



HAL
open science

Identification of materials by LIBS technique applied to nuclear facilities decommissioning

N. Coulon, D. L'Hermite, C. Quere

► To cite this version:

N. Coulon, D. L'Hermite, C. Quere. Identification of materials by LIBS technique applied to nuclear facilities decommissioning. DEM 2018 - Dismantling Challenges: Industrial Reality, Prospects and Feedback Experience, Oct 2018, Avignon, France. <hal-02415492>

HAL Id: hal-02415492

<https://hal.science/hal-02415492v1>

Submitted on 17 Dec 2019

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



HAL Authorization

Identification of materials by LIBS technique applied to nuclear facilities decommissioning

Nadine Coulon^{1*}, Daniel L'Hermite¹, Céline Quéré¹

¹Den – Service d'Etudes Analytiques et de Réactivité des Surfaces (SEARS), CEA, Université Paris-Saclay, F-91191, Gif sur Yvette, France

*Main Author: nadine.coulon@cea.fr

Abstract

The LIBS (Laser Induced Breakdown Spectroscopy) technique allows *in situ* chemical analysis in hostile environments. For the decommissioning of nuclear installations, this technology can be used for waste sorting or to carry out chemical inventories. We have developed a LIBS instrument for remote measurements. In order to achieve future on-site measurements, protocols and data treatments have been perfected.

KEYWORDS: LIBS, decommissioning, *in situ* characterization, material identification

Introduction

The chemical characterization of materials in nuclear facilities under decommissioning, and more generally of nuclear wastes, is currently carried out on samples in laboratory. The use of *in situ* chemical analysis techniques has to be preferred in order to reduce costs, delays, doses and nuisances suffered by operators but also to optimize waste management.

Because of its all-optical character and its capability to perform quick measurements without sample preparation, LIBS technique is very well suited to *in situ* analysis in hostile environment.

We have developed a LIBS device dedicated to dismantling purpose, including a portable probe connected both to the laser and to the spectrometer by two 15m length optical fibers (Figure 1).



Figure 1: LIBS prototype dedicated to nuclear facilities dismantling

The advantage of this set-up is that the electronic part, most sensitive to ionizing radiations (computer laser, synchronization electronics), is located out of the radioactive zone. The probe containing only optical elements is relatively small and can easily be put in contact with the material to be characterized into the radioactive area.

In this paper, two examples of implementation study of this instrument are described: *in situ* identification of the nature of wastes, and discrimination steel / cast iron constituting the thermal shield of a reactor.

***In situ* identification of waste nature for safe storage**

In the framework of nuclear facilities dismantling waste management is a key concern. For safety reasons, long-term storage of waste must be done very rigorously in order to avoid any possible chemical reactions between the wastes and the concrete. This is why, in France, some materials are prohibited or restricted (aluminum, magnesium, zirconium, zinc, brass and galvanized steel ...). CEA has decided to evaluate LIBS technology for *in situ* identification of various wastes stored since a long time in some of these nuclear facilities.

Because operators are supposed to be non-LIBS expert, the LIBS apparatus needs to integrate a specific data treatment for automatic identification of the nature of the waste. We have developed an automatic recognition protocol by acquiring a lot of spectra (900) on various pieces of metals (36) representative of nuclear facility materials. The recognition is based on the comparison of emission lines intensities to threshold values adjusted to obtain the best identification rate. The overall result is displayed as a confusion matrix (See table 1). "Aluminum", "Magnesium", "Zirconium", "Zinc", "Brass", "Galvanized steel" are the material to be identified because of safe storage constraints, "Others" refers to materials that do not induce storage difficulty.

Identified \ Reals	Aluminum	Magnesium	Zirconium	Zinc	Brass	Galvanized steel	Others
Aluminum	126	0	0	0	0	0	0
Magnesium	0	46	0	0	0	0	5
Zirconium	0	0	26	0	0	0	0
Zinc	0	0	0	103	0	0	0
Brass	0	0	0	0	129	0	0
Galvanized steel	0	0	0	0	0	51	0
Others	0	0	0	0	0	0	510

Table 1: Confusion matrix

In this evaluation of performances, only 5 measurements give a wrong result. These wrong results have been attributed to very low LIBS signal due to an incorrect positioning of the LIBS probe. So we have improved the data treatment in order to detect automatically the low signal spectra and to suggest to the operator restarting a measurement.

In perspective, the LIBS system including the automatic identification data treatment has to be tested in more realistic conditions.

Steel / cast iron discrimination

As part of the dismantling of the G1 reactor located on the CEA Marcoule site, the CEA has to confirm the nature of material constituting the reactor heat shield. The objective is to realize *in situ* measurements to discriminate between cast iron (more than 2% carbon content) and steel (less than 2% carbon content). As the carbon emission lines are interfered by many strong iron emission lines, we have decided to detect a carbon line in the UV range, not allowing the use of 15 m length optical fiber. A compact spectrometer was attached to the probe (see Figure 1), in order to reduce the length of the optical fiber. The reactor heat shield has an oxide layer on the surface. An accumulation of laser shots is necessary to remove the oxide layer and reach the substrate. A measurement protocol was therefore developed using cast iron and steel samples with different surface state (more or less oxidized).

From LIBS spectra obtained with these samples we have determined a threshold value of carbon line intensity, corresponding to 2% carbon concentration. When the intensity is above the threshold value, the sample is considered to be a cast iron and below the threshold value as a steel.

These laboratory tests made it possible to establish a LIBS data processing adapted to steel/cast iron discrimination, as well as measurement protocol, in particular concerning the number of laser shots needed to remove the oxide layer. *In situ* measurements on G1 reactor are scheduled for the first half of 2018.

Conclusion

In these two examples of LIBS technology implementation, a preparation phase was carried out, in particular for the development of signal processing. This phase has showed that under laboratory conditions, the LIBS instrument can respond to the needs of *in situ* material identification. Now it is necessary to evaluate the performance of this technology in more realistic conditions and possibly on site.