A FUZZY LOGIC ALGORITHM FOR ANALYZING HANDHELD LIBS SPECTRA OF METEORITES. G. S. Senesi[1], P. Manzari[2], A. Consiglio[3], O. De Pascale[1], [1]CNR - Istituto di Nanotecnologia (NANOTEC) - PLasMI Lab, Via Amendola 122/D, 70126 Bari, Italy, giorgio.senesi@nanotec.cnr.it, [2]Istituto Nazionale di Astrofisica - Istituto di Astrofisica e Planetologia Spaziali (INAF-IAPS), via Fosso del Cavaliere 100, Roma, Italy, [3]CNR - Istituto di Tecnologie Biomediche (ITB), Via Amendola 122/D, 70126 Bari, Italy.

Introduction: In the last decade Laser-Induced Breakdown Spectroscopy (LIBS) has been extensively applied to chemical investigation of meteorites [1 and references therein]. Recently, the use of portable and handheld instruments for the rapid chemical characterization [2,3] has become increasingly important where in-situ analysis is a necessary requirement and the preservation or little loss of material is desirable. The novelty of this work consists in the use of an handheld LIBS instrument for the analysis of meteorites and in the application of a never tested fuzzy logic computational approach to the interpretation and classification of the spectra obtained. As a cost effective technique, LIBS offers the possibility to collect several data, thus reducing the error of the measurements, whereas multivariate analysis of LIBS spectra allows to manage large amounts of data, by facilitating their interpretation.

In particular, any multivariate calibration technique, which included partial least squares regression - discriminant analysis (PLS-DA), principal component analysis (PCA), multi-linear regression (MLR) and artificial neural networks (ANN) [4], would theoretically allow to discriminate classes within a group of LIBS spectra by taking into account different variables and reducing the complexity of the spectra. Multivariate analysis algorithms, however, are not always able to account for the properties of the investigated materials. For example, PCA can not account for between group-to-group and within-group variability [5], PLS-DA can be applied only to linearly separable data [6], while ANN produce black box models [7].

In this work, an approach based on fuzzy sets and genetic algorithms is applied, with the aim of producing an inference system based on fuzzy rules, which allows the interpretation and evaluation of meteorite LIBS spectra and the efficient selection of the most significant features for class discrimination, thus reducing the complexity of the classification procedure.

Materials and methods: The investigated materials consist of a number of classified iron meteorites, including Agoudal, Dronino, Morasko, Muonionalusta, North West Africa 11104, stony meteorites, including North West Africa 1296 and North West Africa 4051, some unclassified meteorites and some cast iron samples (generated from industrial and manufacturing sources) which would be identified as iron meteorites based on a visual identification, but their origin can be certified only by a more deeper chemical analysis in the laboratory (meteor-wrongs).

A portable, handheld LIBS instrument (B&W Tek, Newark, DE, USA) was used to perform the measurements. The instrument consisted of a miniature-diode-pumped, solid-state, short-pulsed laser emitting at a wavelength of 1064 nm with a maximum output of 300 mW, a pulse duration of 500 ps and a maximum pulse energy of 150 μJ, and operating at a high repetition rate, *i.e.* between 1 and 5 kHz. A compact spectrometer allowed to record the spectra in the non-gated mode in a wavelength acquisition range from 180 to 800 nm, *i.e.* from UV to visible, with an overall resolution of 0.4 nm for the entire spectral range. The whole setup was enveloped in a lightweight handheld body.

The workflow used in this work is summarized in Figure 1. The first step consists in the development of a genetic algorithm for the automated selection of the features, i.e. the most significant wavelengths allowing to discriminate samples. In this algorithm, the individuals consist of randomly selected features, the fitness function is the accuracy of the fuzzy inference system built on the selection of features, whereas the generation of new individuals is implemented by the recombination of portions from two individuals, which are selected by roulette wheel extraction.

The fuzzy inference system is composed by a number of sets defined uniformly over the domain of each feature. Some linguistic labels are used for the description of the rules, i.e. the IF-THEN rules take the form "IF feature1 IS very high AND feature2 IS low and feature3 IS medium ... THEN class is A." Each rule has a weight correlated to the strength of the proposed concept.

