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Article

An Optimization Method Based on Spatial Confinement and Spectra Data Screening for Laser-induced Breakdown Spectroscopy Quantitative Analysis of Coal Particle Flow

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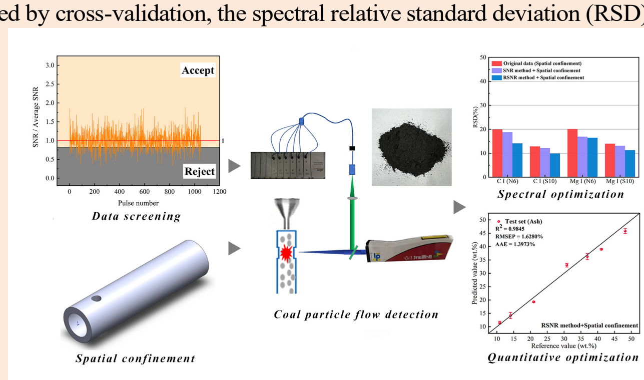
ABSTRACT: Laser-induced breakdown spectroscopy (LIBS) detection on coal in particle flow form is more real-time and beneficial for the effective use of coal. However, the inevitable instability fluctuation of particle flow affects the laser ablation and spectral collection. This work attempted to use particle flow confinement schemes and spectral screening to optimize the quantitative analysis performance of coal particle flow. A cylindrical spatial confinement was developed to stabilize the coal particle flow, laser ablation and plasma evolution. Then a relative signal-to-noise ratio (RSNR) method was proposed for spectral data screening to overcome the inadaptability of traditional signal-to-noise ratio (SNR) method to the samples with large difference in SNR. Based on the RSNR method with the optimal screening threshold obtained by cross-validation, the spectral relative standard deviation (RSD) such as C I 247.86 nm and Mg I 285.21 nm were significantly reduced. Subsequently, quantitative analysis models for coal proximate analysis indexes were established. The models employing RSNR method and spatial confinement showed the superior performance. The root mean square error of prediction (RMSEP) of ash content, fixed carbon content, volatile matter content and calorific value were improved from 3.5667%, 3.3965%, 3.0905% and 1.0188MJ/Kg to 1.6280%, 2.0344%, 1.4999% and 0.4553MJ/Kg. The proposed spatial confinement and RSNR method have applicability and mutual gain for quantitative analysis of coal particle flow.



INTRODUCTION

Coal is one of the most important primary energy sources, and the rational use of coal is related to energy supply and environmental health.[1] The diversity and variability of coal quality make it conditional for its stable and effective application.[2,3] In many scenarios, it is necessary to obtain various coal quality characteristic index quickly. Laser-induced breakdown spectroscopy (LIBS) is considered as a potential leading technology for online coal detection due to its in-situ, rapidly and other technical advantages. It can realize the detection on the indexes of elemental analysis and proximate analysis.[4-7] LIBS has been applied to coal detection for many years, and different detection schemes and optimization methods have been proposed according to different needs for different forms of coal including rock mineral,[8-10] pellet[11-13] and particle flow.[14-19] The coal in the particle flow state is the final form of coal before it enters the boiler and other heat conversion equipment. The detection on particle flow can provide valuable guidance for the optimization of conversion, which is the most real-time and reliable.

At present, relevant researches have been carried out on the LIBS detection of coal particle flow. Zheng *et al.*[14] studied the spectral characteristics of laser induced coal particle flow in air environment, and realized the quantitative analysis of fixed carbon

(RMSE =2.40%). Yu *et al.*[15] compared the effect of matrix effect on coal particle flow and pellet detection. The results indicated that the spectral fluctuation induced by the matrix was less pronounced in particle flow due to the milder effect of laser thermal conduction variation. Up to now, multiple systematic research work about spectral data preprocessing,[16] quantitative analysis methods,[17] and detecting system design[18,19] have carried out and built and innovated several prototype LIBS device for the online scenario detection of particle flow.

