

CALIBRATION TRANSFER FOR ChemCam SPECTRAL DATA FROM DIFFERENT LASER-INDUCED BREAKDOWN SPECTROMETERS VIA A DEEP EXTREME LEARNING MACHINE**

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Laser-induced breakdown spectroscopy (LIBS) technology has been applied to many fields, so it is crucial for quantitative analyses of LIBS spectra. However, there is a problem in the field of LIBS spectra. Even in the same experimental setting, the same sample exhibits different spectra with different instruments, which is mainly reflected in the intensity, wavelength shift, and peak width differences. These differences cause standardization problems in LIBS spectroscopy and serious interference in quantitative analyses. The aim of this study is to correct the difference by applying the extreme learning machine method and the deep extreme learning machine method to two different spectral datasets. The first dataset is the ChemCam calibration target sample set, which contains two spectral datasets produced by using ChemCam on instruments on the Curiosity rover and at the Mars Science Laboratory (MSL). The other dataset comprises spectra obtained from calibration target samples produced by the ChemCam team at MSL. The performance of the two algorithms is tested, and the results show that our calibration transfer methods are stable predictive methods that provide significantly lower prediction error compared with linear transfer and the piecewise direct standardization method. The model established by the partial least square method is used for quantitative analyses of the transferred spectra, and the transmitted spectra showed improved quantitative accuracy.

Keywords: laser-induced breakdown spectroscopy, ChemCam calibration target, piecewise direct standardization, extreme learning machine, deep extreme learning machine.

КАЛИБРОВКА СПЕКТРАЛЬНЫХ ДАННЫХ ChemCam РАЗЛИЧНЫХ ЛАЗЕРНО-ЭМИССИОННЫХ СПЕКТРОМЕТРОВ С ИСПОЛЬЗОВАНИЕМ МАШИНЫ ГЛУБОКОГО ЭКСТРЕМАЛЬНОГО ОБУЧЕНИЯ

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Для устранения различий в интенсивности, смещении длины волны и ширины максимумов, возникающих при измерении лазерно-искровых эмиссионных спектров (LIBS), использованы метод экстремального обучения и машины глубокого экстремального обучения к двум различным наборам спектральных данных. Первый представляет собой набор образцов калибровочной мишени

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ChemCam, который содержит спектральные данные, полученные с помощью ChemCam на марсоходе Curiosity и в Марсианской научной лаборатории (MSL). Другой набор содержит спектры образцов калибровочной мишени, созданных командой ChemCam в MSL. Проверка производительности двух алгоритмов показала, что методы переноса калибровки являются стабильными методами прогнозирования и обеспечивают значительно меньшую ошибку прогнозирования по сравнению с линейным переносом и методом кусочно-прямой стандартизации. Модель на основе метода участичных наименьших квадратов для количественного анализа переданных спектров показала улучшенную количественную точность.

Ключевые слова: лазерно-искровая эмиссионная спектроскопия, калибровочная мишень ChemCam, кусочно-прямая стандартизация, экстремальное обучение, машина глубокого экстремального обучения.

Introduction. Laser-induced breakdown spectroscopy (LIBS) uses an intense laser pulse to ablate a small amount of material from a target of interest, whereby the emission spectrum is collected from the resulting plasma [1, 2]. This spectrum can then be used to study the composition of the target [3]. The ChemCam instrument package on the Curiosity rover [4] (launched in 2012) and the SuperCam [5] instrument on the Spirit rover (launched in 2020), as well as the China Mars 2020 rover with the MarsCoDe [6] (Mars Surface Composition Detection Package) instrument (launched in 2020), all use LIBS technology to identify the compositions of rock and soil targets on Mars.

A LIBS spectral library was developed using a laboratory LIBS instrument, Mars-like atmospheric conditions, and enough standard samples before quantifying the spectra produced by the Mars targets [7]. Ideally, a single quantitative analysis model should be used for both Mars and laboratory datasets, considering the similarities in experimental and instrumental parameters. However, there are differences in the spectra of replicate calibration samples obtained using the Earth and Mars instruments – for example, the spectrum of the shergottite sample on Mars and the spectrum of its replica collected in the laboratory are shown in Fig. 1, which shows how different these two spectra are. It should be pointed out that the synthetic shergottite sample in the Mars dataset is one of the preflight calibration targets that were put on board the rover before it left Earth [8]. The discrepancies between Earth and Mars spectra are clearly visible, and the Mars emission lines are on average more than twice as intense as the Earth emission lines. These differences are caused by the laser energy, inconsistencies in the distance between the instrument and the sample, and the difference between the actual Martian environment and the simulated environment [7].

For LIBS performed in the Martian environment, the linear correction method developed by Clegg et al. is commonly used to solve this problem [7]. The calibration targets on Mars have been analyzed with various laser powers, which causes variations in the spectra. To develop a consistent correction, only calibration target spectra collected at the maximum energy of 14 mJ/pulse were averaged together for the Mars and laboratory instruments, resulting in six Mars and six laboratory calibration target spectra [7]. By averaging the ratios of the six spectra, a single vector containing per-pixel correction factors was obtained; however, this correction method is not very satisfactory over the wavelength range of interest [9].

In the field of spectroscopy, several calibration transfer methods have been proposed in the past decade to solve this problem. Generally, two methods are used. First, each spectrum (instrument) of the whole dataset is resampled to establish a new calibration model. The disadvantage of this method is that the recalibration process is both costly and time-consuming. Second, environmental or experimental differences are corrected through a series of linear or nonlinear correction methods, and then the calibration model is used to identify the target component of the unknown sample [10]. Compared with the first method, this method has the advantage of avoiding the reconstruction of the calibration models. These methods are called calibration transfer in the field of spectroscopy and stoichiometry and transfer learning in the field of machine learning and data mining [11–13]. Due to differences in intensity, wavelength shift and peak width between the secondary instrument and the primary instrument, we assume that the spectra resulting from these two instruments have a nonlinear dependence. To find the relationship between them, two machine learning methods, extreme learning machine (ELM) [14] and deep extreme learning machine (DELIM) [15], are applied to transmit the LIBS datasets produced by two different instruments. Existing transfer methods are mostly used in transferring infrared spectra. When faced with the transfer problem in LIBS spectra, the existing solutions are difficult to use due to high dimensionality, multiple spectral peaks, and interaction between peaks of LIBS spectra.

