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Applications of Portable LIBS for Actinide Analysis

Ashwin P. Rao

Air Force Institute of Technology

John D. Auxier II

Los Alamos National Laboratory

Dung Vu

Los Alamos National Laboratory

Michael B. Shattan

Air Force Institute of Technology

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Applications of portable LIBS for actinide analysis

Ashwin P. Rao¹, John D. Auxier II², Dung M. Vu², Michael B. Shattan¹

1. Air Force Institute of Technology, 2950 Hobson Way, WPAFB OH, USA, 45433

2. C-AAC, Los Alamos National Laboratory, Los Alamos NM, USA, 87545

ashwin.rao@afit.edu

Abstract: A portable LIBS device was used for rapid elemental impurity analysis of plutonium alloys. This device demonstrates the potential for fast, accurate *in-situ* chemical analysis and could significantly reduce the fabrication time of plutonium alloys. © 2020 The Author(s)

1. Introduction

Laser-induced breakdown spectroscopy (LIBS) is a versatile diagnostic technique which has garnered significant interest in the nuclear science community. Recent studies have proven the use of the technique in various nuclear applications, including analysis of surrogate nuclear debris [1], detection of rare earth metals in uranium matrices [2], and elemental impurity analysis of plutonium surrogate alloys [3]. Portable LIBS systems have the potential to conduct rapid chemical analysis in constrained environments, such as a glove box. This capability has attracted the attention of actinide chemists involved in the manufacture and analysis of plutonium alloys. In order for Pu alloys to be used in nuclear defense and security related applications, they must meet a specific set of chemical composition standards. The process of producing Pu alloys involves the careful monitoring of concentrations of dopant elements such as gallium, which is used to stabilize plutonium metal in the delta phase [4]. Additionally, the presence of impurity elements such as iron and aluminium are monitored; the presence of these elements affects the homogeneity and properties of the alloy [5]. Furthermore, since LIBS has been used in analysis of uranium compounds, this technique could also be used to monitor the in-growth of decay products such as U-235, U-236 and Am-241. Such a capability could allow for rapid chronometric analysis of Pu samples, which would greatly benefit the nuclear forensics community. This study presents the results of an initial analysis of Pu spectra using a hand-held LIBS device to identify elemental impurities in the Pu alloys. Chemometric techniques are used to distinguish between alloyed and non-alloyed Pu metal samples.

2. Methodology and Results

A SciAps Z300, shown in Fig. 1, was used to acquire spectral emissions from multiple locations on 7 different Pu alloy samples. The automatic beam rastering feature was used to collect data at 16 different points, ablating each point four times. The average of the 64 recorded spectra was saved for each sample location. One example



Fig. 1. SciAps Z300 portable LIBS device.

spectra is shown in Fig. 2, with the inset plot revealing the region where two subtle Ga emission lines appear. The Ga lines are often dwarfed by the more intense Pu emissions, and are not always well resolved in the spectrum. As a result, it is necessary to implement chemometric techniques to distinguish these data points and conduct a proper analysis of the complex Pu spectrum. Rapid identification and quantification of Ga content in Pu alloys would greatly benefit the production process by yielding metallurgical information about the sample, such as phase of the Pu and uniformity of the distributed Ga. 17 different spectra from 7 different Pu alloy samples

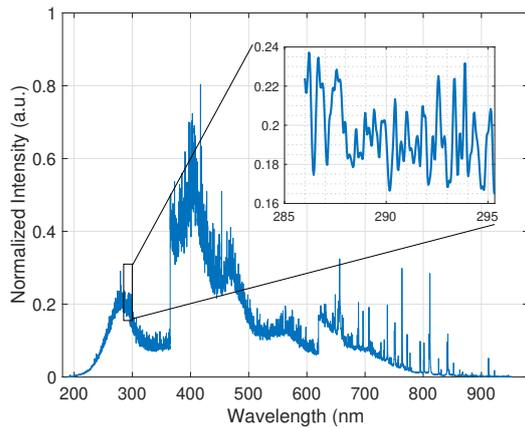


Fig. 2. Overlaid Pu spectra of all samples

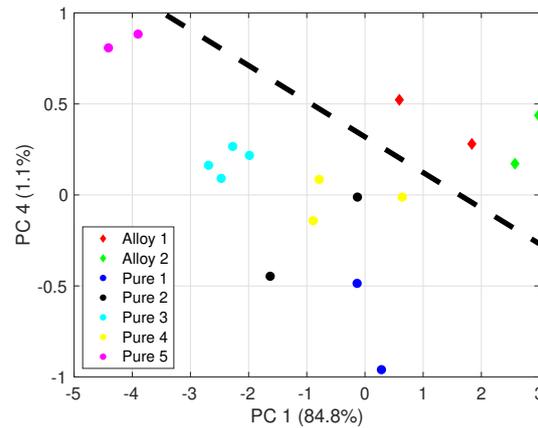


Fig. 3. PCA scores plot of sample set

were analyzed using Principle component analysis (PCA) to classify the sample set. This Multivariate analysis (MVA) technique is commonly used in analysis of LIBS spectra, and was chosen to reduce the dimensions of the complicated Pu spectra for more efficient analysis. PCA scores values, shown in Fig. 3, revealed the clustering of the different spectra by the appropriate Pu sample the spectra was recorded from; a slight overlapping was seen between the spectra from unalloyed samples 1 and 4, indicating similarities in chemical composition. The data points corresponding to the two alloyed samples are significantly separated from the unalloyed sample points, indicating that this technique can be coupled to the hand-held device for rapid *in-situ* classification of Pu metal. The dashed line in Fig. 3 visually demonstrates the separation between the alloyed and unalloyed sample areas, indicating the potential for rapid sample classification using this technique. Despite being "physics-blind," this basic PCA routine was able to resolve the emission line intensity differences between the pure Pu and alloyed Pu metals; this result indicates that a PCA routine could be used to efficiently detect and quantify the presence of dopant and impurity elements in a Pu metal matrix in further studies.

This novel analytical technique presents several advantages to the Pu fabrication community as well as those involved in metallurgical Quality assurance/quality control (QA/QC). Traditional chemical analysis techniques, such as mass spectrometry methods, involve significant sample preparation procedures and require metal samples to be transferred between labs. These steps hinder the production process by adding large amounts of time to the overall analysis timeline. Even existing systems which don't require extensive sample preparation, such as X-ray fluorescence (XRF), are deficient when it comes to detection of lighter metals. Portable XRF devices may not be able to induce fluorescence in gallium. Additionally, these devices must be tuned to identify a particular metal, whereas multi-elemental analysis can be conducted from one LIBS spectra. The portable LIBS device, with its speed, versatility and compact size provides several possible solutions to current problems in the Pu manufacturing process and could help ramp up the production of Pu alloys for the defense community.

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