However, the LIBS detection on particle flow is still suffering from the negative impact of the inevitable fluctuation of particle flow, the ablation on the particle flow does not always guarantee a high ablation rate and a very fixed plasma location. Furthermore, the fluctuation of coal particle flow is also affected by the supply, particle heterogeneity, and environmental disturbances. These factors result in uncertainty in the generation and collection of LIBS spectral signals.

To address these limitations, researchers have primarily attempted to resolve the issue through data screening or optimization schemes for detection. Data screening involves assessing the quality and representativeness of the obtained spectrum using discriminant methods and then determining whether to retain or exclude it. Hahn et al.[20] proposed a method employed the appropriate peak-to-base ratio (P/B) value and signal-to-noise ratio (SNR) value as screening indicators for spectral effectiveness identification on silica particle flow analysis. Alvarez-trujillo *et al.*[21] investigated the applicability of the standard deviation (SD) identification method in aerosol system and the result exhibited a positive impact on quantitative analysis. Lithgow *et al.*[22] applied the peak-to-base ratio (P/B) value to determine whether Mg was detected within the plasma for each laser shot on magnesium chloride aerosols. Zheng *et al.*[14] employed background value and its standard deviation to set a threshold for spectral validity discrimination of coal particle flow. Yao *et al.*[23] proposed a spectra data screening method based on the normal curve distribution of carbon emission intensity. The relative error of prediction (REP) of the ash content prediction set was reduced from 6. 35% to 6. 25%. Chen *et al.*[24] developed an image information-assisted screening method for spectral data identification of coal particle flow, which evaluated the laser ablation based on camera image of plasma. The quantitative models for volatile matter content, calorific value and ash content achieved an RMSE value of 0.2293%, 0.3713MJ/Kg and 1.2202%.

Spectra is an intuitive expression of ablation effectiveness. But the above data screening methods that only refer to the spectral information are easily affected by the receiving instrument, optical setting and sample characteristics. Moreover，low-quality source database is likely to render data screening methods completely unworkable.

The optimization of detection schemes mainly involves stabilizing the flow, excitation, or detection process of particle flow through physical auxiliary means. These methods improve the representativeness and reliability of the original spectral information essentially. In our previous work, Yao *et al.*[25] optimized the selection of the outlet diameter of tapered tube for particle flow generation. The results show that the tube with a diameter of 5.5 mm was suitable to enrich the coal particles for beam-focusing and generate effective spectra effectively. Yu *et al.*[26] installed a metallic target in the particle flow to stabilize and enhance the plasma, thereby improving the spectral relative standard deviation (RSD) and SNR from 116% and 9.89 to 48% and 18.63, respectively. They further optimized the focusing system by using dual lenses, and successfully decreased the signal RSD from 15.85% to 8.24%.[27] Recently, we developed a cylindrical spatial confinement and verified its optimization effect on spectral quality of quartz sand particle flow.[28]

In this work, we further explore the application of cylindrical spatial confinement in quantitative analysis of coal particle flow. And propose a new data screening method that is more suitable for the spectral characteristics of particle flow. In addition, the role and coupling effect of spatial confinement and the proposed data screening method were also investigated.