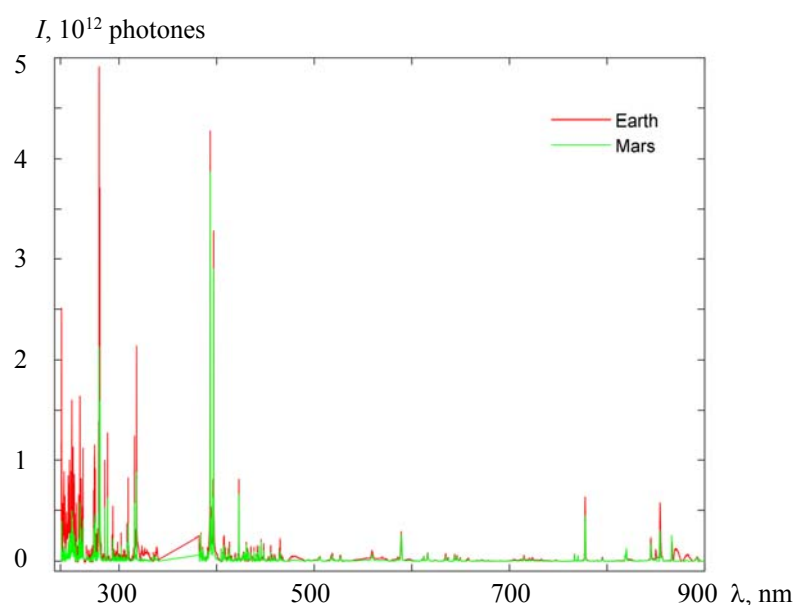


Fig. 1. LIBS spectra of synthetic shergottite glass from the Mars and Earth datasets.

Related work. As mentioned in the preceding, many calibration transfer methods based on standard samples have been reported. Slope and deviation correction (SBC) [16] is an early method used for calibrating transfers between different spectrometers and assumes a strong linear relationship between them. First, for each sample in the standard sample set, the value of the target component is predicted by feeding the spectra from the primary and secondary spectrometers into the original calibration model that is established on these spectrometers. Second, a first-order linear model is established for the two predictions, and slope and deviation parameters are estimated based on the least square methods. In general, the SBC approach works well, and the approach is simple. For those cases in which differences are more complex, direct standardization (DS) [17] and piecewise direct standardization (PDS) [18] methods are used to promote the performance in calibration transfer. For a given sample using the DS method, it is assumed that spectra collected from the spectrometer can be linearly transformed to the corresponding spectra obtained using the primary spectrometer. Based on the standard sample, the linear transformation matrix F can be calculated by the least square algorithm. Once the F value is calculated, the spectrum of a new sample can be projected from the secondary spectrometer to the primary spectrometer. Although PDS is widely used, the real relationships between spectra may be nonlinear in many practical applications. In other words, according to the properties of linear systems, the solution of F may be overfit because the system is underdetermined. To overcome this shortcoming, the PDS method was proposed. The basic idea is to reduce the number of wavelengths by dividing the whole wavelength range into several subintervals. The spectrum of the standard sample scanned at wavelength j on the primary spectrometer is mapped to the wavelength of the small window (subinterval) around wavelength j on the secondary spectrometer. The transformation matrices calculated from each window are spliced into diagonal matrix F . Previous research showed that the PDS method is superior to the DS method in many applications. In addition to methods based on standard samples, several nonstandard calibration transfer methods have been proposed – for example, Liang [19] et al. studied the relationships between spectra collected on different instruments, improved the PDS method, and proposed a new multivariate calibration transfer method. An optimal sample selection method (Rank-KS) was proposed to select the transfer samples. The process of sample selection was based simultaneously on the distributions of spectral space and property space. Chen [12] et al. proposed a new method based on a transfer learning algorithm that combines principal component analysis (PCA), a weighted ELM, and the TrAdaBoost algorithm. The experimental results showed that while objects of the source and target domains were the same, regardless of calibration transfer between different instruments or components, the generalization performance of the target domain model improved greatly, especially when the target domain contained few samples.

Methodology. *Datasets.* Two datasets were used in this study. The spectral datasets are available for downloading from the NASA Planetary Data System [20]. The first is the ChemCam Calibration Target (CCCT, the onboard Curiosity rover calibration target for LIBS) dataset, which consists of the calibration

plates carried by Curiosity. The images are labeled with the names of samples from each disk [21]. Standard sample spectra were obtained at the Mars Science Laboratory on Earth and by the Curiosity rover, and they are called the Earth dataset and the Mars dataset, respectively. The Earth dataset was obtained on Earth under simulated Martian conditions using a ChemCam laboratory clone, whereas the Mars dataset was obtained from the Curiosity rover currently on Mars. Most of the spectra in the Mars dataset for CCCT samples were obtained during the first 450 sols on Mars. It should be noted that all spectral data are from level-1B data on the PDS website; that is, they have been preprocessed. The preprocessed spectra from both the Mars and Earth datasets were preprocessed in the same way using the techniques outlined by Wiens et al. [22]. These spectra were corrected by removing the nonlaser-induced background (“dark subtract”), denoising the spectrum, removing the electron continuum, and calibrating for wavelength and instrument response. Dark signal removal consisted of collecting 50 spectra with the laser turned off but under the same conditions as the experiment. Then, the average spectrum was subtracted from each spectrum recorded with the laser. Wavelet analysis was used for denoising and electron continuum removal. Wavelength calibration was carried out using titanium lines in LIBS spectra obtained by titanium plate ablation. The instrument response was the product of spectral radiance, source image area, solid angle, detector wavelength box width, integration time, and amplification conversion gain.

