on air dried basis, which were analyzed by the ASTM standard method.²³ The chemical composition of gangue is similar to that of coal, with a slightly lower carbon content and a significantly higher concentration of metallic elements than coal.^{24, 25} In contrast, plastics and wood differ significantly from coal in elemental composition. The collected plastic samples are mostly made of polypropylene (PP) and polyethylene (PE), which mainly composed of C and Al.²⁶ The wood primarily contains O, Ca, K, P, and S.²⁷

LIBS experimental setup. The LIBS experimental system’s description has been provided in detail in the literature.²⁸ The ablation source was a Q-switched Nd:YAG laser (Quantel Brilliant Easy, USA) emitting at 1064 nm with the pulse duration of 5 ns. The repetition frequency was set to 2 Hz and the laser energy was adjusted to 55 mJ/pulse to obtain a better spectrum. A lens with a focal length of 150 mm was employed to concentrate the laser onto the sample surface to produce a plasma. Then the four-channel spectrometer (Avantes, Netherlands), which had a nominal resolution of 0.08-0.10 nm and spanned the wavelength range of 178 to 827 nm, received the plasma emission by an optical fiber. The integration time was set to 1.05 ms and the delay time was adjusted to 1 μs. To simulate the collection condition of LIBS performing on the coal conveyor belt, the samples were placed on a motorized translation table. The table was controlled to move the samples in the XY plane at a speed of 3 mm/s. 1 spectrum was taken at each of the 300 locations on the sample surface for each sample.

RESULTS AND DISCUSSION

Characteristic spectral lines classification model. Fig. 4 displays a scatter plot of all samples’ SD values of line intensity of C, Ca, Al, and O. These four elements were chosen by using the strategy of selecting the main elements of each sample. Fig. 4(a) shows that the SD value distribution of C and Ca spectral lines was similar for all spectra, making it difficult to classify the

Table 2. Calorific value of samples in this study

Calibration set		Validation set			
Sample	Calorific value (ad)/MJ kg ⁻¹	Sample	Calorific value (ad)/MJ kg ⁻¹	Sample	Calorific value (ad)/MJ kg ⁻¹
C1	21.66	C10	18.23	C15	23.01
C2	23.72	C11	23.45	C16	23.05
C3	25.03	C12	23.01	C17	24.48
C4	23.18	C13	18.38	C18	23.85
C5	24.57	C14	22.70	C19	23.50
C6	25.92	G1	-	G4	-
C7	25.60	G2	-	G5	-
C8	23.97	G3	-	N2	-
C9	24.39	N1	-	N3	-

Fig. 5 The scatter plot of the SD values of Si I 288.16 nm and Li I 670.78 nm, and their thresholds have been marked with red lines.**Table 3.** Comparison of the classification accuracy using different characteristic spectral lines

Characteristic spectral lines	Accuracy/%
Si I 288.16 nm	82.04
Li I 670.78 nm	99.70
Si I 288.16 nm and Li I 670.78 nm	100.00

Table 4. Comparison of the RF classification accuracy before and after data dimensionality reducing

Preprocessing methods	Coal spectra/%	Gangue spectra/%	All spectra/%
Raw spectra	99.87	94.17	98.24
Feature selection and PCA	99.87	99.67	99.81

spectra by setting a threshold. Fig. 4(b) indicates that the SD values of the O and Al spectral lines were mixed together to some extent. Relying solely on these two spectral lines made it difficult to completely distinguish two kinds of samples.

Figure 5 is a scatter plot of the SD values of Si I 288.16 nm and

Li I 670.78 nm. It shows that the plastic and wood were distinguished from coal and gangue spectra. Orange points represents the plastic and wood spectra that are close to the origin of the axis, while grey points indicates that coal and gangue are farther from it. Obviously, the SD values of these spectral lines are an effective way to separate spectra of different samples. As previously mentioned, the SD values of two characteristic spectral lines, Si I 288.16 nm and Li I 670.78 nm, were used as the basis for classification in this model. Table 3 compares the accuracy of the classification model using these criteria separately and in combination. The accuracy of classification model using the spectral line of Si I 288.16 nm and Li I 670.78 nm was 96.26% and 99.70%, respectively. However, the accuracy of the model using the combination of the spectral lines of Si I 288.16 nm and Li I 670.78 nm reached 100%, with all plastic and wood spectra being screened out.