EXPERIMENTAL

LIBS setup. As shown in Fig. 1, the experimental system of this study includes pulverized coal particle flow setup, cylindrical spatial confinement and LIBS setup. The experimental setup is the same as ref 28. The piezoelectric vibrating feeder (PEF-90A, Sanki, Japan) combined with a tapered tube produces a particle flow with a diameter of 4 mm. A speed control module controls the vibration frequency of the vibrating screen to maintain the average mass flow rate at 0.8 ± 0.05 g/min. The laser source is generated by a Nd:YAG pulsed laser (Brilliant Easy, Quantel, France), which operating wavelength is 1064 nm and the pulse width is 8 ns. The laser pulse frequency is set to 3Hz to reduce mutual interference between pulses while maintaining efficiency, and the laser energy is set to 90 mJ after pre-experimental optimization. A focusing lens with a diameter of 25.4 mm and a focal length of 100 mm is employed to focus the laser pulse to the center of the particle flow and generate plasma. The plasma emission is reflected by a 45° long-pass dichroscope (reflection band 200-785 mm, transmission band 825-1300 nm) and then focused to a fiber optic probe. The emission collection device is a six-channel optical fiber spectrometer (Netherlands Avantes, AvaSpec-2048), which covers a spectral range of 178-827 nm with a nominal resolution of about 0.07 nm. The delay time from the laser output trigger signal to spectrometer scanning and the gate width are set to 1 μs and 1.05 ms, respectively. The cylindrical spatial confinement is a hollow quartz tube with an outer diameter

Fig. 1 Schematic diagram of the experimental setup.

Fig. 2 SNR of C I 247.86 nm emission of each pulse from coal N6 and coal S10 with or without spatial confinement.

of 6 mm and an inner diameter of 4 mm, two slits were symmetrically created on the pipe wall to avoid the effect on laser incidence and propagation while realize spatial confinement.

Specimen preparation. Thirty-nine coal samples from the power plants were employed as the experimental specimens. The proximate analysis data of coal samples are shown in Table S1. Before the experiment, the pulverized coal particles are all ground and screened with a 200 μm diameter screen, and dried in a drying oven at 45°C for 4 h to form air-dried basis. The particle flow of each coal specimen was continuously ablated 1050 times by laser under the condition with or without spatial confinement. In the process of establishing the model for quantitative analysis of coal, 7 coal specimens were randomly selected as prediction set (N1-N7), and the remaining 32 coal specimens were employed as training set (S1-S32).

RESULTS AND DISSCUSSION

Spectral data screening and spatial confinement. Coal particle flow is a kind of gas-solid two-phase flow. The material flow is affected by the supply, environment, equipment vibration, etc., and the flow is inherently random, so the laser ablation process is uncertain. This is the disadvantage of coal detection in particle flow pattern. The result of the instability of particle flow is that not every laser shot ablation is efficient. For the laser ablation process, the spatial distribution of particle in the particle flow is random. The relatively small and limited laser focusing region may not be able to effectively ablate and excite enough particles to generate sufficient spectral information for application. For the spectral information obtaining, the randomness of particle distribution may cause the position and evolution direction of the plasma to shift. To obtain the plasma spectrum more efficiently, the spectra receiving region is relatively small and generally set according to the preset plasma center position. Minor changes in the spatial distribution of plasma may result in the obtained spectral information not being representative enough for further modeling and analysis. In this case, the obtained spectrum is considered invalid. It is of great significance to remove invalid spectra to improve the accuracy of quantitative detection. There have been many data screening methods were proposed for the elimination of invalid spectra, such as absolute intensity method,[14] SNR method,[29] SD method,[16] plasma image assisted screening method.[24]

Among them, the SNR method is the most used data screening method due to its ability to intuitively characterize the degree of particle excitation, simple calculation, no need for additional auxiliary equipment, and the ability to set multiple observation indicators. But the usability of SNR method has some room for improvement. The traditional SNR method use a universal SNR threshold for acceptance or rejection. When the SNR differences between specimens are large due to differences in physical and chemical characteristics, a fixed threshold is not effective for data screening. Moreover, the application purpose of LIBS is for rapid detection. So, the rapid quantitative analysis of unknown test objects is based on preset data processing schemes and models. But the ideal value for the SNR screening of unknown test objects cannot be estimated. The following data analysis case demonstrates this situation.

Carbon is the most important element of coal. As a non-metallic element, carbon excitation is relatively difficult, and part of the carbon is consumed in the generation of molecular components such as CN or C_2.[30] Therefore, when clear carbon atomic emission can be obtained, it means that the excitation has reached a high level. Therefore, in the detection of coal particle flow, carbon atomic emission is usually listed as the line of interest. Fig. 2 shows the SNR of C I 247.86 nm from 1050 pulse on two typical samples with significant differences in the SNR (N6 and S10), the

Fig. 3 RSNR of C I 247.86 nm emission of each pulse from coal N6 and coal S10 with or without spatial confinement.

average SNR is also shown as a red line simultaneously.