Detailed descriptions of the samples and experimental setup are given by Clegg et al. [7]. The physical forms of the samples were powder, glass, and ceramic. The powders were ground and pressed into pellets using approximately 3–5 g of sample. Each prepared sample was then placed in a vacuum chamber filled with CO₂ at 9.3 mbar pressure to simulate Mars atmospheric pressure. The maximum ChemCam pulse energy is limited to 14 mJ/pulse or less. The laser beam was projected onto the targets at a distance of 1.56 m through a telescope with a diameter of 110 mm, which also collected the returning plasma emission [8]. The LIBS spectra were then obtained from ablation at five different spots on each sample, and every spot was ablated 50 times. Each sample was probed at five different locations, each with 50 laser shots, to ensure that the average composition of the sample was represented.

Since there could not be too many samples on the calibration plate, the CCCT dataset was small. To better verify the robustness and adaptability of our calibration transfer method, we selected another dataset containing the spectra of 56 samples measured with different LIBS spectrometers. The dataset, also from the Mars Science Laboratory, consisted of primary instrument spectra from 408 standard samples measured by the ChemCam team in 2015 [7] and secondary instrument spectra from 69 standard samples measured by the ChemCam team in 2010. To enhance consistency, we selected 56 identical samples from the two datasets and formed a transfer sample set called the standards dataset.

Then, we established a PLS2 quantitative regression model based on the primary instrument dataset. For the CCCT dataset, we took the Mars rover instrument as the primary instrument and transmitted the data of the Earth spectrum to the Mars spectrum. For the standards dataset, we used the instrument that produced the 2015 spectral data as the primary equipment and transferred the 2010 spectra. During the establishment of the PLS2 quantitative analysis model, the samples in the transferred dataset should be eliminated from the training dataset, and the transferred sample dataset should be taken as the test set. Therefore, for the CCCT dataset, the selected CCCT samples were used as the transfer set, and after removing 7 CCCT samples from 408 samples, the remaining 401 samples were used as the training set to establish the PLS2 quantitative analysis model. For the standards dataset, the spectral set generated by the main instrument was taken as the primary dataset with which to establish the quantitative model, the transfer set samples were also removed, 56 samples were removed, and the remaining 352 samples were used to establish the PLS2 quantitative analysis model.

Extreme learning machine. An ELM, which was proposed by Professor Huang in 2010, is essentially a single hidden layer feed-forward network (SLFN) [23, 24]. The ELM has both universal approximation and classification capabilities with significant advantages such as fast learning speed, ease of implementation, and minimal human intervention. It also has a general and efficient approximation ability, and its structure is shown in Fig. 2. Unlike backpropagation or conjugate gradient descent training algorithms, ELM theory shows that hidden nodes of generalized feedforward networks are important but do not need to be adjusted, and these hidden nodes can be generated randomly [25, 26]. All hidden node parameters are independent of the objective function or the training dataset. The input data are mapped to an L-dimensional ELM random feature space [27]. In our study, the number of hidden layer nodes used in the ELM was 800. Given a supervised learning problem containing N different data samples, the training process consists of two stages: according to the number of nodes in the hidden layer, the input weight matrix W and bias b_x are generated ran-

domly to calculate the output of the hidden layer, and the output matrix β is obtained by solving the data label, and its model is

$$\begin{cases} H_x = g(W_x x + b_x) \in R_l, \\ Y = \beta h_x \in R^s, \end{cases}$$

where $g(\dots)$ is the activation function (common activation functions include the sigmoid and tanh functions); H_x is the output of the hidden layer; and $\beta = [\beta_1, \beta_2, \dots, \beta_l]$ is the output layer weight. The sigmoid function was adopted in this study: $\text{sigmoid}(x) = 1/[1 + e^{-x}]$.

The output weight β was calculated by the Moore-Penrose generalized inverse matrix of H and $Y = (y_1, y_2, \dots, y_n)$:

$$\beta = H^+ Y.$$

To improve generalization performance and make the solution more robust, we can add a regularization term as shown:

$$\beta = (I/C + H^T H)^{-1} H^T Y.$$

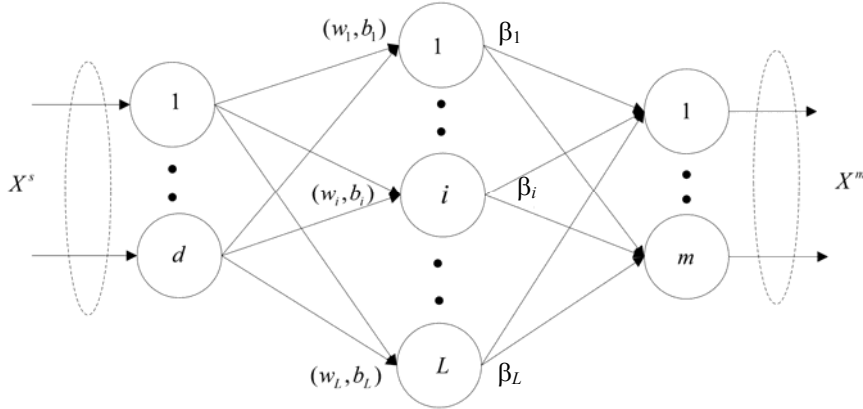


Fig. 2. Network structure for the extreme learning machine.

