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Automatic rock classification of LIBS combined with 1DCNN based on improved Bayesian optimization

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Abstract: To achieve automated rock classification and improve classification accuracy, this work discusses an investigation of the combination of laser-induced breakdown spectroscopy (LIBS) and the use of one-dimensional convolutional neural networks (1DCNN). As a result, in this paper, an improved Bayesian optimization algorithm has been proposed where the algorithm has been applied to automatic rock classification, using LIBS and 1DCNN to improve the efficiency of rock structure analysis carried out. Compared to other algorithms, the improved Bayesian optimization method discussed here allows for a reduction of the modelling time by about 65% and can achieve 99.33% and 99.00% for the validation and test sets of 1DCNN.

1. Introduction

Coal remains one of the main energy sources in wide use in China. In 2020, China's coal consumption accounted for 56.8% of its total energy consumption,[1] while in 2021, there was a small decrease in consumption (to 56.0% of the total energy consumption), and raw coal power generation to ~67.0% of the total power generation (http://www.stats.gov.cn/xxgk/jd/sjjd2020/202210/t20221008_1888971.html). There is a large number of coal mines operating in China and most involve underground mining. To achieve efficient production from such mining and reduce the risk of accidents for underground workers, intelligent mining and unmanned mining are seen as the way forward. Rock and lithology analysis plays an important guiding role in many aspects of the field, such as mining and geological disaster analysis. Through lithology analysis, real-time geological data can be provided for unmanned mining and intelligent mining.[2] The traditional lithology judgment method used relies on the experience of staff to judge the appearance and physical properties of the ore: a task which requires an extremely high level of professional expertise and identification experience from the staff involved, but unfortunately the efficiency of manual identification is not sufficiently high. With the continuous development of compositional analysis techniques as has been seen in the field of spectroscopy, a series of techniques such as X-ray fluorescence spectroscopy (XRF), X-ray diffraction analysis (XRD), and gamma spectroscopy are widely used for rock classification. However, the sample production process using these methods is relatively complex and the measurement time is relatively long. Therefore, the need is for rapid, accurate and in situ geological rock or mineral identification which the technique discussed in this paper can offer.[3-5]

Laser Induced Breakdown Spectroscopy (LIBS) is a laser-based optical spectroscopy technique which operates by shining high-energy pulses on the surface of the sample to create gasification of the sample, to generate a plasma which is studied. This emits a spectrum during

48 the process of plasma diffusion and cooling, allowing the collection of spectral data with a
49 spectrometer, and then conducting a qualitative analysis of the material tested, based on the
50 wavelength and intensity of the spectral peaks seen.[6-8] This approach has the advantage of
51 multi-form analysis and is fast in operation. Further, the approach is non-destructive, shows a
52 low detection limit, with there being no need to use vacuum environment. All this points to the
53 LIBS technique being well suited to field detection applications. Therefore, LIBS is a technique
54 that is widely used in the fields of environmental monitoring,[9-11] metallurgy,[12-14]
55 medicine,[15-18] food,[19, 20] heritage science,[21] planetary exploration missions,[22, 23]
56 and mineralogy.[24-32]

57 Wang C et al. used PCA to reduce the dimension of each spectral signal, and then used
58 linear discriminant analysis, and a random forest and a support vector machine approach to
59 classify the spectral data, after dimensional reduction. The results indicate that SVM could well
60 be applied to LIBS classification of rock.[33] We note that El-Saeid et al. used PCA and Graph
61 Theory methods to classify spectra obtained in rocks using two methods (standard LIBS, and
62 Nanoparticle-Enhanced LIBS), showing that excellent classification of the rocks analyzed (with
63 more than 99% of the spectra correctly classified) could be obtained using standard LIBS,
64 coupled to Graph Theory analysis.[34] Yelameli et al. studied the effect of increasing the
65 number of shots per rock and the detrend operation, showing that the number of dimensions
66 could be effectively reduced by applying PCA. The results obtained indicate that the SVM
67 algorithm with the detrend operation, combined with a specific number of shots, creates a good
68 rock classification effect (with an accuracy greater than 95%).[35] Janovszky et al.
69 demonstrated that LIBS mapping, with spatially resolved local analysis offers an efficient and
70 practical approach for the classification of mineral grains. The results indicate that the
71 classification accuracy obtained is better than 92%, using random forest and linear discriminant
72 analysis. Direct classification by evaluating the presence of feature elements is a powerful
73 approach.[36] In this paper, a classification model optimization algorithm has been proposed
74 and applied to the rock recognition method, combining LIBS and 1DCNN. The positive effect
75 is to reduce the modeling time of the classification algorithm and to improve the efficiency of
76 the rock classification, as an effective means to improve the classification accuracy.

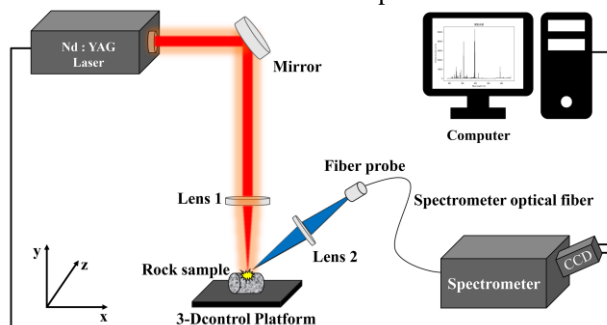
77 In this paper, LIBS technology, combined with a variety of machine learning algorithms
78 (principal component analysis, grid search cross validation, random search cross validation,
79 Bayesian optimization based on Gaussian process, improved Bayesian optimization, support
80 vector machine, one-dimensional convolutional neural network, etc.), is expanded to use to
81 carry out comparative experiments on rock classification for the mining industry. The goal is
82 to automate rock classification and provide reference data that will enhance lithology
83 identification in the mining industry, through a series of straightforward steps that can easily
84 be applied, from inputting raw LIBS rock spectral datasets to outputting rock classification
85 results. In use, a pre-processing operation first is selected to obtain the relevant LIBS spectral
86 data, where the pre-processed spectral data are 'dimensioned-down' through a principal
87 component analysis technique, following which the corresponding number of principal
88 components is selected as the input variable for the classifier, used for different classifier
89 models. Three optimizers were constructed and used to conduct comparative experiments on
90 the classifier model, including the combination of grid search and random search optimization,
91 Bayesian optimization based on Gaussian process and improved Bayesian optimization,[37]
92 which can improve the identification of the samples under consideration.

