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Countering Radiological and Nuclear threats

Compact setup for standoff laser induced breakdown spectroscopy of radioactive material

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Abstract

Radioactive materials present a major threat and can cause severe direct and long term injuries to humans as experienced i.e. in the Fukushima and Chernobyl nuclear plant catastrophes. Furthermore, intended use of radiological dispersal devices may spread radioactive materials over large areas. Detecting these hazards and investigating the status of contaminated areas a remote standoff determination of nuclear fission products would serve as a helpful tool for first responders and damage control teams. Laser induced breakdown spectroscopy (LIBS) offers a unique possibility for the identification of nuclear fission products and can be able to distinguish different isotopes of the same species. Within this scope and based on experiences with a high power / long distance (> 100 m) LIBS setup a compact and low power setup is presented. The compactness allows for handheld operation as well as mounted on a small robot or on an unmanned aerial vehicle (UAV) an advanced setup could be controlled remotely and would be able to safely determine radioactive materials.

Keywords: radioctive materials, Laser-induced breakdown spectroscopy (LIBS)

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1. Introduction

Radioactivity is the ability of an unstable atom to spontaneously emit ionizing radiation while the atomic core decays or changes its state. After its discovery radioactivity enabled many different technologies in diverse fields like medicine, science or energy production [1, 2]. But radioactive material has also the potential to be used as devastating weapon and may cause harm to a large number of people. Since the radiation is not visible and can cause severe injuries different detection systems have been invented [3]. For example the Geiger counter or the dosimeter measures directly the ionizing radiation. These devices are helpful and widespread detection systems but they are often not able to determine the source material and saturate in high radiation fields.

Laser induced breakdown spectroscopy (LIBS) is an approved technique capable of qualitative and quantitative material analysis. The technique was developed shortly after the invention of the laser and since the 1980s interest has grown rapidly, since the technique is fast, can be used for almost all samples in solid, liquid, aerosol or gaseous phase and no sample preparation is needed. Due to this advantageous properties, the LIBS technique was used for many applications ranging from physics to engineering, geology and even in space missions [4–9]. There are many reviews describing different aspects of the technique and comprehensive historical and technical overviews can also be found [10]. Furthermore, within the last years different commercial handheld LIBS devices emerged which are able to detect different materials and identify alloys [11].

Within the scope of radioactive material detection different investigations of the LIBS technique were performed and can be found for example in [12, 13]. For some materials like Uranium (U) and Polonium (Po), which provide large isotope shifts on the order of 0.02-0.2 nm, it is even possible to differentiate between their isotopes [14]. But, for other materials of interest, as for example the fission products lead or strontium, the shift is too small (0.001 nm or smaller) to be resolved with common compact spectrometers. Nevertheless, depending on the scenario an adapted system may be able to help first responders to detect radioactive material and initiate countermeasures quickly or investigate contaminated regions for forensic application fast and from a secure distance [15, 16].

Within this scope investigations utilizing a compact low power setup are presented, examining its detection capabilities of nuclear fission products. The setup is based on the experience with a high power long distance LIBS setup described in [17]. In a first step, a compact low pulse energy laser with output pulse energies on the order of up to 300µJ is used to detect nuclear fission products within a sample. Afterwards it is investigated how well these materials can be distinguished from background materials and a comparison is done with the literature [15] using a different set of sample and background materials. In another step the limit of detection for the used setup is examined by detecting lead within a background material. The compactness and low power consumption of the setup will allow for handheld operation as well as being mounted on a unmanned vehicle as in [5]. An advanced setup could be controlled remotely and would be able to safely detect radioactive materials.

2. Experimental setup

In Figure 1 the experimental setup is displayed. It makes use of a compact, pulsed, diode pumped Nd:YAG laser Crylas FTSS 355-300 (CryLaS GmbH, Berlin, Germany) with output pulse energies of up to 300 µJ, a pulse lengths of approximately 1.7 ns and a repetition rate of up to 60 Hz. A system consisting of three lenses is used to first enhance the beam diameter and than focus the beam onto the sample. A beam diameter of a few 10 µm can be achieved and it is possible to reach a sufficiently high power density to create a stable plasma using relatively low pulse energies of below 300 µJ. To collect the radiation from the excited atoms and ions after relaxiation of the plasma a lens and a reflective fiber collimator is used. It focusses the radiation onto the fiber of an Avaspec multichannel spectrometer from Avantes BV (Apeldoorn, Netherlands). This CCD-based spectrometer is used to analyze the radiation as well as providing the timing for the measurement. The spectrometer sends out a trigger pulse to the laser and starts data collection a few hundred microseconds afterwards. Therefore, it is assured that the plasma has relaxed and the specific radiation of the excited atomic and ionic species is not obscured by the plasma radiation.