(2)

Deep extreme learning machine. DELM is a deep network structure generated by combining the ideas of autocoding with ELM [15], and its structure is shown in Fig. 3. DELM integrates the learning efficiency of ELM with the deep structure of the autoencoder (AE) to achieve better prediction performance. Since the classical ELM algorithm is a supervised learning algorithm and in the deep structure of DELM, the data have no label matrix in the dimension-reduction process, ELM is combined with the autoencoder to approximate the original input matrix by minimizing reconstruction error. The structure of DELM is divided into two parts: unsupervised feature learning and monitoring feature regression. In the first part, the structure of ELM-AE is adopted to obtain a compressed representation of the data layer by layer. First, the input data x is mapped to the hidden layer H through the formula, where H is the output from the random mapping of the input layer, β is the output weight of the hidden layer, X is the input data, l_1 is the norm that reduces the number of hidden nodes to generate more compact data, and the parameters of the hidden layer can be obtained by solving the following mathematical model:

$$\min \|H\beta - X\|^2 + \|\beta\|_{l_1}. \quad (1)$$

In the second part, the classical ELM supervised learning algorithm is used for regression. The ELM algorithm makes full use of the label information of the samples and extracts the hidden information of the sample data after dimension reduction to improve the classification performance of the algorithm. The autoencoder structure here is different from that of the traditional deep autoencoder network. Once the features of the first hidden layer are extracted, the weight and parameters of the layer are fixed, and there is no need for fine-tuning through backpropagation as there is in the deep autoencoder network. Therefore, training is faster for DELM than it is for the backpropagation neural network (BPNN), deep autoencoder (DAE), and other backpropagation algorithms. We used a DELM model structure with three hidden layers, and the number of nodes in each hidden layer was 750, 500, and 750. There were activation functions that act on the outputs

of the hidden layers; the Sigmoid function was chosen as the activation function for each layer. In addition, we selected the number of nodes in each layer according to Eq. (1) to obtain more compact data and a better transfer effect.

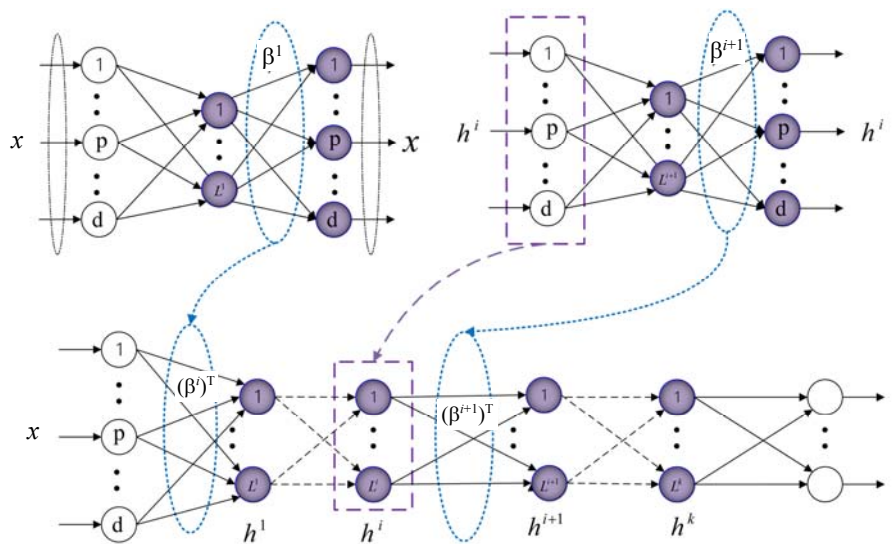


Fig. 3. Network structure for deep extreme learning machine with auto encoding.

Because of the DELM self-coding layer-by-layer dimensionality reduction training method, a better testing effect is often achieved compared with the single-layer ELM for complex high-dimensional data. DELM can be used to extract the compressed representation layer by layer from the original data without supervision and to make full use of the label information in the data when there is supervision classification in the last layer; this provides results better than those from ELM. Therefore, we used the DELM algorithm for the standards dataset with increasingly complex samples.

Results and discussion. Because the LIBS calibration targets must be carried onboard the rover to Mars, there are few samples on the calibration plate. To ensure the running speed and the size of the model, we used the single-layer ELM algorithm for the CCCT sample set, and the expected transfer effect was achieved. To realize LIBS spectral transmission in the field of generalization, we used a more complex sample and more rock sample sets to validate the effects of our method. For the sample set, the feature extraction capability of the single-hidden layer ELM algorithm is not sufficient, so we used the combination of a deep extreme learning machine and an automatic encoder for transfer. Then, the partial least squares algorithm was used to carry out quantitative regression of the principal components of the transmitted spectrum, and the optimal number of potential factors was determined by the 10-fold cross-validation method. The quantitative regression model was developed from sample spectra generated by the primary instrument, and then the performance was evaluated using the root mean square error of prediction (RMSEP) on the independent test set.

CCCT dataset. In Figure 4, we selected two spectral segments (440–490 and 520–650 nm) to show the detailed differences between the laboratory and Mars spectra before and after transfer. As seen from Fig. 4, the laboratory spectrum of the same sample was very different from the Mars spectrum; there were differences in intensity, spectral peak shape, and peak shift, and the differences were obviously nonlinear. After treatment with the ELM algorithm, these differences were well corrected.

In addition to overall comparisons between pretransfer and post-transfer laboratory spectra with Mars spectra, we also show a comparison of spectra for the shergottite standard sample obtained under Mars conditions as well as laboratory conditions with corrected spectra (Fig. 5). The differences between the actual and predicted spectra are shown in Fig. 5. Among them, the correction method used in Figs. 5a–c is the ELM algorithm proposed in this paper, and the correction method used in Figs. 5d–f is the algorithm using linear factors [7]. We can see that data corrected by the ELM algorithm (1) are more consistent with the Mars data (2). We can also see from the difference chart that the difference for corrected data (1) is much smaller than the difference for the uncorrected data (3).