93 **2. Experiment**

94 *2.1 Experimental setup used*

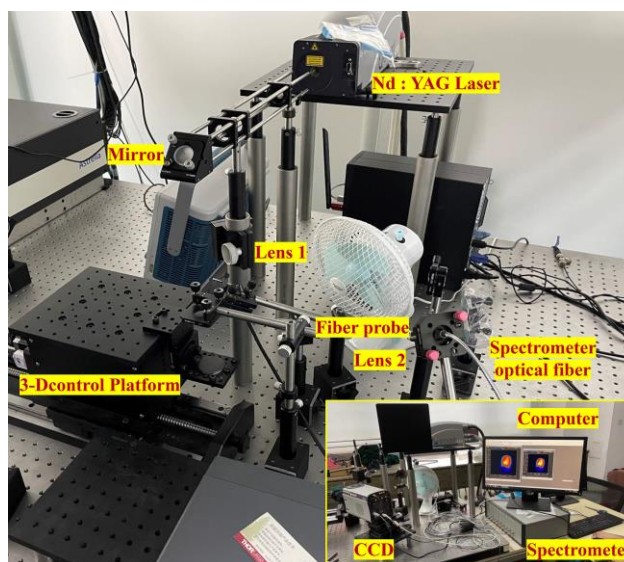
95 Fig.1 and Fig.2 respectively illustrate a schematic diagram and show photographs of the LIBS
96 experimental setup. The laser source used in this work was a Q-switched Nd: YAG (Q-smart
97 450) pulsed laser (from Quantel, France), which was used to generate a laser beam with pulse
98 repetition rate of 8Hz, a pulse width of 7ns and an output wavelength of 1064nm. A laser beam

99 is focused on the surface of the rock to be tested (using a quartz lens with a focal length of
100 100mm) to generate the plasma. When the species in the plasma make a transition from an
101 excited to a low-level or ground state, light is emitted and this is collected by using a lens (with
102 a diameter of 10mm and a focal length of 15mm). Here the optical signal is collected using a
103 fiber optic probe and fed to an 8-channel spectrometer (Avantes-usb8) equipped with a CCD
104 detector, where it is converted into an electrical signal and transmitted to a computer for
105 analysis and processing. The pulsed laser used emits a laser pulse energy of 35mJ, where the
106 wavelength range of the spectrometer used was 181-673nm, thus covering a wide range. Further,
107 the delay time of the acquisition of the spectrum is 160 μ s, the integration time is 1.05ms, and
108 the spectral resolution of the different channels of the spectrometer is 0.058-0.068nm.



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110

Fig. 1. Schematic diagram of the experimental setup.



111
112

Fig. 2. Photographs of the experimental setup.

113 2.2 Sample preparation

114 In this paper, 10 different and representative types of sedimentary rock samples, taken from the
115 rock that forms the roof of Yanzhou coal mine (owned and operated by Shandong Energy
116 Group) were selected for analysis, in consultation with mining experts: these being Siltstone,
117 Oil shale, Argillaceous siltstone 1, Gritstone 1, Argillaceous siltstone 2, Mudstone 1, Red
118 sandstone, Mudstone 2, Gritstone 2, and Fine sandstone, as detailed in Table 1 and Fig.3. To
119 remove dust or any environmental effects from the samples, several 'cleaning' shots (i.e. 10
120 laser pulses applied to six different locations of the samples) were fired at them, before any
121 data sets were acquired. To overcome the heterogeneity of the rocks, 40 laser irradiations were

122 performed on the same acquisition site in the course of the experimental work – where here the
 123 laser focus position was controlled by a laser ranging feedback control 3D platform. In order
 124 to eliminate the apparently unstable plasma spectra, it was necessary to eliminate the anomalous
 125 spectra and to do so, the specific process was as follows:

- 126 ① The M spectral information of a measurement sample, after 40 repeated measurements,
 127 was used as the independent variable matrix to obtain a $40 \times M$ matrix, following which
 128 the average value of each column was calculated to obtain a matrix, of row vector $1 \times M$.
 129 Each element of this row vector is the average spectral intensity of the corresponding row;
- 130 ② The sum of the squares of the differences between each measurement value and the mean
 131 value were calculated;
- 132 ③ The measured spectra which were larger than 1.1 times the mean of the sum of squares
 133 were removed to complete the screening of abnormal spectral data. This process was found
 134 to remove about two-fifths of the original data. Finally, 960 sets of spectral data were
 135 obtained as the original data set used in the subsequent analysis.

136 To eliminate the influence of the time taken for the work to be done on the spectral data
 137 collected, 50 sets of data were extracted from each rock sample in sequence. Thus, in total, 500
 138 spectral data sets were formed from the 10 different rock samples (seen in Table 1) to ensure
 139 that each was equally sampled.

140 **Table 1. Rock category information: illustrating Rock Category Label and Rock Type Name**

Rock Category Label	Rock Type Name
Rock 0	Siltstone
Rock 1	Oil shale
Rock 2	Argillaceous siltstone 1
Rock 3	Gritstone 1
Rock 4	Argillaceous siltstone 2
Rock 5	Mudstone 1
Rock 6	Red sandstone
Rock 7	Mudstone 2
Rock 8	Gritstone 2
Rock 9	Fine sandstone



Fig. 3. Photographs of 10 kinds of rocks.

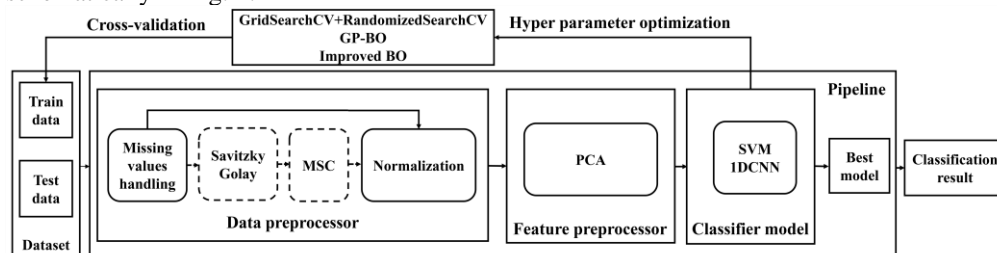
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142

143 *2.3 Spectral data pre-processing*

144 The experimental system error of the instrument used can interfere with the LIBS spectrum
 145 received. This can result from changes in the external environment, random noise in the spectral
 146 line signature seen and the diffuse reflection of the solid. As a result, the spectral data collected
 147 inevitably contain irrelevant information, such as from the pump light, from stray light that has
 148 been collected and molecular vibration effects from the spectral species involved. Spectral data
 149 from material other than the rock samples themselves i.e. background spectral data will be
 150 superimposed on, and can potentially interfere with, the collected spectral data. All this will
 151 negatively affect the accuracy of the classification of the rock samples and slow the speed of
 152 iteration of the classification model created. Therefore, it is necessary to eliminate signals
 153 which can cause such errors in the spectral signals received from the LIBS process, while still
 154 retaining the characteristic spectra of the samples themselves, to provide an appropriate basis
 155 for the subsequent analysis of the LIBS data.

156 In this paper, it is recognized that missing values in the data sets will occur and this is
 157 undesirable. However, to compensate, and thus to choose appropriate values to complete the
 158 sets, a Savitzky Golay smoothing filter (polynomial order is 4, window width is 5), and a
 159 Multiplicative Scatter Correction (MSC) approach, coupled with normalization as the optimal
 160 pre-processing method, with basic pre-processing was carried out to enable the results from
 161 comparative experiments to be evaluated. The experimental process used is shown
 162 schematically in Fig. 4.



163
 164

Fig. 4. Flow chart of the experiment.

165 To prevent data leakage, only the training set, in the pre-processing and cross-validation
 166 hyperparameter optimization, is processed. The validation set and test set are used for
 167 hyperparameter optimization and model testing experiments through the pipeline mapping
 168 transformation. The set is transformed through a pipeline map for hyperparameter optimization
 169 and model testing experiments. In this approach, 35 sets of spectral data were randomly selected
 170 from the 50 sets of spectral data from each type of rock studied (see Table 1), to form the
 171 training set. This training set, comprising 350 sets of spectral data, was used to train the model
 172 and to optimize the hyperparameters; the remaining 15 sets of spectral data (from each type of
 173 rock) form the test set. As a result, 150 sets of spectral data were used finally to test the accuracy
 174 of the classification model used. The algorithms described in this paper were all implemented
 175 in Windows10, 64-bit system, Python 3.9.7 version, and using the Jupyter Lab 3.3.2
 176 development environment.