It can be intuitively found a significant difference of the single pulse SNR and the average SNR between coal N6 and coal S10. In the case of employing spatial confinement, the spatial confinement limits the active area of particles to a fixed and relatively small area and controls the shape and expansion direction of the plasma. Based on the constraint effect on particle flow and plasma evolution, the spatial confinement optimizes the ablation effect, enhances plasma intensity and stability, and controls plasma position for effective spectra harvesting. The SNR is improved to a certain extent, but cannot make the SNR of different specimens reach similar values. Namely, if the same screening threshold is adopted, the N6 specimen with smaller SNR may have too much pulse data removed, while the S10 sample with larger SNR cannot get an effective spectral screening. This is obviously unreasonable and ineffective and ultimately affect the accuracy of the following analysis.

To address the issues while retaining the advantages of SNR method, we made simple and effective improvements to the SNR method. We proposed a relative signal-to-noise ratio (RSNR) method for spectral data screening, which can achieve the purpose of avoiding complex methods for setting dynamic thresholds while overcoming the interference of large SNR differences. The RSNR is the ratio of SNR of each single pulse to the average SNR from multiple pulse, the calculation formula is as follows:

$$RSNR = \frac{snr_i}{\overline{snr}} \qquad (1)$$

where snr_i is the SNR of the *i* pulse, and $\overline{snr}$ in this work is the average SNR of specific characteristic spectral lines of 1050 pulses. Fig. 3 shows the RSNR of C I 247.86 nm from 1050 pulse on N6 and S10 with or without spatial confinement, and the average RSNR is also shown as a red line simultaneously. The mean SNR represents the average ablation effect, which is generally better than the application requirements. It will change with the detection object and time. The RSNR value characterizes the degree of deviation between each pulse and the average level, and serves as a discrimination criterion for data screening. Importantly, the RSNR values of different specimens are similar, and a fixed screening threshold could achieve more efficient and accurate representative spectra screening for different specimens. Furthermore, data screening can also be performed by setting thresholds for single or multiple indicators as needed based on RSNR values.

To clearly display the screening performance of SNR method and RSNR method, the spectral data from coal N6 and coal S10 are screening in accordance with the setting of a certain gradient increase threshold for C I 247.86 nm (Tables S2 and S3). The results show that the difference of the rejection rate of N6 and S10 is great under various screening thresholds in the case of SNR method. When the threshold setting basically meet actual demand and conform to the common value of the rejection rate of N6, the rejection rate of S10 is very low. It is not suitable for coal specimens with obvious differences, and the SNR improvement by spatial confinement cannot overcome such defects. In contrast, the RSNR method can achieve similar rejection rate when the SNR of different specimens vary greatly, and ensure the effect of eliminating enough unrepresentative and weak intensity spectra, while avoiding over-screening. This method is more convenient and accurate for the spectra screening and removal of various coal specimens. More importantly, effective screening can be achieved in both the modeling process of known objects and the testing process of unknown objects.

Screening threshold optimization. The selection of screening threshold determines the screening effect and ultimately affects the accuracy of the quantitative analysis. Setting an optimal threshold is the premise of establishing an accurate quantitative analysis model.[31,32] In this work, different thresholds for SNR and RSNR method were employed to screen the original spectral data, and the best thresholds in different cases were confirmed based on the cross-validation result. Using spatial confinement can significantly improve SNR, so the cross validation based on SNR method and spatial confinement appropriately increases the screening threshold. Due to the importance of carbon emission to coal detection, and carbon emission can effectively characterize the excitation effect and signal quality. The screening reference of the following screening still employed C I 247.86 nm. The verification which employed volatile matter content (one of the most important proximate analysis indexes of coal) as verification index was carried out using partial least squares ten-fold cross validation method (PLS-LTO-CV). The first 30 coal specimens in the Table S1, namely S1-S30, are employed as the cross-validation

Fig. 4 The RMSE of the cross-validation based on SNR or RSNR method with different threshold.

