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Double-pulse laser-induced breakdown spectroscopy of multi-element sample containing low- and high-Z analytes

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ABSTRACT

Laser-induced breakdown spectroscopy (LIBS) is a portable, remote, non-invasive analytical technique which effectively distinguishes neutral and ionic species for a range of low- to high-Z elements in a multi-element target. Subsequently, LIBS holds potential in special nuclear material (SNM) sensing and nuclear forensics requiring minimal sample preparation and detecting isotopic shifts which allows for differentiation in SNM (namely U) enrichment levels. Feasible applications include not only nonproliferation and homeland security but also nuclear fuel prospecting and industrial safeguard endorsement. Elements of higher mass with complex atomic structures, such as U, however, result in crowded emission spectra with LIBS, and characteristic emission lines are challenging to discern. Preliminary research suggests double-pulse LIBS (DPLIBS) improves signal sensitivity for analytes of lower atomic mass over conventional single-pulse LIBS (SPLIBS). This study investigates signal sensitivity for low- and high-Z analytes in a glass matrix containing U (1.3%) comparing DPLIBS to SPLIBS. DPLIBS involves sequential firing of 1064 Nd: YAG (FWHM 9 ns) pre-pulse and 10.6 μm TEA CO₂ (FWHM 50-100 ns) heating pulse in near collinear geometry; SPLIBS entails only the Nd:YAG laser. Optimization of experimental parameters including inter-pulse delay and energy follows identification of characteristic lines for bulk analytes Ca, Na, and Si and trace analyte U for both DPLIBS and SPLIBS. Temporally-integrated excitation temperature and electron density as well as neutral-to-ionic species ratio constitute relative figures of merit for both DPLIBS and SPLIBS plasma characterization. Temporally-resolved studies provide insight into high-Z U analyte persistence and signal enhancement with DPLIBS as compared to low-Z bulk analytes. The study predicts and discusses optimal emission conditions of U lines and relative figures of merit in both SPLIBS and DPLIBS.

KEYWORDS

Double- and single-pulse laser-induced breakdown spectroscopy (LIBS), nuclear forensics, Uranium, signal sensitivity