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Development of LIBS for planetary explorations: the effects of pulse energy on the accuracy of elemental analyses

Yuichiro Cho[1*], Seiji Sugita[2], Ko Ishibashi[3], Sohsuke Ohno[3], Shunichi Kamata[1], Koji Wada[3], Tomoko Arai[3], Hiroki Senshu[3], Masanori Kobayashi[3], Noriyuki Namiki[3], Takafumi Matsui[3]

[1]Earth and Planetary Science, Univ. Tokyo, [2]Complexity Sci. and Eng., Univ. Tokyo,
[3]PERC, Chiba Institute of Technology

Laser-induced breakdown spectroscopy (LIBS) is a recently developed elemental-analysis method. LIBS measurement is carried out by irradiating high-intensity laser pulses focused on a target materials, creating a light-emitting plasma, which will be observed by a spectrometer. Since LIBS has several advantages including remote measurements, rapid analyses, the ability to remove dust layers on the surface of a target, it is planned to be on board for the future landing missions toward the Moon and Mars.

Pulse energy of laser is one of the most essential parameters of on-board LIBS instrument, However, the effects of the laser energy on accuracy of LIBS measurements have not been evaluated quantitatively. In addition, the influences of the laser energy on LIBS spectra were not fully understood. These problems have been a bottleneck in the development of an on board LIBS instrument.

In order to resolve this problem, Cho et al. [1] investigated the effect of laser energy on LIBS spectra by changing the laser energy on the surface of a basalt target. They found that (1) the total spectral intensity increases proportionally to the laser energy over the threshold for breakdown of the target, (2) the local thermal equilibrium condition may be applicable to the plasma vapor allowing the emission intensity to be described with one plasma temperature for each irradiation condition, and that (3) the same rock sample may generate different spectra for different laser energies because different plasma temperatures would occur. However, the relationship between accuracy of elemental analyses and the laser energy has not been examined systematically yet.

Thus, in this study, we changed the laser energy from 40 to 70, 100, 150, and 250 mJ and obtained five spectra for these laser energies in order to investigate the effects of laser energy on the accuracy of the elemental analyses. The laser energy of 40 mJ was near the lower limit to generate observable laser plasma. The targets were 16 pellets of igneous rocks including basalts and andesites. The samples were placed in a vacuum chamber filled with 9 mbar CO_2 to simulate the Martian atmosphere. We acquired the spectra of the five laser energies and predicted the concentrations of Si, Fe, Mg, Ca, Al, Ti, Na, and K using partial least squares regression (PLSR) method [2,3]. We then compared the PLSR results with elemental concentrations obtained with the X-ray fluorescence (XRF) method. Thus we estimated the accuracy (root mean square error of prediction; RMSEP) and precision (error bars of predicted concentrations) of LIBS measurements for the above-described five laser energies.

Our preliminary results reveal that the accuracy and the precision of LIBS measurements have few relations with laser energy. As laser energy is decreased from 250 mJ down to 40 mJ, the accuracy and the precision of the measurements for some elements are improved but those for others are deteriorated. For instance, RMSEPs of measurements for the laser energy of 250 mJ, 1 00 mJ, 40 mJ are 4.54, 7.09, 7.55 wt% for SiO_2; 3.05, 8.05, 8.29 wt% for Fe_2O_3; 2.27, 2.25, 2.80 wt% for MgO; 1.80, 2.49, 2.58 wt% for Al_2O_3; 1.19, 0.182, 0.208 wt% for TiO_2; 1.91, 1.21, 1.10 wt% for CaO; 0.47, 0.56, 0.63 wt% for Na_2O; 0.629, 0.051, 0.146 wt% for K_2O, respectively. The standard deviations of predicted concentrations also fluctuate as the laser energy is changed, but no clear trend was observed.

The results of this study indicate that elemental analyses are possible even if the laser energy is not so high. This energy-unaffected nature would suggest that we can use low-energy laser without compromising a measurement accuracy. This would further leads to reduction in both engineering and economical costs of an on board LIBS instrument.

References

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[3] Clegg et al., 2009, Spectrochim. Acta. B 64

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