177 The pre-processing operation used can be described as follows. Firstly, the negative value
 178 in the spectral intensity value of the data to be processed is defined as NaN, and then converted
 179 to a value of 0, following which the Savitzky Golay was used for smoothing filtering, and then
 180 the effect of scattering was eliminated by use of the MSC process. Here Multiplicative Scatter
 181 Correction (MSC) is one of the common methods of spectral data pre-processing used. Due to
 182 the diffuse reflection and surface inhomogeneity of the solid sample on which the measurement
 183 was carried out, the spectral differences caused by the presence of the different scattering levels
 184 could be eliminated by the use of the MSC algorithm. Thus, the phenomenon of baseline drift
 185 in the spectrum could be dealt with, thereby enhancing the correlation between the spectrum
 186 obtained and the original data. The specific process by which MSC is carried out can be
 187 described as follows.

188 1) The mean value of the spectral data used was taken as the ‘standard spectrum’. Thus,
 189 the LIBS standard spectra for the 10 rock types used in this study are shown in Fig. 5
 190 below.

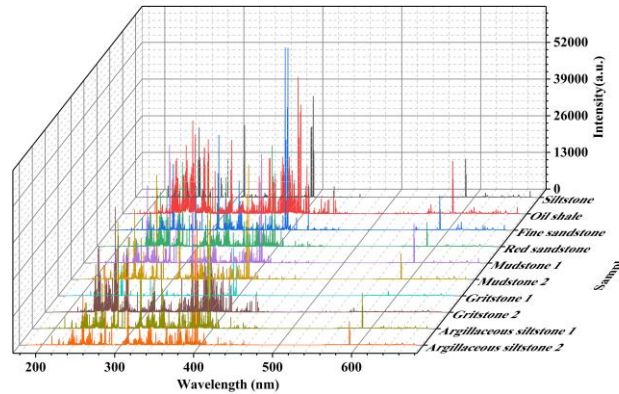
191
$$\bar{A} = \frac{\sum_{i=1}^n A_i}{n} \quad (1)$$

192 2) A univariate linear regression is performed on the average spectrum by importing the
 193 Linear Regression module in the sklearn library, and the linear translation and tilt
 194 offset of each spectral data set, relative to the average spectrum are obtained, by
 195 solving the least squares problem.

196
$$A_i = m_i \bar{A} + b_i \quad (2)$$

197 3) A Multivariate Scattering Correction is applied for each spectral data point: the
 198 corrected spectral data are obtained by subtracting the linear shift from the spectral
 199 data and dividing by the regression coefficient.

200
$$A_{i(MSC)} = \frac{(A_i - b_i)}{m_i} \quad (3)$$



201

Fig. 5. LIBS standard spectra for 10 rock types.

202
 203 Each spectral data point is corrected, with reference to the average spectrum (this process
 204 does not affect the information each absorbs), to improve the signal-to-noise ratio of the spectral
 205 data overall. The spectral data are then normalized by use of the following formula:

206
$$x_1 = \frac{x - x_{min}}{x_{max} - x_{min}} \quad (4)$$

207
$$x_2 = x_1 * (Max - Min) + Min \quad (5)$$

208 In the above formula, x represents the value of the intensity of each group of spectral data
 209 obtained at each wave point; x_{min} is the minimum intensity of this group of spectral data; x_{max}
 210 is the maximum intensity of this group of spectral data; this is normalized to the interval 1, and
 211 x_2 represents the normalized spectral data.

212 2.4 Models and Algorithms

213 Principal Component Analysis (PCA) allows for creating a data dimensionality
 214 reduction, and thus feature extraction, by mapping n-dimensional features to k-
 215 dimensions, which are brand-new orthogonal features, also known as principal
 216 components. In this experiment carried out, each group of spectra contains 10239 wave

217 points. If this is used as the input of the classification model, problems such as
218 dimensionality disaster, low efficiency of the classification model, and long training
219 time can be seen.

220 The experimental work carried out and reported in this paper uses two machine learning
221 classification models (SVM, 1DCNN), combined with three hyperparameter optimization
222 methods (GS-RS, GP-BO and Improved BO) to conduct comparative experiments on data
223 that are pre-processed, or not pre-processed. Support Vector Machine (SVM) is a
224 machine learning algorithm, based on Statistical Learning Theory (STL) (and created by
225 Vapnik) to deal with binary classification problems. The SVM approach realizes the
226 classification of samples by finding the optimal classification hyperplane that satisfies
227 the constraints. In the linear classification, the classification surface is selected by taking
228 the farthest distance from the two samples. In the case of a nonlinear classification, it is
229 processed through a transformation in high-dimensional space. To reduce the amount of
230 calculations needed and the complexity of the model, a kernel function is introduced to
231 replace the dot product process in the high-dimensional feature space. The kernel
232 function formula is given by.

$$233 \quad K \left[x^{(i)}, x^{(j)} \right] \equiv \Phi \left[x^{(i)} \right]^T \cdot \Phi \left[x^{(j)} \right] \quad (6)$$

234 In the above Equation (6), $\Phi \left[x^{(i)} \right]^T$ and $\Phi \left[x^{(j)} \right]$ are n -dimensional features $x^{(i)}$ and
235 $x^{(j)}$ are mapped to \bar{n} dimensional features. Following that, the SVM classifier model was
236 optimized by adjusting the penalty coefficient, C , and the kernel function parameter, γ .
237 In this paper, a Radial Basis Function (RBF) kernel function was used, which is given as shown
238 below (where δ is the kernel width, $\delta > 0$)

$$239 \quad K \left[x^{(i)}, x^{(j)} \right] = e^{-\frac{\|x^{(i)} - x^{(j)}\|^2}{2\delta^2}} \quad (7)$$

240 A Convolutional Neural Network (CNN) is a deep neural network model with the
241 characteristics of neuron weight sharing and local connection. It is important because it has
242 powerful feature extraction capabilities in high-dimensional data. When the traditional fully
243 connected network model processes data, due to the large number of parameters present,
244 problems such as a large amount of calculation needed, the low model efficiency, and local
245 invariance are seen. Convolutional Neural Networks reduce the number of parameters by
246 locally connecting each neuron, sharing weights for each group of connections, and adding
247 pooling layers, while improving the robustness of the model and the ability of the network to
248 generalize. For one-dimensional spectral data from the rock samples considered, it creates a
249 kind of multivariate sequence data, using a one-dimensional convolutional neural network
250 (1DCNN) model. By performing the one-dimensional convolution operation on the input
251 spectral data, the feature information can then be extracted to create an effective classification
252 from the different types of rocks considered.

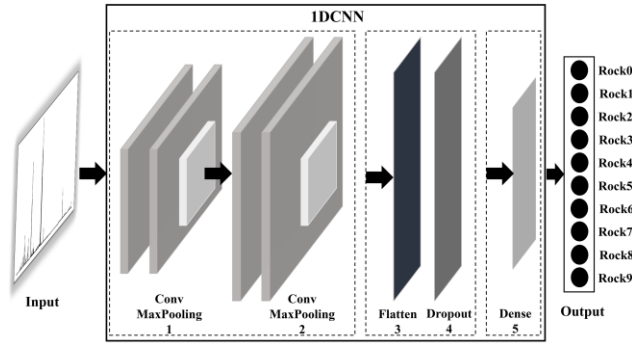


Fig. 6. The structure of one - dimensional convolutional neural network.

