An experimental study on development of a LIBS instrument for planetary explorations: effects of pulse repetition rate

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The Japanese planetary exploration program in the past has been largely based on remote sensing approaches using either flyby or orbital observations. However, after the touchdown on asteroid Itokawa by Hayabusa, a number of landing plans on our Moon or Mars have been discussed. For a landing mission, measurement of the elemental compositions of rocks around the landing site promptly and precisely over a wide range is extremely important. Among several composition-measurement methods, LIBS (Laser-Induced Breakdown Spectroscopy) outstands for its characteristics meeting these requirements sufficiently. LIBS is a method carried out by shooting a laser beam focused on a target rock, creating a light-emitting plasma plume, which will then be broken down into spectra with a spectrometer. The advantages of LIBS include stand-off measurements, rapid analyses, no need for chemical pretreatment, and the capability of analyzing a layered structure. However, there are unresolved issues. For instance, a decrease in signal intensity under lower atmospheric pressures has been reported [1]. Another example is a relation between laser pulse repetition rates and spectral intensity of vaporized plasma, which is reported by Kamata et al. [2]: (1) The total spectral irradiance of laser-induced plasma increases by the factor of 1.1 to 10 as the laser pulse repetition rate rises from 1 Hz to 10 Hz. (2) Some spectral lines, especially those of ions, appear at 10 Hz, but they are unseen at 1 Hz. (3) The accuracy of the result obtained by elemental analysis improves when the laser-induced plasma is brighter. Thus, raising the repetition rate is an excellent means for improvement of analysis accuracy. However, the reasons why these phenomena occur have not been understood well yet. Revealing the physical mechanisms of the enhancement in spectral intensity is important for future improvement of LIBS and for finding the optimal pulse repetition rate for a LIBS system on board a planetary exploration spacecraft so as to maximize the scientific output for a given engineering costs.

Because the timescale of laser pulse interval (0.1 - 1 sec) is much longer than the lifetime of laser vapor plumes (1 - 10^3 micro sec), the pulse repetition is likely to influence the laser plume via solid target, most likely in a form of temperature rise. Rise in target temperature, may cause the followings: (1) heating of a subsequent laser plume, (2) rise in the degree of ionization of the plume, (3) increase in plume mass. First, in order to probe the target temperature rise, we changed the pulse repetition rates from 0.1 Hz up to 10 Hz, tracing the surface temperature with a thermal imaging camera. Second, we irradiated an ilmenite target and measured the electron temperature of Fe I and Ti I using Boltzmann plot in order to investigate (1). As for (2), we chose a serpentine as a sample. The electron density was measured using Stark broadening effect with Mg+ spectral line (448 nm). We repeated the measurements changing the pulse repetition rate from 1 Hz to 10 Hz.

Our preliminary experimental results show; (i) the increase in target temperature is 20 deg. at 1 Hz, 100 deg. at 10 Hz, respectively. (ii) The electron temperature is 6800 plus minus 1000 deg. for both Fe I and Ti I regardless of the repetition rates. (iii) The ionization rate of Mg+ grows by no more than 20 % although the spectral intensity rises by the factor of 4.

The 100 deg. rise in target temperature can lead to substantial elevation in evaporation efficiency. However, alterations in plasma temperature and ionization rates are not sufficient for explaining the growth of spectral intensity due to the increase of pulse repetition rate unfortunately. These results imply that the change in evaporated mass by the increase in the repetition rate contributes to the signal enhancement.

References

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[2] Kamata et al., LPSC, (2008) in press