Laser-induced Breakdown Spectroscopy Analysis for Addressing Copycat Crime of Alexander Litvinenko (Polonium-laced Tea)

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1. Introduction

According to the international atomic energy agency (IAEA) incident and trafficking database (ITDB), unauthorized access to radioactive materials for malicious use is getting easier [1]. One of the most shocking accidents is murdering a person named Alexander Litvinenko using radioactive materials. The cause of death was poisoned by radionuclide polonium-210 in tea. In his body, significant amounts of the rare and toxic element were founded by Health Protection Agency. Through the Alexander Litvinenko poisoning with polonium-210, the era of nuclear terrorism started [2].

2. Experimental and Results

2.1 Experimental

To address the radiological threat of food poisoning rapidly, the laser-induced breakdown spectroscopy (LIBS) was considered. LIBS is a type of atomic spectroscopy. The atomic spectra were emitted through the returning to the ground state. In this process, a laser is used as excitation source the samples by forming a plasma. The emitted spectra were transmitted to a spectrograph and recorded to intensified-CCD that can control a few ns time gate.

For excitation the sample, a compact Nd:YAG laser (Quentel Q-smart 450 with the 2nd harmonic module, 532 nm, 5.5 ns, 220 mJ, 20 Hz) was used. For acquiring the spectra, two plano-convex lenses were used and the emissions were analyzed by a spectrograph (Andor, ME5000) with nano-scale of time resolution (Andor, iStar DH334T).

Fig. 1. Experimental setup for aqueous form of samples.

The target material was selected as SrCl₂ which is typically used as a radiation source. Since it has high solubility in water (53.8 g/100 mL), easy to use in terrorism such as diluting radionuclides in water tank installing rooftop of buildings and copycat crime of Alexander Litvinenko as known for polonium-laced tea. To calculate the limit of detection for the LIBS technique, numerous concentration was used. In addition, only 4 μL of the sample was investigated by a single laser pulse and it takes only 30 μs for each experiment.

2.2 Optimization

To find a trace of radionuclides in something aqueous form as much as LIBS can, the optimization process was conducted by finding the most signal-to-noise ratio point of LIBS condition. The three parameters of delay time, laser power, and gate width were investigated. As shown in Fig. 2, the time delay of 180 ns, the laser power of 40.4 mJ, and the gate width of 30 μs were obtained as an optimization condition. Fig. 2 depicts an average of wavelength including 338.07, 346.44, 407.77, 421.55, 430.55, 460.73, and 481.18 nm of Sr persistent emission.
2.3 Results

Among seven emission line of Sr which is used to find optimization process, the three emissions of 407.77, 421.55, and 460.73 nm were used to calculate the limit of detection (LOD) because these exist clearly in the spectra. Fig. 3 depicts the calibration curves fitted by LIBS. For each point, 30 times of investigation were conducted with numerous concentration. The calibration curve was well fitted with good linearity ($R^2=0.9889$ for 407.77 nm, $R^2=0.9933$ for 421.55 nm, $R^2=0.9834$ for 460.73 nm) as shown in Fig. 3.

![Fig. 2. Optimization points for energy, time, and width.](image)

![Fig. 3. Calibration curve of SrCl$_2$ in water.](image)

The calculated LODs for each emission line was 197.805 ppm for 407.77 nm, 285.579 ppm for 421.55 nm, 862.108 ppm for 460.73 nm. The lowest LOD was obtained at 407.77 nm due to the highly stable emission for blank measurements overcome.

3. Conclusion

This preliminary result shows the LIBS can be used for analyzing the presence of radionuclides in aqueous medium rapidly. The LODs look like quite high to analyze the trace amount of radioactive materials. However, this limitation can be overcome by accumulating the laser pulse certain times because the LOD can be enhanced by integrating laser pulse can enhance the LOD exponentially.

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