Random forest classification model. After successfully eliminating all plastic and wood spectra, an RF classification model was built to classify coal and gangue using the remaining 4200 spectra of coal and 900 spectra of gangue in the calibration set as input. The model's performance was checked by inputting the remaining 1500 coal and 600 gangue spectra from the validation set. In addition, data preprocessing methods of feature selection combined with PCA were applied to reduce the dimensionality of the raw spectral data and extract the valid information. Table 4 compares the classification accuracy of RF models without and with data dimensionality reduction. The number of trees (n_{tree}) and random variables (m_{try}) used in the RF model were empirically set to 200 and 3, respectively. The results demonstrated that the raw spectra combined with the RF model already provided a high classification level. However, the model calculation was time-consuming and unsuitable for LIBS's fast and online measurements. It was necessary to reduce the raw data dimensionality. The feature selection combined with PCA was an effective method, obtaining a classification accuracy of 99.81%. Therefore, in the subsequent RF model optimization, the feature selection combined with PCA was applied throughout the process.

To further optimize the model, the classification accuracy of the RF model was contrasted as a function of n_{tree} at different levels of m_{try} , as shown in Fig. 6(a). The model achieved the highest

Fig. 6 Relationship of (a) step-by-step and (b) non-step-by-step classification accuracy with n_{tree} and m_{try} .

Fig. 7 The results of the calorific value analysis using 4 inputs: (a) experimental group, (b) control group A, (c) control group B, (d) control group C.

accuracy of 99.95% when m_{try} was set to 2 and n_{tree} was set to 380, 480, and 500, with only one spectrum in the validation set being incorrectly classified. The ideal value of n_{tree} was 380 to accelerate the model calculation. Combining the characteristic spectral lines and RF classification model, the step-by-step classification

method achieved an accuracy of 99.96%.

Figure 6(b) illustrates the classification results without employing the step-by-step classification method. Here, all preprocessed spectra of miscellaneous materials and coal were

used as input to construct the classification model. The validation set contained 9 samples with 2700 spectra. The classification accuracy was 98.30% with 46 spectra misclassified. This may be due to the large spectral differences and the preprocessing methods reducing the information contained in the spectra. In comparison, the model without employing the step-by-step classification method was slower to calculate and had a lower accuracy. These results demonstrated that the proposed step-by-step classification method was effective.

Calorific value quantitative analysis. To assess the necessity of spectral classification in coal analysis, a comparative experiment was performed for calorific value analysis. This was done by adding false spectra generated by gangue with varying proportions and comparing them with classified spectra. The raw coal production process typically yields 15% to 30% associated gangue production. Based on the degree of spectral interference, three control groups were formed. Control group A had 300 spectra of each sample with a random substitution of 10% gangue spectra for coal spectra, resulting in the ratio of coal and gangue spectra was 9:1. The ratios of coal and gangue spectra in control groups B and C were 8:2 and 7:3, respectively. The blended gangue spectra were randomly selected from all spectra of gangue samples. The blending ratio of each gangue sample was the same, and all spectra did not repeat each other. The spectra of plastic and wood were effectively excluded, so their effect was not considered here.

In the practical application of LIBS to coal analysis in power plants, the quantitative model for calorific value is typically built from laboratory data. Then the data collected in the field is fed into the model to predict the calorific value. In this work, the LIBS data from coal samples of the calibration set was used as the independent variable to build the quantitative analysis model, and the dependent variable was the calorific value. Because of the complicated and intimate relationship between calorific value and the content of various elements in coal, the full LIBS spectra were utilized for quantitative analysis.²⁹ A quantitative analysis model for calorific value was built based on partial least square (PLS) to minimize the spectral dimensionality. When predicting elemental content in LIBS, PLS is a regression analysis method that contains correlation analysis and was frequently used in LIBS predicting elemental content.^{20,30} Three averaged spectra were input for each sample while modeling the spectra of the calibration set coals to reduce the influence of data fluctuations on the calorific value prediction.^{31,32} For the quantitative model of control groups, the spectra of each coal sample and the blended gangue spectra were first divided into three equally. Then the spectra were averaged, which means the blending ratio of the gangue spectra of 3 averaged spectra was the same.