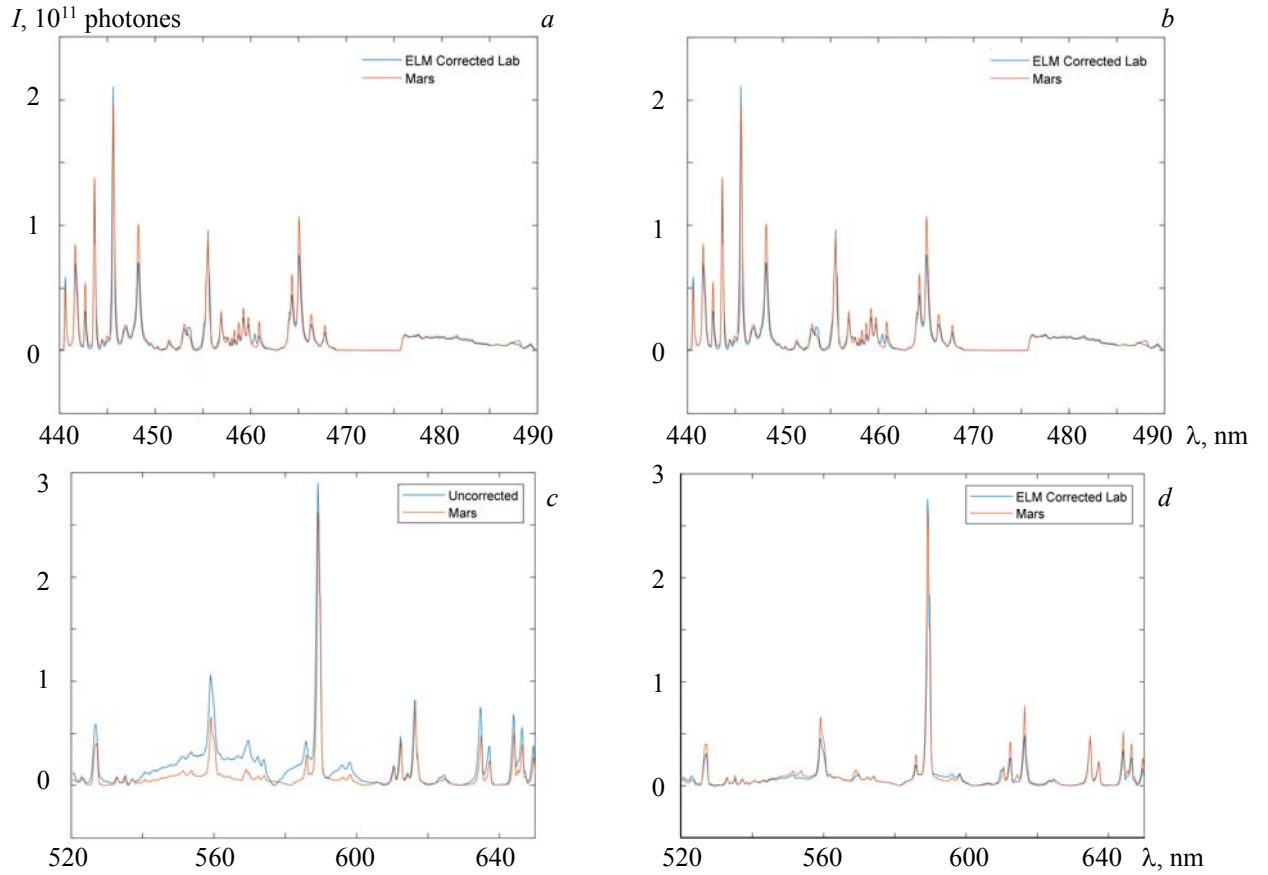


Fig. 4. Uncalibrated laboratory spectra of the shergottite samples at 440–490 and 520–650 nm compared to the spectra obtained from Curiosity on Mars (a,c) and laboratory spectra calibrated with the ELM algorithm compared to spectra obtained from Mars (b,d).

We compared the similarities between the spectra obtained from Mars to those obtained in the laboratory. To compare the correction effect and explain the intensity variation between the spectra being compared at each wavelength point, we introduce the concept of intensity difference. In this study, the intensity difference is equal to the sum of the intensity difference between the predicted data and the actual data at each wavelength point compared with the sum of the intensity of the actual spectrum. The formula is:

$$\text{Intensity error} = \frac{\sum(|\text{intensity}_{\text{pre}} - \text{intensity}_{\text{act}}|)}{\sum(\text{intensity}_{\text{act}})}.$$

Table 1 shows the differences between transmitted spectra and actual spectra for CCCT samples with different calibration transfer methods, including linear correction, the ELM algorithm used in this study, and the classic PDS algorithm frequently used with infrared spectra, as mentioned in section ‘Related work’. The results listed in Table 1 clearly show that the prediction error from using the ELM algorithm was much lower than that of either linear correction or the PDS algorithm and that the performance of the PDS algorithm was better than that of the linear correction. To show the effect more clearly, we made a test set for each sample and conducted a total of 7 experiments. The results for each sample are shown in Table 1. Table 2 shows the Pearson correlation coefficients [28] for the Mars spectrum and the corrected spectrum for each sample, and the ELM algorithm exhibited the best transfer effect. The Pearson correlation score finds the ratio between the covariance and the standard deviation for both objects. In mathematical form, it can be described as follows:

$$\text{Pearson}(x, y) = \frac{\sum xy - \frac{\sum x \sum y}{N}}{\sqrt{(\sum x^2 - (\sum x)^2 / N)(\sum y^2 - (\sum y)^2 / N)}}.$$

Standards dataset. For the standards dataset with more samples and more types, the network structure with a single hidden layer has difficulty achieving satisfactory transmission, so we used the deep extreme learning machine with an autoencoder. Table 3 shows the transfer performance of ELM and DELM with standard datasets, where 20, 30, and 40 standardized samples were considered to evaluate the impacts of the number of subset samples on different calibration methods. The results showed that more transferred samples resulted in a better transfer effect. DELM outperformed ELM regardless of the number of standardized samples.