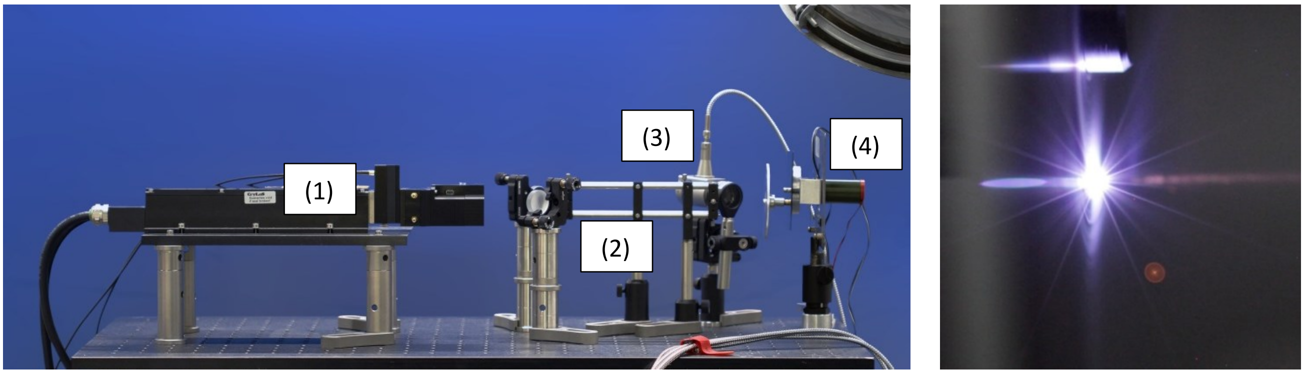

Figure 1: Left: Experimental setup consisting of the laser (1) , focussing optics (2), collection optics (3) and the sampleholder (4); Right: picture of plasma created in the laboratory.

The system parameters are summerized in the following table:

System Parameters	
Laser system	Crylas FTSS 355-300
Laser pulse energy	< 300 µJ
Repetition rate	20 Hz
Focus diameter	< 20 µm
Collection optics diameter	2.41 cm
Spectrometer	Avantes multichannel
Standoff distance	5 cm
Acquisition time (100 spectra)	< 10 sec

For the investigations different metal and plastic samples as possible background materials and different salts are used, namely $BaSO_4$, Cs_2CO_3 and $PbCl_2$ as source for the nuclear fission products Ba, Cs and Pb. Powders of the used salts were mixed with KBr powder with a ratio of 20 % to 80 % with a mortar and than pressed under vacuum with a weight of 10 tons resulting in pellets of 1 cm diameter. For the metal samples no preparation was used. A picture of the samples is displayed in Figure 2.

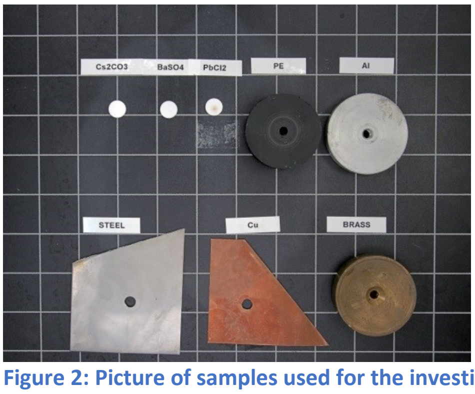

Figure 2: Picture of samples used for the investigations. The white pellets are prepared from mixtures of 80 % KBr and the specified sample material. PE stands for Polyethylene and the metal samples are specified in the picture.

2. Experimental results

With the presented measurements the performance of the compact setup for detection of nuclear fission products is investigated, as well as the distinction of these materials from possible background materials. Therefore, LIBS spectra of the samples containing barium, cesium and lead are measured and compared to toe ones from aluminum, steel, polyethylene, copper and brass samples. The recorded spectra of Barium and Cesium are in agreement with Gaona et al. [15] and Lang et al. [16] with differences due to slight variations in source material, detection optics, used laser wavelength and pulse energy to create the plasma. The presented spectra (see Fig. 3) are averaged over 100 measurements that are acquired with a sample rate of 20 Hz.