The performance of quantitative analysis model of the calorific value was assessed by the determination coefficient (R^2), the root mean square error of prediction ($RMSEP$), the average absolute error (AAE), and the average relative error (ARE), which has been

introduced detailed in literature.¹⁵

Figure 7(a)-(d) compares the prediction results of calorific value using spectral data from the experimental group, control group A, control group B, and control group C, respectively. As can be seen, the R^2 remained unchanged at 0.99, the $RMSEP$ increased from 0.42 MJ kg⁻¹ (after removing) to 0.50 MJ kg⁻¹ (mixing with 10% gangue spectra), 0.56 MJ kg⁻¹ (mixing with 20% gangue spectra), and 0.57 MJ kg⁻¹ (mixing with 30% gangue spectra), the AAE increased from 0.42 MJ kg⁻¹ (after removing) to 0.50 MJ kg⁻¹ (mixing with 10% gangue spectra), 0.54 MJ kg⁻¹ (mixing with 20% gangue spectra), and 0.54 MJ kg⁻¹ (mixing with 30% gangue spectra), and the ARE increased from 1.80% (classified spectra) to 2.14% (mixing with 10% gangue spectra), 2.35% (mixing with 20% gangue spectra), and 2.35% (mixing with 30% gangue spectra).

Since the model was built from the same calibration set, R^2 remained unchanged at 0.99. However, the value of $RMSEP$, AAE , and ARE increased with the proportion of doping in the gangue spectra. This is because the LIBS spectra of both coal and gangue contains emission lines such as C, H, and O and the mineral elements Si, Al, and Ca, but the content of which differs significantly. The maceral and mineral makeup of the coal affects the calorific value as a coal-rank metric. In actuality, the calorific value drops as the mineral matter rises due to the endothermic breakdown processes and the heat capacities of such minerals during burning.³³ Still, it increases as the carbon content does.³⁴ Therefore, the kinds of samples profoundly affected LIBS spectra and the calorific value prediction. The results demonstrated that the proposed step-by-step classification method was effective in eliminating the interference of the miscellaneous materials spectra and improving the accuracy of calorific value prediction.

Furthermore, the conventional method of coal analysis utilized in power plants requires sampling from the conveyor belt, sample preparation, and analysis employing a calorimeter in the laboratory. In contrast, the step-by-step classification method proposed in this study requires less than 5 s to classify the entire spectra. When integrated with the LIBS measurement device installed on the coal conveyor belt, the entire process from spectrum acquisition and screening to coal analysis can be completed within an average of 1 minute. Therefore, the proposed classification method in conjunction with LIBS measurement resolves the issues of time-intensive analysis and poor sample representativeness associated with conventional methods to satisfy the substantial demand for flexible peak-shaving in power plants.

CONCLUSION

A step-by-step classification method to effectively screen the miscellaneous materials spectra was proposed to improve power

plants' coal analysis on the conveyor belt. The spectra of plastic and wood were screened using the SD values of specific characteristic spectral lines, and the classification accuracy reached 100%. Subsequently, after reducing the data dimensionality using feature selection combined with PCA, an RF classification model was used to classify the gangue and coal spectra. The accuracy was 99.95%, with only one gangue spectrum misclassified by optimizing the number of trees and random variables. Finally, PLS was employed as the calibration model to compare the predicted performance of calorific value using the spectra without and with classification as input. Compared to spectra mixed with coal and gangue, the classified spectra resulted in optimal *RMSEP*, *AAE*, and *ARE* values, which were 0.42 MJ kg⁻¹, 0.42 MJ kg⁻¹, and 1.8%, respectively. The results confirmed the need to identify coal and miscellaneous materials' spectra and provided an experimental basis for practical application. The proposed step-by-step spectral classification method can further improve the accuracy of coal analysis and support the flexible peak-shaving in power plants.

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Notes

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