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The model, 1DCNN uses 4 convolutional layers to extract features, according to the spectral data from the rocks investigated. Each convolutional layer adds an activation function to improve the ability of the neural network to express an effective classification model. After each of the 2 convolutional layers, a maximum pooling layer was added. To preserve the main features and reduce the computational cost, a flattening layer and a Dropout Layer were added in the middle, where the flattening layer converted the high-dimensional data into one-dimensional data and acts as a transition from the convolutional layer to the fully connected layer. Further, the Dropout Layer randomly assigns 50% of the neurons in the network to zeros. By resetting the weight to zero, the weight reduces the sensitivity of the convolutional neural network to any small changes experienced in the data and further improves the accuracy of the processing of unknown data. After that, a fully connected layer was then added and the softmax function was further added as the output layer, to obtain each feature, with data matching the feature category with the highest probability. The Cross Entropy Loss function was selected as the model loss function, where the formula used is as shown in Equation (9). The corresponding network structure of the 1D-CNN was as is shown in Fig. 6 above, where the detailed parameters of the network structure used are as shown in Table 2. The Softmax function formula employed was as follows.

$$Softmax(x_i) = \frac{\exp(x_i)}{\sum_{i=1}^n \exp(x_i)} (i = 1, 2, \dots, n) \quad (8)$$

In Equation (8), x_i represents the input of the Softmax function, and the n-dimensional real vector was mapped to the (0, 1) interval through the Softmax function. The sum of all probabilities is 1 and consequentially the probability distribution of multi-classification can be obtained.

$$Loss = -\sum_{i=1}^n y_i \cdot \log y_i \quad (9)$$

278

Table 2. The parameters of 1D - CNN

Number	Network layer	Parameter	Conv kernel	Step size
	Input layer	145×1	-	-
1	Conv-1	5×1	100	1
	Conv-2	5×1	100	1

	Max-Pool-1	3 × 1	-	1
	Conv-3	5 × 1	150	1
2	Conv-4	5 × 1	150	1
	Max-Pool-2	3 × 1	-	1
3	Flatten	-	-	-
4	Dropout	0.5	-	-
5	Dense	10 outputs	-	-

279 Grid Search Cross Validation (GS) is a basic hyperparameter optimization technique. Here,
280 through defining an n-dimensional grid, each grid has a hyperparameter map, and the optimal
281 hyperparameters of the model are obtained by exhausting all the hyperparameter combinations,
282 through cross-validation. In this technique, assuming that there are n hyperparameters, and each
283 hyperparameter has P_i values, then with N-fold cross-validation, the number of
284 hyperparameters to be evaluated is $N \times (\prod_{i=1}^n P_i)$. When this number of hyperparameters is
285 large, although the optimal hyperparameter combination can be obtained in the end, this will
286 lead to dimension explosion and low model efficiency. Randomized Search Cross Validation
287 (RS) obtains the optimal hyperparameter combination in the sampling group by randomly
288 selecting the hyperparameter combination in the hyperparameter space for a given number of
289 iterations n_iter with cross-validation. The use of a random search solves the problems of a
290 large number of grid search combinations, a large amount of computation, and a long time over
291 which it is done, but only suboptimal solutions can be obtained. In this paper, the SVM
292 classification model is used to optimize the super parameters, C and gamma through both grid
293 search cross validation and random search cross-validation. First, the grid range of the optimal
294 combination is determined through a grid search, following which an optimal solution is
295 obtained by use of a random search. Therefore, the use of grid search cross-validation combined
296 with random search cross-validation can be used to shorten the time taken and thus obtain better
297 optimization results.

298 Bayesian Optimization (BO) regards the function of the optimized object as a random
299 process that satisfies the prior distribution and allows obtaining a new distribution by updating
300 the posterior probability with the Bayesian formula, by solving the function value. Following
301 this, it then judges the most likely value, according to the new distribution. The extreme point
302 and its function value can be calculated to form a new function value observation history. The
303 next stage is to set the number of iterations, repeat the above process, and thus to obtain the
304 optimal solution. It can be noted that the formula from Bayes' theorem can be given, as follows.

$$305 \quad P(f | D_{1:t}) = \frac{P(D_{1:t} | f) P(f)}{P(D_{1:t})} \quad (10)$$

306 Where in the above formula f is the objective function; $D_{1:t} = \{(x_1, y_1), \dots, (x_t, y_t)\}$
307 represents the observed set, x_t represents the decision vector, $y_t = f(x_t) + \varepsilon_t$ represents the
308 observation value, ε_t represents the observation error; $P(D_{1:t} | f)$ represents the likelihood
309 distribution of y ; $P(f)$ represents the prior probability distribution of f , which is used
310 to describe the assumption made about the state of the unknown objective function; $P(D_{1:t})$
311 represents the posterior probability distribution of f which is used to describe the confidence
312 of the unknown objective function after the prior probability is corrected by the measured data
313 set.

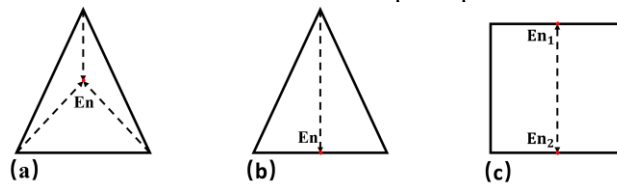
314 Gaussian Process-Based Bayesian Optimization (GP-BO) uses Gaussian Processes (GP)
 315 to build a surrogate model to simulate the objective function for black-box optimization and in
 316 that way speed up the convergence. The specific idea behind its use is as follows: by obtaining
 317 the posterior probability of the observed point x , the mean μ and variance σ of the x point can
 318 be calculated, following which the value of x is determined according to the extraction function
 319 u . After that, the process continues to sample the objective function and evaluate the objective
 320 function value, then integrate the data and update the Gaussian surrogate model for fast
 321 convergence. The formula of the function, u , is given as shown below in Equation (11).

$$322 \quad u(x) = \operatorname{argmax}_{x \in D} \mu_{t-1}(x) + \beta_t^{\frac{1}{2}} \sigma_{t-1}(x) \quad (11)$$

323 In this paper, an improved Bayesian optimization algorithm (a Bayesian fast automatic
 324 hyperparameter optimization method based on simplex optimization domain) is proposed. In
 325 this approach, first, we construct the simplex optimization domain and sample the partition in
 326 an internal division. Then create a work queue through the heappop function of the heapq stack
 327 column module in Python; As shown in Fig. 7 below, the two division methods of simplex and
 328 square are respectively selected to optimize the domain partition, and on the assumption that
 329 each partition can be represented by the sampling points of its vertices, the partition efficiency
 330 can be defined as E . The formula for E is given as Equation (12):

$$331 \quad E = \frac{En'}{En} \quad (12)$$

332 In the above formula, En' is the number of new partitions, and En is the number of sampling
 333 points required to divide the new area. It can be noted that when bisecting the square
 334 optimization domain, two sampling points are required; when bisecting from the inner third of
 335 the simplex or an edge, only one sampling point is required. The next step is to divide the
 336 interior of the simplex and the sides of the simplex and square respectively, and the three
 337 partition efficiencies E are $(n+1)/1$, $2/1$, $2/2^{n-1}$. This gives $E_a > E_b > E_c$.
 338 Therefore, it is seen as most efficient to divide the simplex optimization domain internally.