Fig. 5 Comparison of RSD of C I 247.86 nm and Mg I 285.21nm of coal N6 and coal S10 processed by different screening method.

set. The cross-validation set was divided into 10 parts on average, 9 of which were selected as the training set to establish the PLS regression model of volatile matter content, and the remaining part was employed as the test set. The root mean square error (RMSE) of the test set is regarded as the analysis result error of the cross-validation, and the specific formula is as follows:

$$RMSE = \sqrt{\frac{\sum_{i=1}^{n}(f_i - y_i)^2}{n}} \tag{2}$$

where n is the total sample size of the test set, f_i is the predicted value of the i sample in the test set, and y_i is the true value of the i sample.

The results are shown in Fig. 4, where the error bar is the relative standard deviation of RMSE for 10 cross-validation test sets. Overall, the RMSE of the cross-validation based on the RSNR method is generally smaller than that of the SNR method, which indicates that the spectral data screened by the RSNR method is more conducive to the accuracy of the quantitative model. The main reason is that as analyzed above, the SNR of C I 247.86 nm is differ widely between different coals. Under the same exclusion threshold, too much data is removed from the samples with low SNR or the threshold is too tolerant to effectively exclude unrepresentative data for specimens with relatively high SNR. In this case, the retained spectral data, whether averaged or used separately, will have a negative impact on the analysis performance. The result also found that, the RMSE decreases first and then increases with the increase of the threshold, and the smallest error occurs at the middle threshold. When the threshold is relatively small, some specimens still have a lot of invalid data in the spectral database after screening, which affects the spectral representativeness. On the other hand, when the threshold increases to a certain extent, excessive threshold eliminates part of the effective spectral data, resulting in a decrease in the database size and the utilization rate, thus affecting the accuracy of quantitative analysis. The minimum value of RMSE achieves 1.978 when employing spatial confinement and RSNR screening method with the threshold of 0.80.

To exhibit the optimization effect of data screening, the RSD of C I 247.86 nm and Mg I 285.21 nm of spectral data filtered by different screening methods with the optimal threshold are compared. As shown in Fig. 5, the RSD decreases obviously after the addition of spatial confinement, which has been proved to be one of the optimization effects of confinement.[28] The spatial confinement optimizes the average level of ablation effect and reduce the degree and number of pulses deviating from the mean. Therefore, the minimum RSD value is 9.87% in the case of RSNR method couple with spatial confinement. In general, the RSNR method can overcome the influence of the fluctuation of SNR between different samples. More importantly, RSNR method couples with the spatial confinement that has the optimization effect on spectral quality can achieve a mutually reinforcing effect.

Quantitative analysis. To further verify the availability of the above spectral screening methods and spatial confinement, quantitative models of coal proximate analysis indexes were established based on multivariate partial least squares regression (PLSR). Then the model performance was compared, and the optimization effect of spatial confinement and RSNR method was evaluated. PLSR is a method based on multiple linear regression, combining the characteristics of principal component analysis and multiple linear regression, which has been proved to be a common and effective modeling method for LIBS coal detection.[4-7] The input database for modeling included the whole spectra data of all six channels of spectrometer, and the quantitative models were established employing four different processing: SNR method, RSNR method, spatial confinement + SNR method or spatial confinement + RSNR method.

When coal is used as fuel, the increase of ash content means the corresponding reduction of combustible substances and the increase of mineral in coal. The combustion of ash as well as the

Fig. 6 Prediction result of ash content in each case: (a) SNR method, (b) RSNR method, (c) SNR method and spatial confinement, and (d) RSNR method and spatial confinement.

slag generation lead to heat consumption. Therefore, the ash content is one of the important indexes to evaluate coal quality. In this work, a quantitative model for ash content was firstly established based on PLSR to verify the improvement of the above optimization means.

