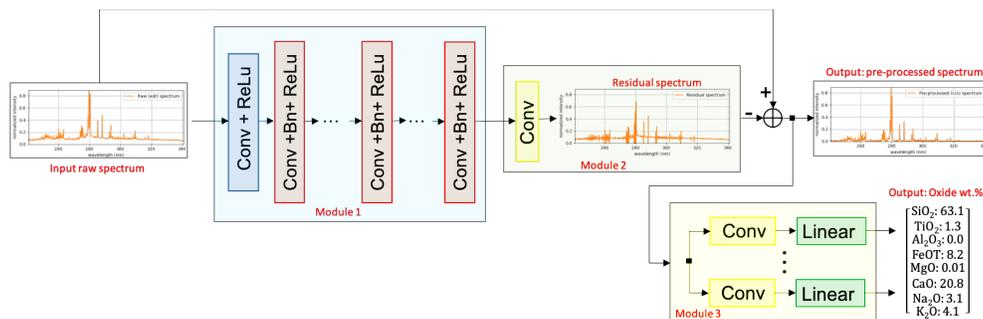**DEEP LEARNING FOR CHEMCAM ANALYSIS.** J. Castorena[1], D. Oyen[1], N. Klein[1], A. Ollila[1], C. Legget[1] and N. Lanza[1], [1]Los Alamos National Laboratory, Los Alamos, NM 87545, United States.

Figure 1: LIBS Pre-processing and calibration with CNN architecture.

Introduction: The ChemCam instrument onboard the Curiosity rover has shown excellent capabilities to remotely analyze the geochemical composition of rocks and soil samples on Mars. To this date, it has collected over 800000 laser induced breakdown spectroscopy (LIBS) signals encompassing the ultraviolet (UV), visible (VIO) and near infrared (NIR) bands between 240-905 nm. The analysis of these signatures to extract the geochemical composition is challenging due to sensor noise, including dark current, white noise, continuum removal, and sensor-to-target distance variations. The mixing effects of combinations of chemical elements also introduce uncertainties. The works of [2-3], among others, have aimed to reduce the impact of sensor noise on calibration (i.e., extracting chemical content from LIBS signatures) through the pre-processing of LIBS signals to adjust for the sources of instrument variation. In this work, we summarize our findings from [1] aiming at exploring the power of deep learning (DL) methods working under supervised strategies to learn both to pre-process and calibrate.

Approach: The proposed approach is a supervised deep learning method using a convolutional neural network (CNN) architecture that operates and is trained on LIBS signal data. The proposed architecture is capable of performing both the task of pre-processing and calibrating just by adding a head module of linear regressor layers. Below we include the problem formulation and CNN architecture.

Pre-processing. A discriminative learning approach is employed that learns to remove the noise $\mathbf{z} \in \mathbb{R}^N$ (including dark current, white noise, background continuum) from the signal $\mathbf{x} \in \mathbb{R}^N$ where N is the number of wavelengths in the spectrum. The pre-processing model of the raw measurements $\mathbf{y} \in \mathbb{R}^N$ is

$$\mathbf{y} = \mathbf{x} + \mathbf{z}. \qquad (1)$$

Training for pre-processing optimizes the parameters of a CNN architecture using a dataset of M raw-clean (rdr or ccs) spectral signal pairs $\{(\mathbf{y}_m, \mathbf{x}_m)\}_{m=1}^M$. The ℓ_2-norm loss function optimized is:

$$f_1(\mathbf{\Theta}) = \sum_{m=1}^M \left\| \mathbf{y}_m - \mathbf{x}_m - \underbrace{\mathcal{R}_{\mathbf{\Theta}}(\mathbf{y}_m)}_{\hat{\mathbf{z}}} \right\|_{\ell_2} \qquad (2)$$

where $\mathbf{\Theta}$ denotes the CNN parameters of the function $\mathcal{R}_{\mathbf{\Theta}}: \mathbb{R}^N \rightarrow \mathbb{R}^N$ and $\hat{\mathbf{z}}$ is the noise estimate.

Calibration. The model we propose to map LIBS signatures to the chemical constituents is the model $\mathbf{v} = \mathcal{F}(\mathbf{x})$ with LIBS input $\mathbf{x} \in \mathbb{R}^N$ representing either a raw or preprocessed signal and an output vector $\mathbf{v} \in \mathbb{R}^C$ of the chemical element composition (in % oxide) with C being the number of chemical elements. The problem is treated as that of learning a calibration function $\mathcal{F}_{\mathbf{\Theta}'}: \mathbb{R}^N \rightarrow \mathbb{R}^C$ with CNN parameters $\mathbf{\Theta}'$. This is achieved by training using M raw (edr) or preprocessed (e.g., rdr, ccs) LIBS signal and its corresponding truth chemical compositions. The loss function optimized to learn the parameters $\mathbf{\Theta}'$ with the training example pairs $\{(\mathbf{x}_m, \mathbf{v}_m)\}_{m=1}^M$ is:

$$f_2(\mathbf{\Theta}') = \sum_{m=1}^M \| \mathbf{v}_m - \mathcal{F}_{\mathbf{\Theta}'}(\mathbf{x}_m) \|_{\ell_2} \qquad (3)$$

where $\mathcal{F}_{\mathbf{\Theta}'}(\mathbf{x}_m)$ denotes the estimate of chemical constituents.

The CNN architecture we employ is shown in Figure 1. It comprises three modules: Module 1-2 which are used for pre-processing the raw LIBS signal and Module 3 of linear regressors whereby adding it as a head to Module 1-2 adds the capability to obtain chemical constituents through calibration. Module 1 consists of a convolution and rectifier linear unit (ReLu) layer followed by a sequence of D convolution, batch normalization (Bn) and ReLu interleaved layers, Module-2 is a convolution and subtraction of the residual estimation and Module 3 consists of a convolution layer followed by a fully connected (FC) layer to perform a linear regression.