339
 340

Fig. 7. Optimized domain partitioning efficiency comparison.

341 Following this, the hyperparameter space can be sampled according to Inverse Distance
 342 Weighting (IDW), and local interpolation is then performed in a simplex manner. The inverse
 343 distance weighted interpolation is a spatial interpolation method that can be used to estimate
 344 the location of the next point to be measured by using the measured sample points with
 345 corresponding weights. The inverse distance weighted interpolation formula used is shown
 346 below.

347
$$x^* = \frac{w_1x_1 + w_2x_2 + w_3x_3 + \dots + w_nx_n}{w_1 + w_2 + w_3 + \dots + w_n} \quad (13)$$

348 Where in the above formula, x^* is the next sample point to be measured; x_i ($i = 1, 2, \dots, n$) is
 349 the i -th measured sampling point; w is the weight. The formula for the weight of the i -th
 350 measured sampling point used is shown below.

351
$$w_i = \frac{1}{d_{ix^*}^p} \quad (14)$$

352 Here, the weight w is inversely proportional to the p power of the distance d (from the point to
 353 be measured x^* to the known sampling point x_i), where the Euclidean Metric is selected to
 354 calculate the distance d , and the distance formula is as given in Equation (15) shown. As a
 355 result, the corresponding weighting decreases as the distance increases and the rate of descent
 356 depends on the pre-set constant p . When $p = 0$, the relative weight is 1, and the prediction
 357 point is the average value of the measured data in the search field; and when p is too large,
 358 the weight decreases rapidly. It is only the sampling points of the nearest points which have an
 359 effect on the predicted points. Therefore, the default value is $p = 2$, and the interpolation
 360 method is the inverse distance square weight interpolation.

361
$$d = \sqrt{(x_2 - x_1)^2 + (y_2 - y_1)^2} \quad (15)$$

362 In the above formula, d is the Euclidean distance between the point (x_2, y_2) and the point
 363 (x_1, y_1) .

364 Next, the number of iterations is set and the local interpolation value which gives the
 365 highest information gain is selected from the priority queue of the points to be measured
 366 through each iteration, and the acquisition function is used to prevent the acquisition of a sub-
 367 optimal local interpolation value, to ensure the faster convergence of the objective function.
 368 The acquisition function used is shown below.

369
$$f(x^*) = x^* + C * T \quad (16)$$

370 In the above formula, C is the weight of the optimization domain exploration, which is used
 371 to inform the optimizer of how much attention should be paid to the current exploration
 372 optimization domain to prevent the pursuit of the first Sub-optimal value explored by the
 373 optimizer. C is an adjustable constant, and the default is $C = 0.1$, where T is the exploration
 374 cost. The formula used for the value of T is as shown below.

375
$$T = (X_1 - X_2) * \log_{EN} (F_1 * F_2) \quad (17)$$

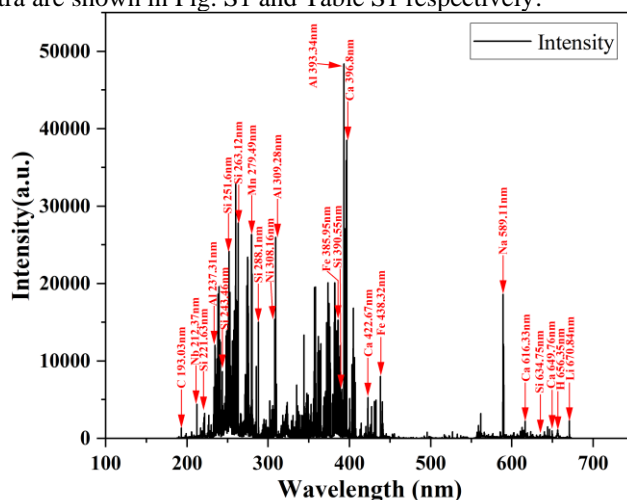
376 In the above formula, X_1 and X_2 are the best sampling point and the worst sampling point
 377 in the simplex sampling domain respectively, EN is the number of all the vertices in the
 378 simplex optimization domain, and F_1 、 F_2 are the scores of the simplex parent domain and
 379 subdomain, respectively. Through local IDW interpolation, the simplex parent domain can be
 380 divided into several simplex subdomains, and the content of each simplex subdomain then
 381 comes from the parent domain, so the exploration cost is the cost of each simplex space domain
 382 containing the collection points to be measured. A relative measure of how much has been
 383 explored in this work.

384 The specific process employing the improved Bayesian optimization algorithm is as
 385 follows: the classification accuracy of the classification model is used as the objective function
 386 of the optimizer, constructing a simplex optimization domain from the hyperparameter space
 387 and creating a simplex work queue. Then, the surrogate model of the optimizer is constructed

388 by dividing the simplex optimization domain into independent local interpolations, according
 389 to IDW. The local interpolation with the highest acquisition function value is then obtained
 390 from the simplex work queue, and the objective function can be evaluated by this local
 391 interpolation. As the exploration information is updated, the parent interpolation is split into
 392 smaller and more accurate child interpolations, and then added to the simplex worklist. With a
 393 given number of iterations, when the model training meets the number of iterations or the
 394 optimal local interpolation gets the optimal hyperparameter combination.

395 3. Results and analysis

396 Building on the above, the average spectrum of 10 types of rock samples was obtained and then
 397 the characteristic wavelengths of each element were determined according to the NIST atomic
 398 spectrum database. It is found through experiments that the elements affecting lithology
 399 identification include Nb, Si, Al, Mg, Ca, Ti, Na, Ba, H, Li, Mn, Fe, etc. It is not easy to quickly
 400 select the analysis line as the identification and classification model under the principle of
 401 characteristic spectral line screening (which involves high spectral line intensity, high element
 402 transition probability, and no interference and overlap of other element spectral lines around
 403 the spectral line). Typical LIBS spectra of rock samples and emission spectra of main elements
 404 in the LIBS spectra are shown in Fig. S1 and Table S1 respectively.



405
 406

Fig. S1. Typical LIBS spectrum of rock samples.

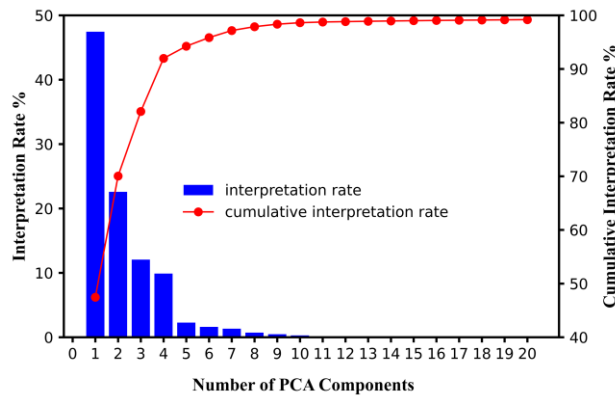
407

Table S1. Emission lines of main elements in the LIBS spectrum

Species	Wavelength (nm)	Species	Wavelength (nm)
Si	221.63, 243.46, 251.6, 263.12, 288.1, 385.56, 390.55, 634.75	Al	237.31, 309.28, 393.34, 394.39, 396.09
Fe	385.95, 438.32	Ca	396.8, 422.67, 616.33, 649.76
Na	261.18, 288.1, 589.11,	Ni	308.16
Mn	279.49	Nb	212.37, 260.06
H	656.35	Li	256.23, 670.84
Mg	279.55, 350.07	Ba	273.93, 614.25
O	240.62, 373.68, 404.6	Cr	275.52, 383.42

408 3.1 Principal Component Analysis

409 In this experiment, the PCA has been used to extract features from the spectral data, and the
 410 pre-processed spectral data are used as the input of PCA for dimensionality reduction
 411 processing, to achieve rapid convergence of the classification model, on the premise of
 412 retaining most of the original spectral information. Fig. 8 shows the interpretation rate and
 413 cumulative interpretation rate of the first 20 principal components, and Fig. 9 shows the two-
 414 dimensional scatter plot of the 10 types of rocks when the first two principal components were
 415 selected.