Qualitative LIBS Analysis

Spectra of nuclear fission products as well as different background materials are displayed in Figure 1.

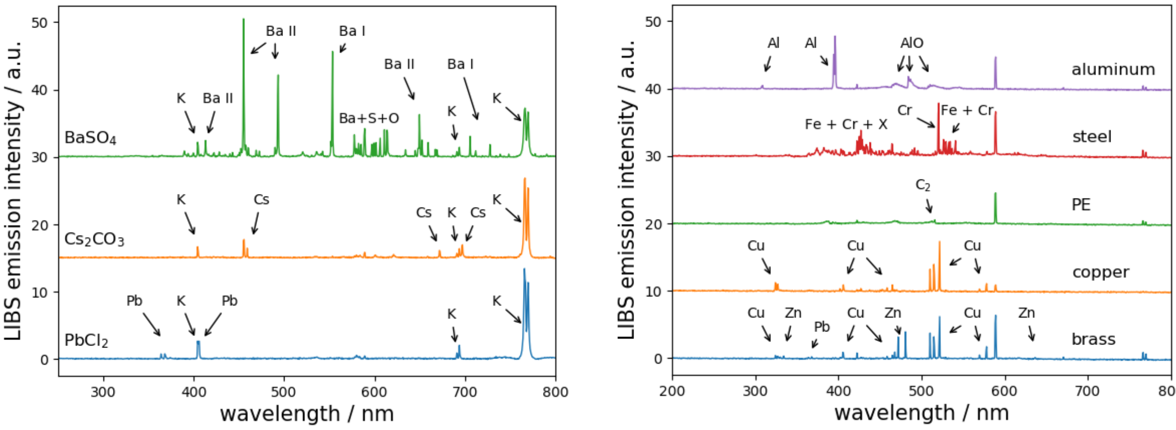

Figure 3: left: spectra of nuclear fission products, right: spectra of possible background material.

A peak finding algorithm implemented in the python programming language is applied to extract the peak positions with a peak height larger than 0.05 within the measured datasets. These positions where assigned to atomic and ionic emission lines using the NIST Standard Reference Database [18]. Except for steel there is no overlap of the dominant peaks with the nuclear fission products Barium (554 nm) and Cesium (445 nm, 697.3 nm). Within the brass spectrum a small lead content could be detected, typically added to brass to enhance the machinability. For the other samples no interference between the lead spectrum and the metal samples is observed. To further investigate the possible interference of the background material spectra with the ones of the nuclear fission products we plotted the peak positions of the background material in gray and the peak positions of the nuclear fission products in the same colors as the ones in the left part of Figure 3.

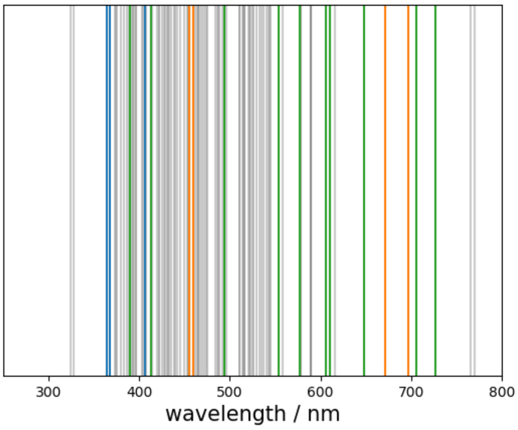

Figure 4 peak positions of spectral lines of the measured background materials (gray) and the nuclear fission products, blue: lead, orange: Caesium, green: barium

It can be seen that the spectral region between 360 nm and 370 nm only contains the lead peaks at 363.8 nm and 367.9 nm, which can therefore be used to detect lead. For barium and cesium the spectral region from 640 nm to 730 nm can be used. As was shown by Gaona et al. [15] within this region calcium lines may be present. But for the cesium peaks at 697.3 nm and the barium one at 705.7 nm there are no interferences with calcium emission lines listed in the NIST atomic database [18] and therefore these peaks can be used to distinguish barium and cesium from the investigated background. As described by Lang et al. for cesium a strong peak at 894.4 nm could also be used for identification.