Experimentation: Validation of the proposed approach is performed on the 'Mars' and 'Calib' ChemCam datasets in [4]. In our experiments we use combination pairs between edr, rdr, ccs, ground truth

a) raw LIBS signal

b) pre-processing comparison
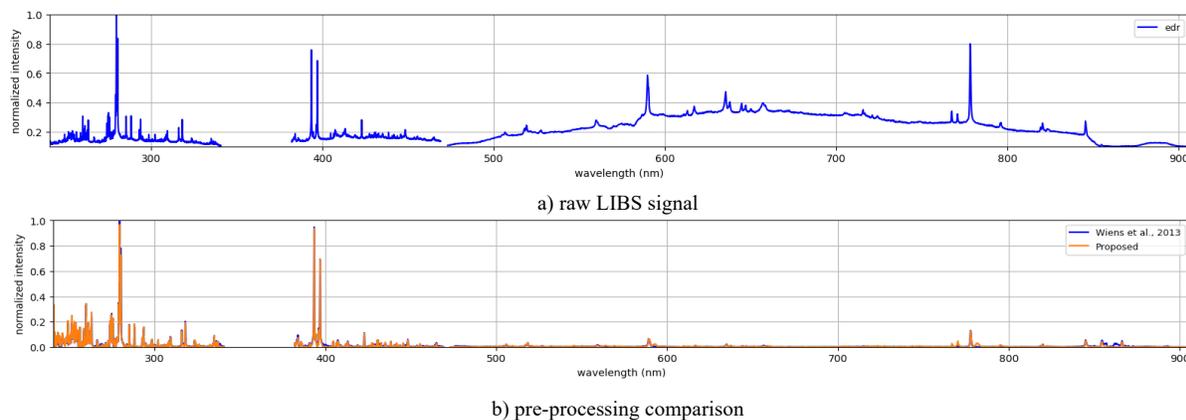
Figure 2: Pre-processing example (sols: 0293, target: Duluth, dist: 2.68 m) from Mars dataset.

chemical content depending on the task. The 'Calib' dataset is obtained in a laboratory setting on Earth using 408 calibration standards of known and certified qualitative and quantitative chemical reference composition. Experiments on both pre-processing and calibration tasks are performed to validate the capability of the proposed method. Evaluations take place on individual laser pulse shots (non-averaged).

For learning pre-processings, the training stage uses pairs of raw and pre-processed signals, this later using the method of [2] as ground truth. Other alternatives could also be learned by combining a variety of cherry-picked methods. Figure 2 shows a representative example of a result and comparison against that of [2] in the test 'Mars' dataset. Note in 2b that the proposed method was able to learn and predict the pre-processings of [2]. The results of the average RMSE errors for pre-processing in a test set comprising 10,000 held out from training shots where 0.078 and 0.079 in the Mars and Calib dataset, respectively, with a max RMSE normalization of unity. These, small RMSE values demonstrate that the learned function and proposed method is generally a good approximation to the pre-processing in [2].

Evaluations on calibration are conducted using the 'Calib' dataset with the 8 major chemical elements only. Comparisons are made against ICA [3], SM-PLS [3] and our method trained under two strategies: (1) that first learns to pre-process and then to calibrate each step trained independently or (2) an end-to-end trained network without supervision on the pre-processing. The reason of adding a comparison between the later two training strategies is to evaluate if any gains in performance could be achieved by reducing potential information loss at pre-processing. Our findings are summarized in Table I using a test set of 7,500 shots from targets held out at training. The results demonstrate our method is capable of learning calibration functions with performance comparable to

state of the art. The end-to-end learning strategy (i.e., without pre-processing supervision) in the last column of Table I offers a marginal but superior performance. In terms of computational complexity and network size, the added linear regressor layers were insignificant relative to pre-processing. In fact, all learning schemes proposed can generate estimates at rates of 50 Hz on CPU with memory storage of 1MB per model.

Table I. Calib. performance RMSE in oxide wt. %

Element	pre-Proc.[2] SM-PLS[3]	pre-Proc.[2] ICA [3]	Our Pre-proc + linear reg.	Our end2end
SiO_2	4.33	8.31	4.716	4.193
TiO_2	0.94	1.44	1.654	0.478
Al_2O_3	2.85	4.77	3.279	1.913
FeO_T	2.01	5.17	1.672	2.787
MgO	1.06	4.08	1.21	0.947
CaO	2.65	3.07	2.283	1.244
Na_2O	0.62	2.29	1.075	0.745
K_2O	0.72	0.98	1.841	0.796

We conclude by summarizing the benefits of the proposed approach being two-fold. (1) the streamlining of the entire processing pipeline and (2) the capability to achieve in-situ, real-time processing. For calibration, our experiments show a marginal but superior error performance relative to existing methods.

References:
[1] Castorena, J., Oyen, D., Ollila, A., Legget, C. and Lanza, N. (2021). *Spectrochimica Acta Part B: Atomic Spectroscopy*, *178*, 106125. [2] Wiens, R. C. et al., D. (2013). *Spectrochimica Acta Part B: Atomic Spectroscopy*, *82*, 1-27. [3] Clegg, S. M., Wiens, R. C., Anderson, R., Forni, O., Frydenvang, J., Lasue, J. and Maurice, S. (2017). *Spectrochimica Acta Part B: Atomic Spectroscopy*, *129*, 64-85. [4] http://pds-geosciences.wustl.edu