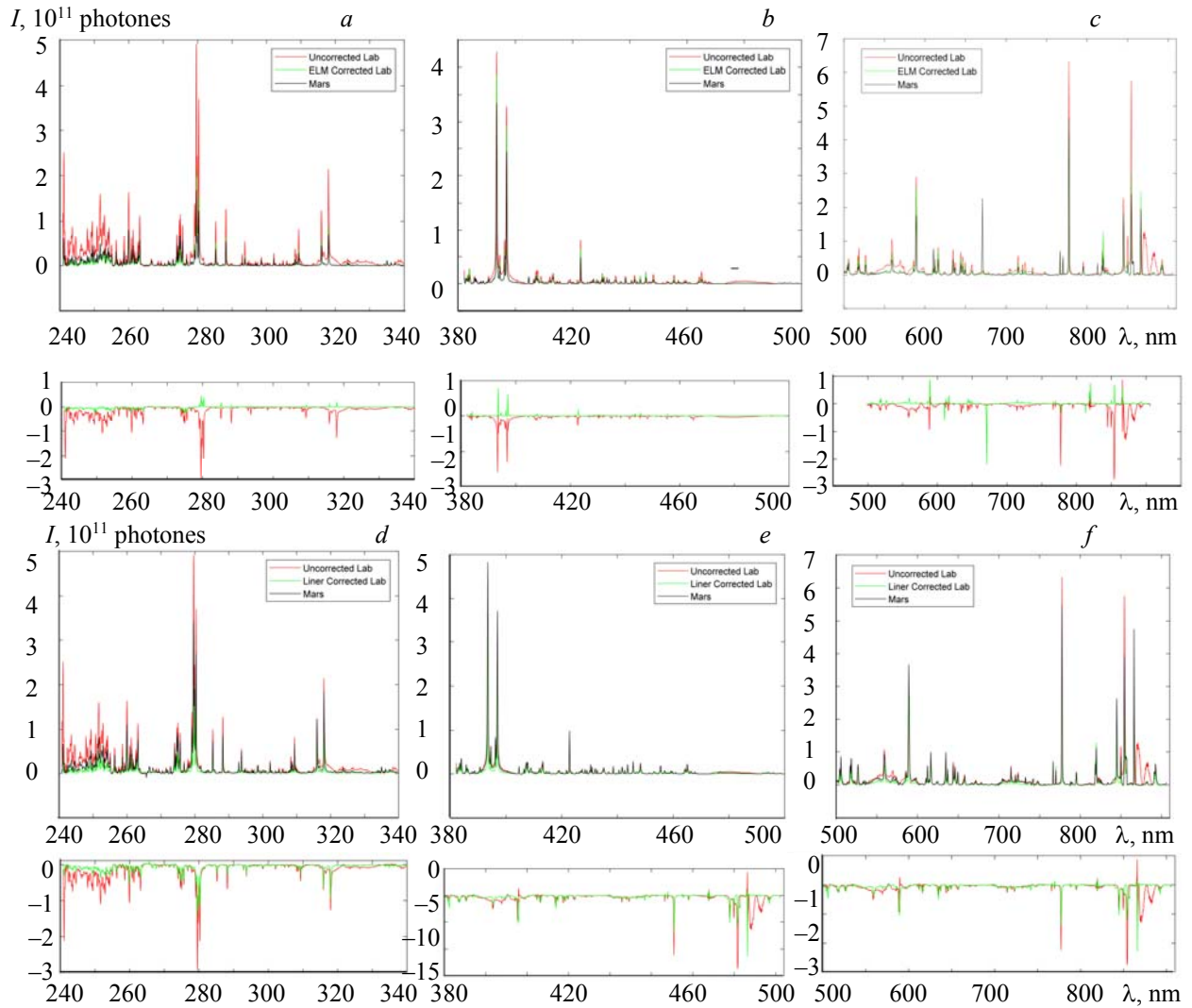


Fig. 5. The improvements using the ELM algorithm to alter the relationship between spectra for the sher-gottite calibration target on Mars and in the laboratory (a–c) and the linear correction performance (d–f)

TABLE 1. Intensity Errors for CCCT Spectra Datasets with Different Transfer Methods

CCCT	Linear correction, %	PDS, %	ELM, %
Norite	175.86	86.42	2.77
Picrite	149.31	38.19	2.32
Shergottite	101.56	93.31	6.46
KGa-2-Med-S	53.40	52.65	5.92
Nau-2Lo-S	162.89	63.16	9.09
Nau-2Med-S	86.76	45.65	1.68
Nau-2Hi-S	140.10	79.53	5.26

TABLE 2. Correlation Coefficients for CCCT Spectra Datasets with Different Transfer Methods

CCCT	Linear correction, %	PDS, %	ELM, %
Norite	0.8054	0.8706	0.9853
Picrite	0.8675	0.9223	0.9832
Shergottite	0.9085	0.9534	0.9885
KGa-2-Med-S	0.8565	0.9612	0.9833
Nau-2Lo-S	0.8974	0.8709	0.9864
Nau-2Med-S	0.8613	0.9057	0.9871
Nau-2Hi-S	0.8879	0.9412	0.9814

TABLE 3. Transfer Performance of ELM and DELM with Standard Datasets

Standardization samples	Standardization spectra	ELM, %	DELM, %
20	80	34.79	19.14
30	120	22.68	16.88
40	160	18.65	16.04

Figure 6 shows the correlation between the predicted total integrated intensities and actual total integrated intensities for all samples in the full spectral range 240–900 nm. All samples on the one-to-one correspondence line showed a high correlation between the actual and predicted values. We note that this result was obtained despite significant differences in the spectra of some samples due to heterogeneities and sampling differences (i.e., ablation site/location selection of these samples).