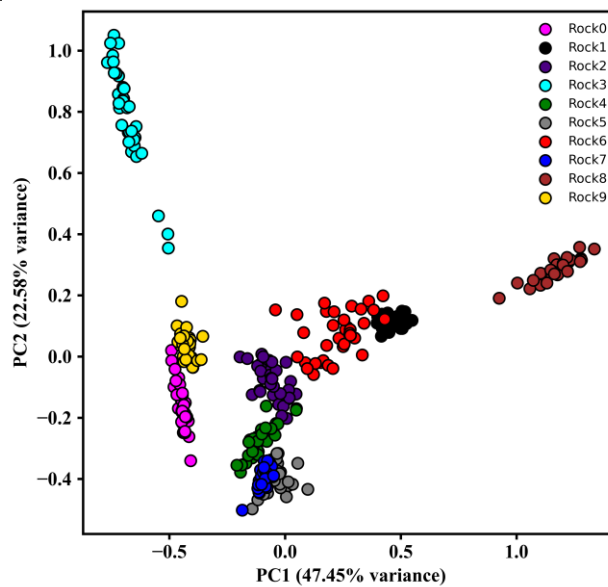


416

417

Fig. 8. PCA analysis results of LIBS data for 10 types of rocks.

418 By observing Fig. 8 above, it can be seen that the cumulative interpretation rate of the first
 419 four principal components has the fastest growth rate, and the cumulative interpretation rate
 420 reaches 91.96%; when the first ten principal components were taken, where the cumulative
 421 interpretation rate reaches 98.62%, and the cumulative interpretation rate of the principal
 422 components rate growth is extremely slow.

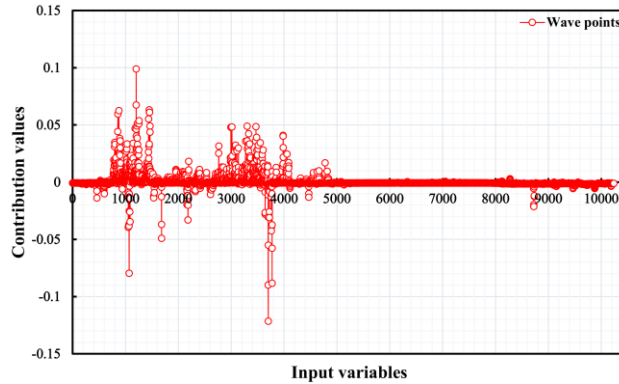


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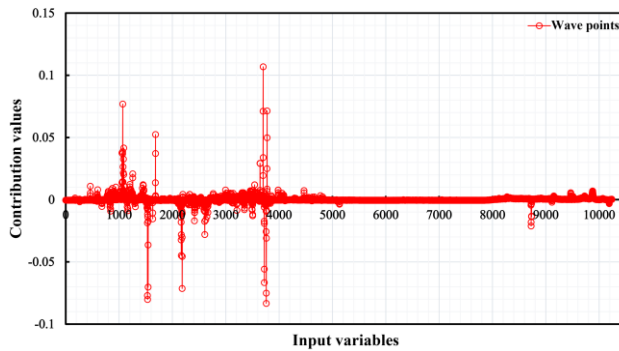
424

Fig.9. Two-dimensional scatter plot of the first two principal components.

425 By observing Fig. 9 above, it can be seen that when the first two principal components
 426 were selected as features after dimensionality reduction by using the PCA, it is found that ten
 427 types of rock data points appear to show a regional clustering phenomenon, among which
 428 Rock2 and Rock4, Rock1 and Rock6 have very little information which is overlapping in nature,
 429 but most of the information on Rock5 and Rock7 overlap, where most of the sample intervals
 430 are small. This is because the cumulative explanation rate of the first two principal components
 431 is only 70.03%, and it is necessary to increase the number of principal components and use the
 432 classification algorithm to further analyse the data after feature extraction has taken place.



433 Fig. 10. The loadings of PC1.



434 Fig. 11. The loadings of PC2.

435 **Table 3. Contributions of the elements to the PC**

Contribution values	Si	Fe	Al	Ca	Nb	Mn	Na	C	Li	Others
PC1	0.183	0.264	0.045	0.152	0.068	0.081	0.049	0.098	0.015	0.045
PC2	0.151	0.166	0.019	0.122	0.015	0.025	0.111	0.007	0.036	0.348

438 By calculating the loads of PC1 and PC2, the contribution values of the input variables
 439 (10,239 wave points) to the first two principal components were obtained. Following that and
 440 with reference to the NIST atomic spectrum database, the contribution value of the main
 441 elements in the rock to the PC was obtained, where the loads on PC1 and PC2 are, respectively,
 442 as shown in Fig. 10 and Fig. 11 above, and the contribution of elements to the PC is as shown
 443 in Table 3. It can be seen from Table 3 that Si, Fe, Al, Ca, Nb, Mn, Na, C and Li are the key
 444 elements of the first two principal components. Among them, Si, Fe, Al, Ca, Nb, Mn and C
 445 provide the major contributions to PC1. When the number of principal components is increased,
 446 the contribution value of Na, and Li and other elements to PC2 increases, and this process adds

447 new important features. Therefore, the appropriate number of principal components should be
 448 selected as the input to the classification model.

449 3.2 Classifier Model Optimization

450 The LIBS spectral data of 10 types of rocks can be seen as basic pre-processing operations and
 451 PCA dimensionality reduction with different numbers of principal components is used as the
 452 input of the classification model – and there are 500 sets of spectral data used in total. Among
 453 them, there are 350 groups in the training set and 150 groups in the test set, and the proportion
 454 of each type of rock in both the training set and the test set is equal. The classification accuracy
 455 R_{ACC} was used as the evaluation index of the classification model, the model was evaluated
 456 with ten-fold cross-validation, and the confusion matrix was selected for the final evaluation of
 457 the classification effect of the test set. The classification accuracy formula is given by:

$$458 \quad R_{ACC} = \frac{\sum_{i=1}^{10} T_i}{\left(\sum_{i=1}^{10} T_i + \sum_{i=1}^{10} F_i \right)} \quad (18)$$

459 In the above formula, T_i is the total number of correct classification⁴⁰ of class i rocks,
 460 F_i is the total number of incorrect classifications of class i rocks, $\sum_{i=1}^{10} T_i$ represents the
 461 total number of correct classifications of 10 types of rocks, and $\sum_{i=1}^{10} F_i$ represents the total
 462 number of incorrect classifications of 10 types of rock samples.