In the prediction process of the test set, the 1050 pulses of each coal specimen were divided into 5 groups. Five repeated predictions were made for the preset indicator, and the final prediction result was the average value of the five repeated predictions. The coefficient of determination (R^2), root mean square error of prediction (RMSEP), average absolute error (AAE) and standard deviation (SD) of the five repeated predictions were employed to evaluate the model performance. The closer R^2 is to 1, the better the regression effect of the model is. The RMSEP, AAE and SD represent the error of the predicted value and repeated prediction volatility, respectively. The calculation formula is as follows:

$$R^2 = 1 - \frac{\sum_{i=1}^{n}(\hat{y}_i - y_i)^2}{\sum_{i=1}^{n}(y_i - \bar{y})^2} \tag{3}$$

$$RMSEP = \sqrt{\frac{\sum_{i=1}^{n}(\hat{y}_i - y_i)^2}{n}} \tag{4}$$

$$AAE = \frac{\sum_{i=1}^{n}|\hat{y}_i - y_i|}{n} \tag{5}$$

$$SD_i = \sqrt{\frac{\sum_{k=1}^{m}(\hat{y}_{ik} - \hat{y}_i)^2}{m-1}} \tag{6}$$

Where n is the total sample size of test set, m is the number of repeated measurements, $\hat{y}_{ik}$ represents the k^{th} time predicted value of the i sample, $\hat{y}_i$ represents the average of the 5 times repeated predicted value of the i sample, and SD_i represents the standard deviation of the 5 times repeated predicted value of the i sample. The y_i is the reference value of the i sample, then $\bar{y}$ represents the average of all reference values for n samples.

Figure 6 and Table S4 shows the prediction results of quantitative ash analysis for all test set samples in each case; SD is the standard deviation of the predicted value for 5 times prediction. The error bar is the standard deviation of the predicted value for 5 times prediction. The results in Fig. 6 and Table 4 show that the RSNR method and spatial confinement can significantly improve the model performance. When SNR method was used with spatial confinement, R^2 increased from 0.9225 to 0.9834, RMSEP and AAE decreased from 3.5667% and 2.7720% to 1.6866% and 1.3750%, respectively. In the case of RSNR method, spatial confinement increased the R^2 from 0.9334 to 0.9845, RMSEP and AAE decreased from 3.3730% and 2.6350% to 1.6280% and 1.3973%, respectively. The spatial confinement effectively stabilizes the particle flow and plasma generation and propagation, which not only greatly reduces the fluctuation of the spectra, but also improves the intensity and reliability. Therefore, the quantitative analysis model established in spatial confinement condition is more accurate, and it also achieves a smaller standard deviation in repeated prediction. Taking specimen N4 as an example, under no-confinement condition, the SD values of SNR method and RSNR method are 3.68 and 3.17, respectively, indicating that the fluctuation in predicted values is relatively large. After the addition of spatial confinement, the SD are reduced to 0.95 and 1.14, respectively. In terms of the data screening method optimization, the RSNR method also significantly improve the model performance. In the condition of employing RSNR method, the R^2 of the prediction model was increased from 0.9225 and 0.9834 to 0.9334 and 0.9845 under no-confinement and spatial confinement condition, and the RMSEP were also decreased. The optimization on detection can effectively improve the original spectral data, while data processing method provide greater fault tolerance for spectral data. In the use of a single method, the detection system optimization has an upper limit on the quality improvement of source data, and the data screening method cannot achieve good processing effect on low quality source data. The optimization of detection needs to be combined with appropriate and high-quality data screening method.