Quantitative LIBS Analysis

Determination of ratios of different atomic species within a sample can be done by calibration using different peaks from known samples. LIBS spectra were acquired for different mass percent of $PbCl_2$ in KBr samples (see Figure 5 left). The lead peaks around 360 nm can be clearly seen, and are rising for higher amounts of $PbCl_2$. From the ratio of the peak heights for lead around 368.4 nm and the potassium peak around 693.9 nm a calibration curve was extracted, shown in Figure 5 right. This curve serves for the determination of the relative quantities of lead and potassium.

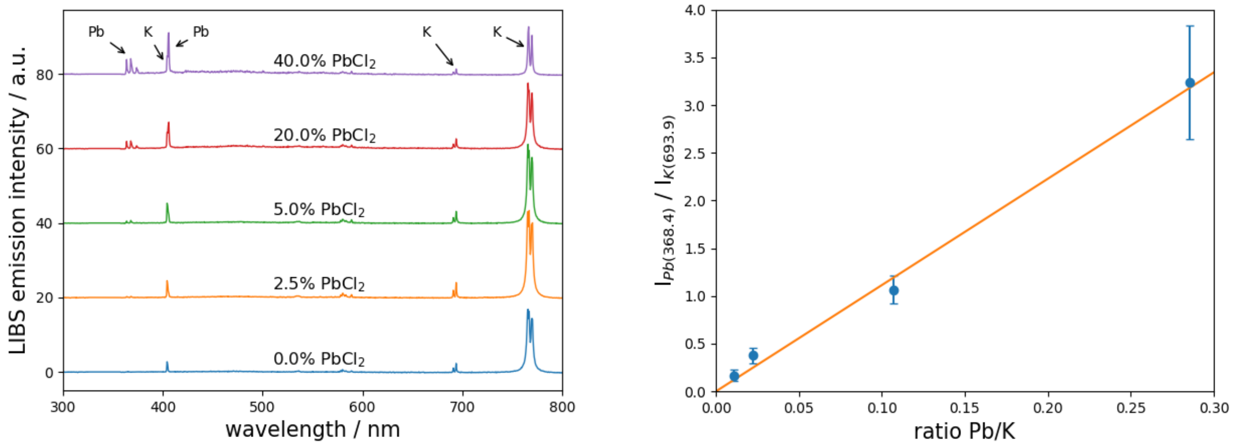

Figure 5: LIBS Spectra of samples with different ratios of $PbCl_2$ in KBr (left). Ratio of the peak hights of the lead peak at 368.4nm divided by the Potasium peak at 693.9nm over the ratio of the number of lead and Potassium atoms (right). The orange line is a linear fit that might be used to determine the lead contant of comparable samples.

For the smallest measured lead concentration, namely 2.5 % $PbCl_2$ in KBr corresponding to approximately 4000 ppm lead atoms, the signal to noise ratio of the peak at 368.4 nm is approximately 10 and shows that this and even lower concentration of lead can be detected at a 5 cm distance using this setup. Further investigations are necessary to determine the limit of detection more accurately for lead and for the other nuclear fission products.

Summary and Outlook

In this paper, investigations on the utilization of a compact LIBS setup to determine nuclear fission products and different background materials are presented. Here, it is demonstrated that the spectra of the nuclear fission products barium, cesium and lead exhibit spectral regions where they can be distinguished from different background materials. The region from 360 nm to 370 nm can be used to detect lead and the spectral region from 640 nm to 730 nm can be used to detect barium and cesium. Furthermore, a detection limit of 4000 ppm lead was determined for the presented system by measuring the peak at 368.4 nm with a signal to noise ratio of 10. Additionally, a calibration curve was extracted, which can be used to determine the lead content in a KBr sample.

One of the next steps is the implementation of different self-optimizing algorithms, i.e. artificial neural networks or support vector machines, to automatically distinguish different spectra acquired for various samples. More advanced algorithms can be used for quantitative analysis of these materials. Furthermore, choosing a specific wavelength range enables the utilization of a more adapted spectrometer. Thereby, the spectral resolution of the system can be increased to enhance the possibility to distinguish isotopes of materials with small isotope shifts. In another approach a high sensitivity spectrometer can be chosen leading to a better signal to noise ratio, and thereby lower limits of detection may be achieved. Additionally an optimized beam profile of the laser may possibly enhance the signal to noise ratio lowering the limit of detection for nuclear fission products within different background materials of a few 100 ppm and below.

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