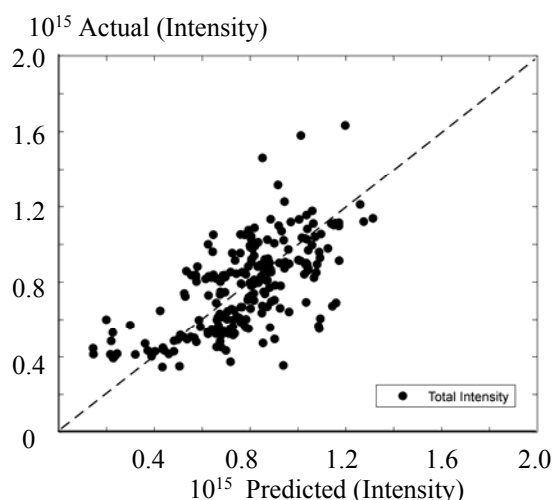


Fig. 6. Predicted total integrated peak intensities compared to actual integrated intensities over the actual full spectral range 240–900 nm for all samples.

In addition to comparing total integrated areas, we also compared spectral peak intensities of some characteristic elements of interest. The abscissa is the predicted intensity, and the ordinate is the true intensity. Four characteristic elements, Fe, Mg, Si, and Na, were selected, and comparisons of their spectral peak intensities are shown in Fig. 7. The ratio generally followed the 1:1 trendline. However, we noticed from this figure that sample points with high total integrated areas had higher degrees of dispersion. We believe that this is because there were certain differences between these samples and most training samples. ELM and DELM are supervised learning techniques used to predict test cases based on examples. Therefore, insufficient examples of the original spectrum may result in poor performance with the predicted spectrum – for example, in a spectral range where there are no similar examples in the training data, and the spectral accuracy after prediction processing may be poor. When the oxide concentration of the test sample exceeds the concentration range of the training set, the predicted spectra may not be accurate because the spectrum of the oxide was not considered when generating the model.

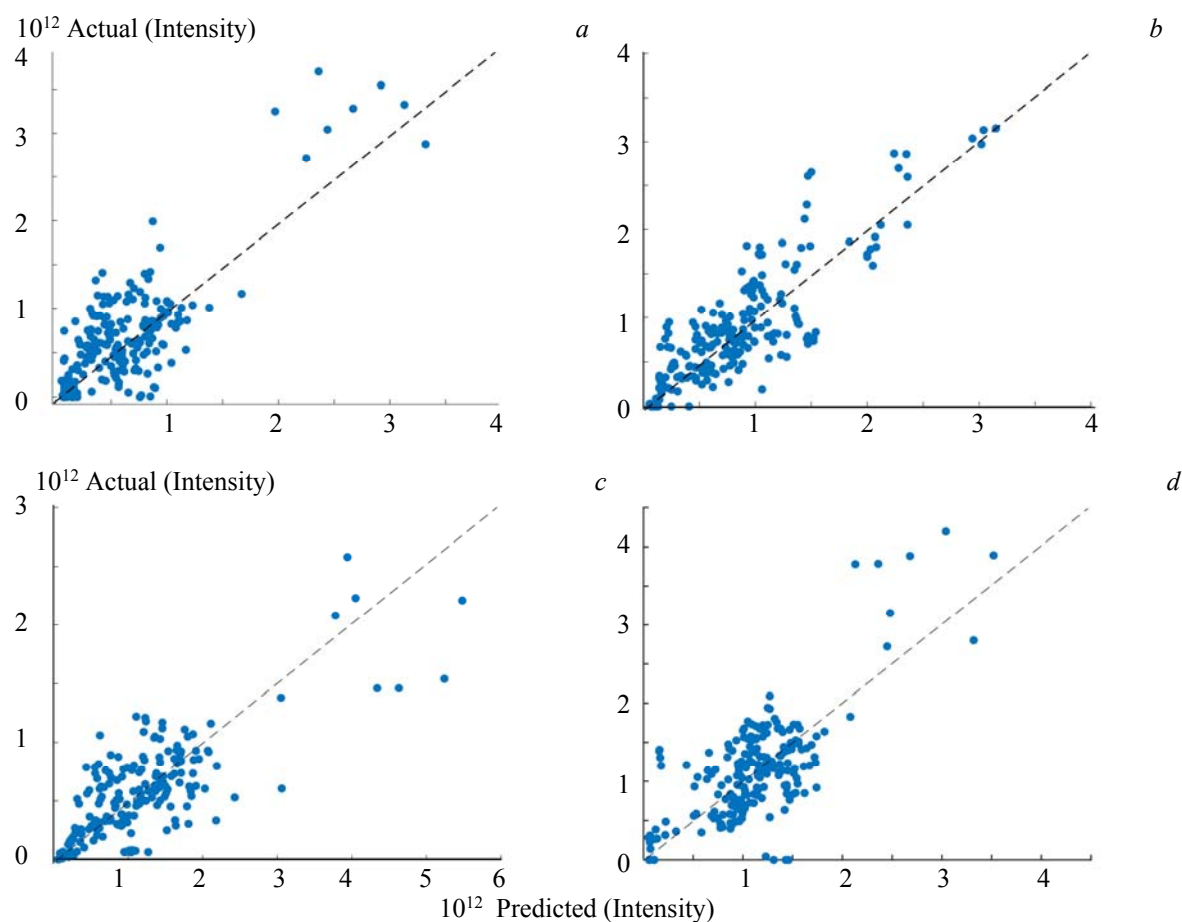


Fig. 7. Comparison of the correlations between predicted and actual integrated spectra in selected wavelength ranges containing Fe 275 nm (a), Na 285.2 nm (b), Si 288 nm (c), and Ti 334.8 nm lines (d).