463 The SVM in this experiment can be implemented, based on the sklearn library in Python
 464 3.9.7. The classification accuracy was used as the objective function of GS and RS, and the
 465 SVM model was optimized by adjusting the hyperparameter space and the number of iterations.
 466 Table 4 shows the 10-fold average classification and recognition accuracy of the sub-test set
 467 when different numbers of principal components were selected as the input variables of the
 468 SVM classification model, and the training set was subjected to ten-fold cross-validation as the
 469 sub-training set and the sub-test set, respectively.

470

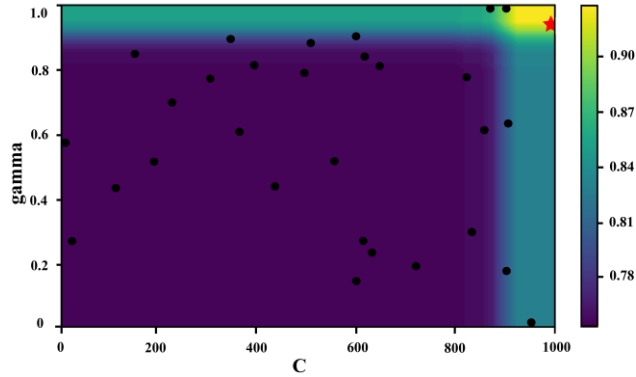
Table 4. SVM recognition results with different input variables

Numbers of variables	Cumulative interpretation rate /%	Average accuracy rate /%
1	47.45%	55.14%
2	70.03%	74.57%
3	82.08%	79.14%
5	94.23%	83.31%
10	98.61%	87.00%
15	98.99%	86.20%

471 It can be seen from Table 4 above that with the increase of the principal component score,
 472 the cumulative interpretation rate is increasing, and the average classification accuracy of the
 473 SVM classification model is also improving. When ten principal components were selected as
 474 the input of the classification model, the average classification accuracy of the SVM reaches
 475 87.00%. When the principal component scores continue to increase, the average classification
 476 accuracy of the classification model was seen to decrease. This arose because with the increase
 477 of principal component fraction, the increase of noise and dimension will reduce the
 478 classification accuracy.

479 Through the comparative analysis in Table 4, the top 10 principal components are selected
 480 as the inputs of GSCV-RSCV-SVM and GP-BO-SVM models. Firstly, the range of penalty
 481 factor C in SVM is [0.01, 0.1, 1, 10, 100, 1000, 10000], and the range of parameter gamma was
 482 [0.001, 0.01, 0.1, 1, 10, 100]. Through grid search cross validation, the best parameter
 483 combination $C=1000$, and $\text{gamma}=0.1$ is obtained. At this time, the average accuracy of cross

484 validation of the classification model is 87.20%. Then set $10 < C < 1000$, $0.01 < \text{gamma} < 1$, were
485 set, and through 30 iterations of random search cross validation, the optimal parameters of SVM
486 were found to be $C=743$, $\text{gamma}=0.38$. At this time, the average accuracy of the cross
487 validation set of the classification model was 87.71%.

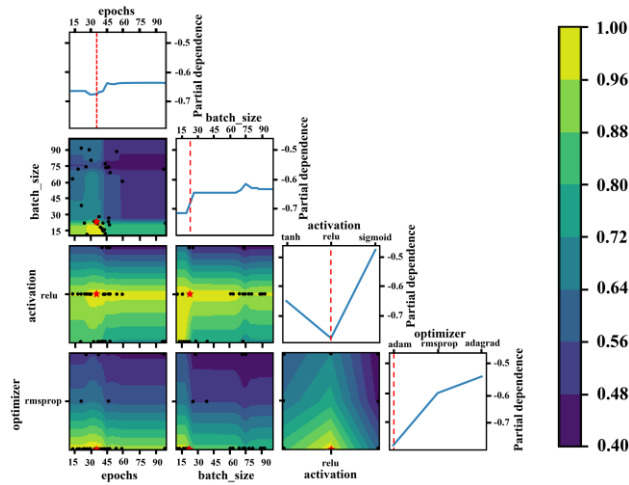


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Fig. 12. BO-GP-SVM optimal parameters optimization process.

490 To further improve the classification efficiency of SVM, Bayesian optimization based on
491 the Gaussian process was used to optimize the hyperparameters of SVM. Following that, it was
492 necessary to establish the hyperparameter C and gamma two-dimensional hyperparameter
493 space. As shown in Fig. 12 above, the best parameter combinations $C=990.101$ and
494 $\text{gamma}=0.919$ were obtained after 30 iterations. At this time, the average accuracy of cross
495 validation of the classification model was 90.20%. Although BOGP-SVM has improved the
496 classification accuracy compared with GS-RS-SVM, the modelling times of the two models
497 were 300.611s and 298.023s respectively, so the model efficiency of the two methods was not
498 high.

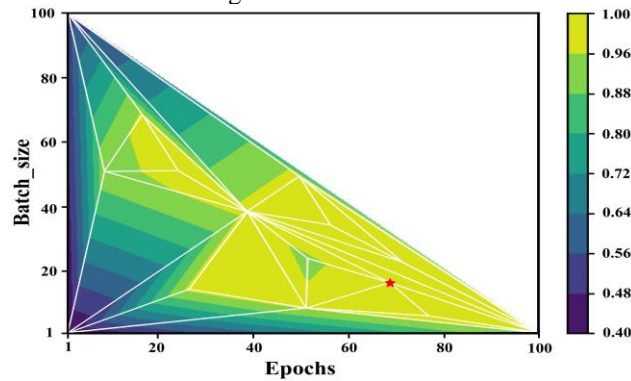
499 Next, the Bayesian optimization based on the Gaussian process was used to optimize the
500 hyperparameters of 1DCNN, and thus construct a four-dimensional parameter space for the
501 convolutional layer activation function, optimizer, batch size, and number of model training
502 rounds through the Bayesian optimizer. The process searches within the parameter space serve
503 as a surrogate model. The blue area in the figure is the area with a 'poor model' effect, the
504 yellow area is the 'better area', and the black point is the sampling position of the Bayesian
505 optimizer. The Bayesian optimizer fitted the model by using the prior probability at the higher
506 sampling density. parameters and the red five-pointed star was obtained as the best
507 hyperparameter combination position. The optimal hyperparameter combination (pre-
508 processing) was as follows: the activation function of the convolutional layer was Relu, the
509 optimizer was Adam, the batch size was 23, and the number of model training rounds was 35.
510 At this time, the average cross-validation accuracy of the classification model was 98%. The
511 hyperparameter optimization process is shown in Fig. 13 below.



512
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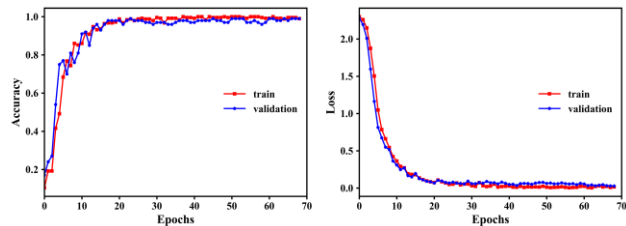
Fig. 13. BO-GP-1DCNN optimal parameters optimization process.