The combustible species in coal are the main component for combustion heat generation, such component is called fixed carbon. This work also compared the performance of fixed carbon model, as shown in Fig. 7 and Table S5. The effect of employing spatial confinement and RSNR method on the model performance is consistent with the effect on the above ash model. In the case of spatial confinement + RSNR method, the R^2 achieves 0.9736, RMSEP and AAE also reach the minimum at 2.0344% and 1.6777%, respectively.

Fig. 7 Prediction result of fixed carbon content in each case: (a) SNR method, (b) RSNR method, (c) SNR method and spatial confinement, and (d) RSNR method and spatial confinement.

Fig. 8 Prediction result of volatile matter content in each case: (a) SNR method, (b) RSNR method, (c) SNR method and spatial confinement, and (d) RSNR method and spatial confinement.

Fig. 9 Prediction result of calorific value in each case: (a) SNR method, (b) RSNR method, (c) SNR method and spatial confinement, and (d) RSNR method and spatial confinement.

As one of the important indexes to evaluate the coal quality, volatile matter in coal mainly comes from organic components and has a direct relationship with non-metallic elements such as C, H, O and N, which determine the ignition and combustion characteristics of coal. The prediction results of volatile matter models are shown in Fig. 8 and Table S6. The spatial confinement + RSNR model achieves the best R^2 at 0.9815. Suitable and effective data screening and the detection optimization also show its effectiveness in improving the quantitative analysis of volatile matter content.

The calorific value of coal refers to the heat released per unit mass of coal when the fuel product is cooled to the ambient temperature after complete combustion. Calorific value is mainly related to the organic component of coal, which is one of the important criteria to evaluate coal quality and determine coal pricing. As shown in Fig. 9 and Table S7, the model employing spatial confinement and RSNR method shows the best performance, the R^2, RMSEP and AAE achieve 0.9897, 0.4553% and 0.3794%.

On the whole, detection optimization essentially optimizes the laser-particle flow interaction and plasma evolution process to achieve source data quality optimization, but it cannot completely overcome the interference to the detection caused by the inevitable instability of particle flow. The data screening scheme is based on the evaluation of data quality and representativeness after the acquisition of spectral information. When the quality of the source spectral signal is low, the effective data acquisition is extremely inefficiency or even impossible. Therefore, the two optimization methods need to be coupled with each other to achieve mutual gain.

The above quantitative analysis results of ash, fixed carbon, volatile matter and calorific value verify the practicability and the positive effect of RSNR method and spatial confinement on the LIBS quantitative analysis of particle flow. Employing the spatial confinement, the influence of particle flow fluctuation can be effectively reduced from the root, the effective ablation on the sample can be promoted, thereby optimizing the spectral signal quality including the repeatability, SNR and intensity. High quality spectral data compatible with RSNR methods can achieve better quantitative results.

CONCLUSION

The inevitable fluctuation of particle flow affects the stability and effectiveness of laser ablation and spectral information acquisition in LIBS detection. In this work, spatial confinement and data screening were employed to optimize the LIBS quantitative analysis of coal particle flow. Firstly, the drawbacks of spectral data screening based on fixed SNR values were discussed. Then proposed a new RSNR screening method and optimized spectral quality with spatial confinement to achieve effective screening of spectra with fixed RSNR values. Subsequently, the screening threshold was optimized using cross validation and the optimization effect on spectral quality was verified. Finally, quantitative analysis of coal particle flow was carried out based on coal proximate analysis indexes. The results showed that the spatial confinement and RSNR effectively optimize the quantitative model performance. The RMSEP of ash content, fixed carbon content, volatile matter content and calorific value were improved from 3.5667%, 3.3965%, 3.0905% and 1.0188MJ/Kg to 1.6280%, 2.0344%, 1.4999% and 0.4553MJ/Kg. The innovation and combined application of spatial confinement and data screening have reference significance for optimizing the application of LIBS in quantitative analysis of coal particle flow.

ASSOCIATED CONTENT

Supporting information (Tables S1-S7) is available at www.at-spectrosc.com/as/home

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Notes

The authors declare no competing financial interest.

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