Quantitative analyses. To perform quantitative analyses, we established a PLS2 [29, 30] model to test the effects of spectral transmission. PLSR is used to extract the principal components from two matrices (i.e., samples (X) and their corresponding labels (Y)) and then models the relationship between the two matrices. For the CCCT datasets, we used 401 standard samples to establish a quantitative analysis model without 7 transfer samples and then used 7 CCCT samples as the test set. In the standards dataset, 56 samples were transferred from spectra generated by the instrument in 2010 to the data distribution of the spectra generated by the instrument in 2015. The training data for the quantitative analysis model were established with spectral datasets for 352 samples from the 408 ChemCam samples generated in 2015 after 56 transfer samples were removed. The spectra of CCCT samples before and after transfer and the LIBS spectral data of the 56 samples released by ChemCam in 2010 were used as the test set to determine whether quantitative analysis accuracy was improved after transfer. In the partial least square quantitative regression model, eight potential factor numbers were selected.

As seen in Table 4, after the transfer of the CCCT dataset with ELM and transfer of the standards dataset with DELM, the performance of the transferred spectra in quantitative analyses was significantly improved. Only the KGa-2-Med-S sample in the CCCT dataset showed an opposite trend; however, after quantitative analyses with the recalibrated spectra of the sample, we found that the performance of the sample in quantitative analyses was still poor even after recalibration. As shown in Fig. 8, KGa-2-Med-S spectra generated on Mars and in the laboratory revealed the emergence of several new emission lines, including those for Mg (280 and 285 nm) and Ca (315, 317, 393, 396, and 422 nm). There may be two reasons for the new lines observed in the Mars data. First, KGa-2-Med-S is not as homogenous as other targets [7], and it is possible that the probe surfaces of the two KGa-2-Med-S targets were indeed chemically different. Another possibility is that ubiquitous Martian dust may have embedded itself on the surface of the target, changing the average composition and leading to new emission lines. For the standards dataset, it should be noted that the

two datasets were obtained in 2010 and 2015 at MSL. Although the settings of the experimental instruments were the same, there were some changes in the experimental conditions and sample conditions. The spectra from the CCCT dataset were very different, thus the transfer of this dataset was difficult. We chose to increase the complexity of the network and add the autoencoder to extract deeper and more accurate features to improve the effect.

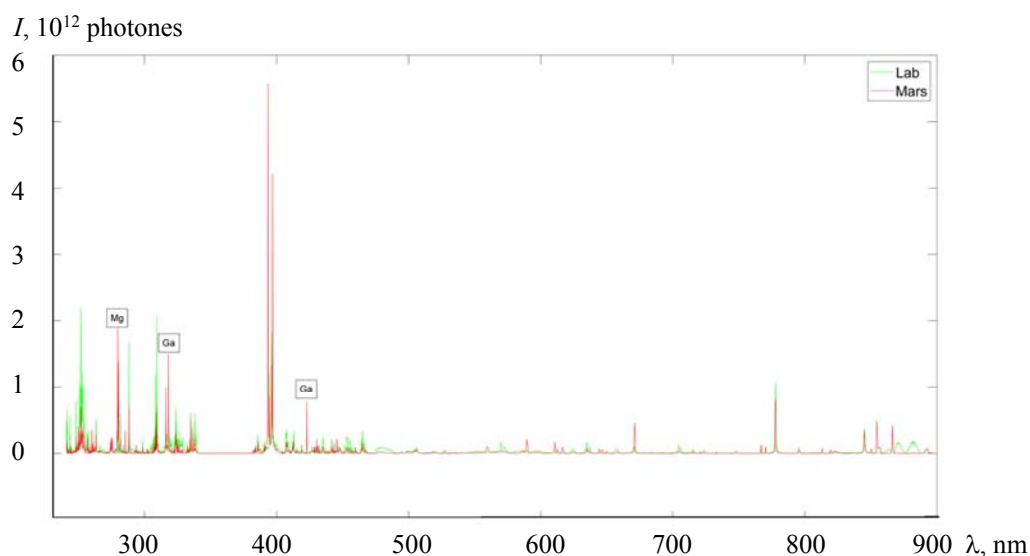


Fig. 8. Spectra showing some of the new emission lines observed from the Mars KGa-2-Med-S target relative to the laboratory target.

TABLE 4. RMSEPs of SiO₂ for Two Spectra Datasets with Different Transfer Methods

RMSEP	ELM		Uncalibrated	Recalibrated
<i>RMSEP for CCCT dataset (Mars to Earth)</i>				
Norite	3.2455		6.7547	3.0694
Picrite	1.0522		9.0632	6.6892
Shergottite	4.808		11.4345	3.8152
KGa-2-Med-S	10.9788		2.0141	7.3716
Nau-2Lo-S	3.512		9.8846	1.8186
Nau-2Med-S	5.324		14.2857	2.6320
Nau-2Hi-S	4.7282		22.1249	4.9326
Average	4.8069		10.7945	4.3326
<i>RMSEP for the standards dataset (2010 to 2015)</i>				
RMSEP	ELM	DELM	Uncalibrated	Recalibrated
$N = 20$	6.589	5.3549	7.6357	3.8942
$N = 30$	6.134	4.8993		
$N = 40$	5.887	4.1752		

Conclusions. We propose the application of an extreme learning machine and a deep extreme learning machine for small and large Laser-induced breakdown spectroscopy spectral data transmission, respectively. The calibration results of the CCCT dataset and standards dataset show that these two methods are stable in two dimensions and successfully correct the systematic differences between spectra obtained with different instruments. On the one hand, they are effective in calibrating the spectra themselves, while on the other hand, we input the corrected spectrum into the quantitative analysis model and obtained better results. In most cases, extreme learning machines and deep extreme learning machines can also use a small number of calibration targets to establish transfer relationships and obtain the best RMSEPs. These advantages ensure that the method proposed in this study is accurate and practical for transferring spectra from an instrument to

a well-established and maintained calibration model with very few transferred samples, which eliminates costly and time-consuming recalibration.

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