514 As shown in Fig. 14 below, a Bayesian optimizer was constructed, based on a simplex
 515 optimization domain to determine a two-dimensional hyperparameter space for the batch size
 516 and the number of model training epochs in the 1DCNN classification model, where the
 517 $\text{Batch_size} \in [1,100]$, $\text{Epoch} \in [1,100]$. Then the simplex optimization domain was
 518 constructed according to the hyperparameter space, and the optimal hyperparameter
 519 combination was obtained through 15 iterations. Here the batch size was 17, and the number of
 520 model training rounds was 69. At this time, the average cross-validation accuracy of the
 521 classification model was 99.33%, and the modelling time was 103s. The hyperparameter
 522 optimization process is as shown in Fig. 15 below.



523
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Fig. 14. Improved BO-1DCNN optimal parameters optimization process.



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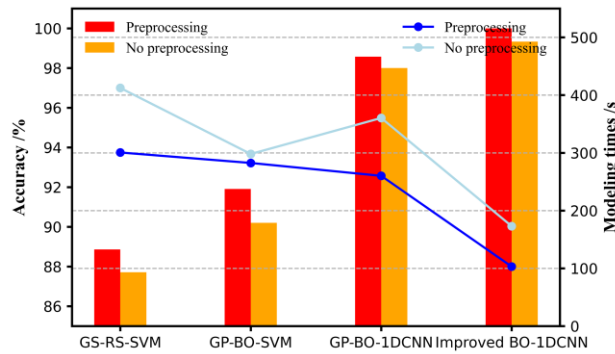
Fig. 15. Improved BO-1DCNN model training process.

527 By examining the training process of the Improved BO-1DCNN model in Fig. S2, it can
 528 be seen that when the network was trained through ~20 rounds, the cross-entropy loss

529 difference of the network tends to be stable, and the network accuracy at this time was 0.001.
 530 Although the actual set network error value was met at this time, the training set and the
 531 classification accuracies of the validation set were 97.2% and 98%, respectively, falling into a
 532 local optimum. The global search was carried out through the improved Bayesian optimizer.
 533 When the network was trained to 69 rounds, the network accuracy is 0.00098, which is the
 534 global optimal value. The confusion matrix for the classification and recognition of 10 types of
 535 rocks in the test set after model optimization is shown in Fig. S2. It can be seen that the
 536 Improved BO-1DCNN model can recognize and classify 10 types of rocks. The accuracy rate
 537 can reach 99.33%, and the model recognition time was significantly reduced, compared to the
 538 first three models, which can be used to quickly identify and classify 10 types of rock samples.

539 3.3 Comparative and analysis

540 To further verify the ability of the Improved BO-1DCNN model to automatically classify rocks,
 541 the four classification models are compared with the basic pre-processing and optimally pre-
 542 processing rock LIBS spectral data. Fig. 16 below shows the validation set accuracy and
 543 modelling time of the four methods obtained. Table 5 is the validation set efficiency comparison
 544 of the four models. (1) in Table 5 is pre-processing, and (2) is no pre-processing. It can be seen
 545 that SVM has lower classification accuracy than 1DCNN. This is because SVM performs
 546 classification and recognition by mapping the input to a high-dimensional space. Moreover,
 547 when the SVM input variable dimension is too high, the efficiency of classification recognition
 548 and hyperparameter optimization was low. The 1DCNN adopted a convolution calculation and
 549 deep architecture and achieves high-accuracy classification by extracting spectral features to
 550 form high-level semantic category information.



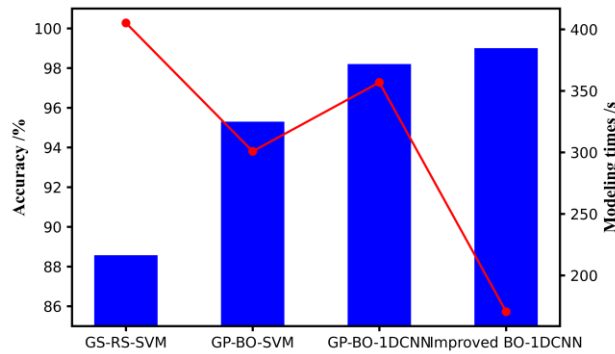
551
 552 Fig. 16. The validation set accuracy and modelling time of the four methods.

553

Table 5. Comparison of the validation set efficiency of the four models

Models	Accuracy /%	Modelling times /s
GS-RS-SVM (1)	88.86	300.611
GS-RS-SVM (2)	87.71	412.391
GP-BO-SVM (1)	91.91	282.493
GP-BO-SVM (2)	90.20	298.023
GP-BO-1DCNN (1)	98.57	260.145
GP-BO-1DCNN (2)	98.00	360.381
Improved BO-1DCNN (1)	99.46	103.858
Improved BO-1DCNN (2)	99.33	173.299

554 By comparing the classification accuracy and modelling time of different classification
 555 models, it can be seen that the optimization efficiency of the three optimizers follows the
 556 following pattern: GS-RS<GP-BO<Improved BO. By comparing the basic pre-processing
 557 experiment and the optimal pre-processing experiment, it has been found that although the
 558 classification accuracy of the optimal pre-processing experiment was higher, the modelling
 559 time also increased, and a large number of experiments were found necessary to obtain the
 560 optimal pre-processing combination. Fig. 17 below shows the test set accuracy and modelling
 561 time of the four methods. Table 6 is the test set efficiency comparison of the four models.
 562 Therefore, compared with other methods, the Improved BO-1DCNN model proposed in this
 563 study not only needed no optimal pre-processing, but also has the highest classification
 564 efficiency, used to realize LIBS for the automatic and rapid classification of rock samples.



565
566
567 Fig. 17. The test set accuracy and modelling time of the four methods.

Table 6. Comparison of the test set efficiency of the four models

Models	Accuracy /%	Modelling times /s
GS-RS-SVM	88.57	405.231
GP-BO-SVM	95.30	300.824
GP-BO-1DCNN	98.20	356.912
Improved BO-1DCNN	99.00	170.521

568 **4. Conclusions**

569 In this paper, an improved Bayesian optimization algorithm has been put forward and proposed
 570 – then it has been applied to create an identification method combining laser-induced
 571 breakdown spectroscopy with a one-dimensional convolution neural network. It has been used
 572 and evaluated for the classification of 10 different types of rocks, such as siltstone, oil shale,
 573 mudstone, argillaceous siltstone, red sandstone, coarse sandstone, fine sandstone, etc., to solve
 574 the shortcomings of traditional Bayesian algorithms, such as global dynamic optimization and
 575 low efficiency. By selecting different classifiers and optimizers to build classification models,
 576 including GS-RS-SVM、GP-BO-SVM、GP-BO-1DCNN and improved BO-1DCNN,
 577 comparative experiments were carried out on 10 types of rock spectral data that were pre-
 578 processed and unprocessed respectively. The experimental results obtained from that work
 579 were then able to show that LIBS technology, combined with the 1DCNN classification model,
 580 does not need to select the optimal pre-processing combination, and the improved Bayesian
 581 optimization algorithm can achieve high accuracy and fast classification between rocks.
 582 Through global dynamic optimization, the modelling time of the improved Bo algorithm has
 583 been seen to be greatly reduced, and the classification accuracy obtained was higher after
 584 optimization, which improves the classification efficiency of rocks.

585 **5. Back matter**

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592 *5.3 Disclosures*

593 The authors declare no conflicts of interest.

594 *5.4 Data availability statement*

595 **Data availability.** Data underlying the results presented in this paper are not publicly available
596 at this time but may be obtained from the authors upon reasonable request.

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