The accuracy of the method is evaluated by leave-one-out cross validation, i.e. the model is trained repeatedly on all the samples of the dataset but one, and it is tested on the observation excluded. A mean of the resulting accuracies is considered as the overall accuracy of the model.

The whole workflow was developed by using the environment for statistical computing R [8]. Further, the package “FRBS” [9] has been used in the fuzzy inference system for the training and testing steps.

Results: The method proposed was tested on the dataset of iron meteorites, and compared to the other meteorites and meteor-wrongs. The genetic algorithm was set so to select randomly 20 features (wavelengths) at most, which were used as the input for the fuzzy inference system. The model generated by the genetic algorithm reached an accuracy of 100% (using the leave-one-out cross validation) with a selection of 7 features. Thus, the final model was able to distinguish all the input samples by involving only 7 wavelengths in its IF-THEN rules.

Conclusion: In this work, an innovative strategy has been proposed for meteorite classification based on the LIBS spectra measured by an handheld instrument. The approach used integrates automated feature selection and supervised learning in a fuzzy rule based classifier. The method has been tested to build a model for the automated classification of iron meteorites based on LIBS spectra. The final model showed to classify correctly iron meteorites using a set of fuzzy rules that can be easily interpreted by humans. Further, the identification of the elements yielding the 7 wavelengths selected, would allow the fuzzy rules to be exploited for further chemometrics studies.

In conclusion, the use of handheld LIBS instruments appears particularly promising for performing in-situ measurements to identify and classify meteorites, and so to decide previously any further interventions in the laboratory.

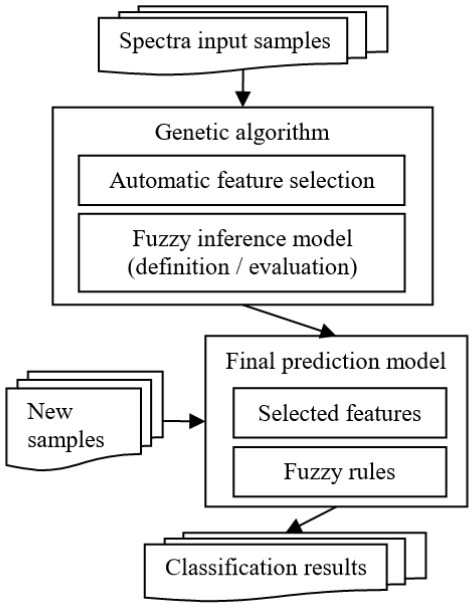

Figure 1 - *Workflow for the feature selection and the definition of the fuzzy-rule-based inference model for meteorites classification. The model obtained can be used to classify unknown new samples.*

References: [1] Senesi G.S. (2014) *Earth Sci. Rev., 139*, 231-267.

[2] Gemelli M., D'Orazio M., Folco L. (2015) *Geostand. Geoanal. Res., 39*, 55-69.

[3] Senesi G.S., Tempesta G., Manzari P., Agrosì G. (2016) *Geostand. Geoanal. Res., 40*, 533-541.

[4] Dyar M.D., Carmosino M.L., Breves E.A., Ozanne M.V., Clegg S.M., Wiens R.C. (2012) *Spectrochim. Acta B, 70*, 51-67.

[5] Barker M. and Rayens W. (2003) *J. Chemom., 17*, 166–173.

[6] Cover T.M. (1965) *IEEE Trans. Electron. Comput., EC-14*, 326-334.

[7] Linda O., Manic M., McJunkin T.R. (2011) *Proceedings of the 4th International Symposium on Resilient Control Systems (IEEE, 2011),* 35-41.

[8] R Core Team (2015). R: A language and environment for statistical computing. *R Foundation for Statistical Computing*, Vienna, Austria.

[9] Riza L.S., Bergmeir C.N., Herrera F., Benítez Sánchez J.M. (2015). frbs: Fuzzy rule-based systems for classification and regression in R. *American Statitical Association*.