2024 International Nuclear Atlantic Conference – INAC 2024
Escola de Guerra Naval, Rio de Janeiro, RJ, May 6 - 10, 2024

ISBN: 978-65-594-1256-3

Radioactive Waste Sampling by Laser Ablation Sample Transfer (LAST)

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1. Introduction

The appropriate handling and storing of radioactive waste are important for the protection and safety of human lives and to avoid environmental impacts. For this purpose, correct characterization of the contaminants is fundamental as it impacts the storage methods and the personal protective equipment necessary to handle it. Said characterization pertains to determining the chemical and physical characteristics of the desired material, along with its radioactivity [1].

The most common method is scrubbing, in which an operator utilizes a porous material, such as a paper filter, to rub against the surface of the target whilst applying pressure. The material becomes a sample, which is then meticulously analyzed in search of specific radionuclides [2].

This method, however, is unable to consistently ascertain the amount of contaminants present in the original material as it heavily relies on the dexterity of the operator, the amount of pressure applied, the texture of the material and the chemical interactions between the radionuclides and the material's surface.

This project seeks to develop a new method of sampling metallic surfaces for radiological contaminants through laser ablation.

Laser ablation is a process in which the energy emitted by a laser is focused into a small area. This concentration of energy deposition is capable of making the material around the center of the laser-affected area heat up above its point of vaporization. Consequently, part of the material's surface is removed in the form of gas, plasma, liquid or small solid molecules. These can then be deposited in another surface such as a microscope glass slide [3] in a process often referred to as Laser Ablation Sample Transfer (LAST). The process can be visualized in Fig. 1.

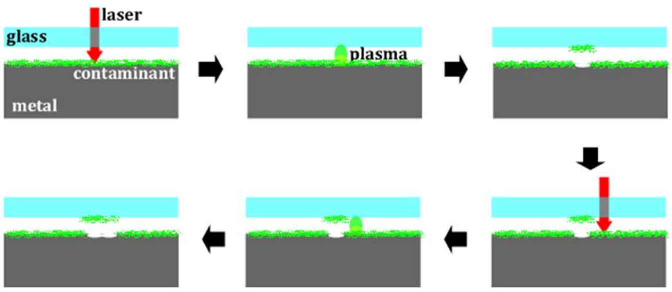

Figure 1: Visualization of the laser ablation sample transfer process on a glass substrate.

We expect that laser ablation could prove itself as a more reliably consistent method for sampling of contaminated objects since, in principle, the removal of a thin top layer of the surface of the material should remove the contaminants at fixed proportions. This process would be more reliant on physical properties of equipment and experimental setup than the dexterity of the human operator. Therefore, the aim of the present study is to evaluate the viability of the LAST technique and its potential.

2. Methodology

For this experiment three Cobalt-60 radioactive samples were used. These samples were created at the Radioactive Waste Management Department (GRR) of IPEN. They consist of 1 inch metallic disks about 0.5 mm thick contaminated with Cobalt-60 through the process of electrodeposition and are shown in Fig. 2. The activity of these samples was verified through the use of an emission counter, a Ludlum model 2929 dual-channel scaler [4]. Before the measurement of each sample the equipment measured the local background radiation by counting the emissions for 5 minutes. Afterwards, the same amount of time was used to analyze each sample. The results of the equipment for background and sample activity are shown in Table I.

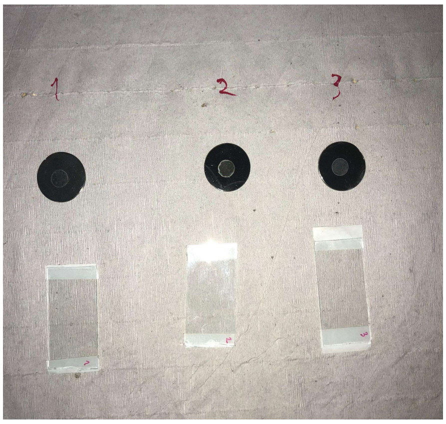

Figure 2: Photograph taken from the 3 circular samples and the corresponding microscope slides used as substrate for each of them. The darker areas on the center are where the electrodeposition took place.

These samples were held in place by a vacuum pump on a programmable mechanical arm along with the microscope slide which served as the substrate where the ablated material was deposited (Fig. 3). The substrate was maintained at a fixed distance from the sample by double sided tape placed on other two microscope slides glued to the translation stage, also by double sided tape.

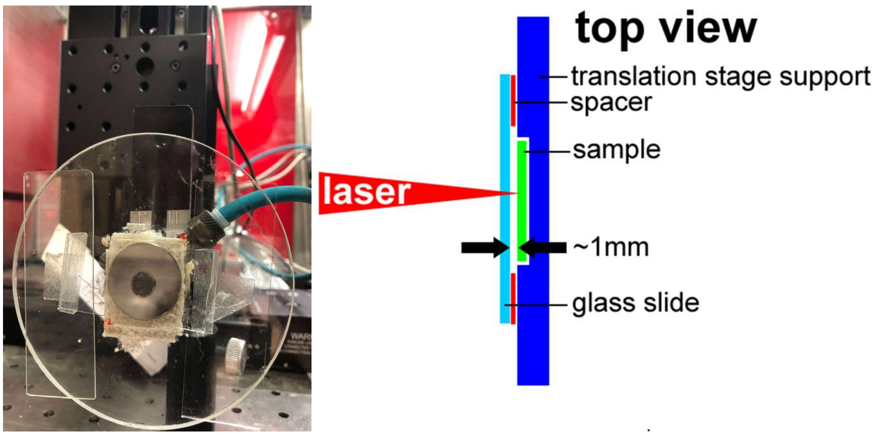

Figure 3: At left: Photograph taken from sample placed in the mechanical arm with corresponding substrate in place. Right: a schematic representation of sampling system where the spacer used was also a microscope glass slide.

The laser setup used consisted of a QUantel Ultra 100, Nd:YAG 1064 nm laser system lined up with a mirror to reflect the beam at a 90 degree angle, followed by a convergent lens which focused the laser at the sample. The focusing system promoted a focal spot of 0.3 mm and the pulse energy was set in order to result in a fluence of 10 J/cm^2 .

While the laser was active with a repetition rate of 20 Hz the translation stage moved at 6 mm/s in a direction perpendicular to the laser beam with a lateral step of 0.4 mm, the latter concept can be visualized in Fig. 4. This was done in order for the laser to ablate all of the surface of the sample i.e. these parameters were calculated so the overlap between shots is set at ~50% (of the radius), allowing for enough energy to be deposited throughout the surface.

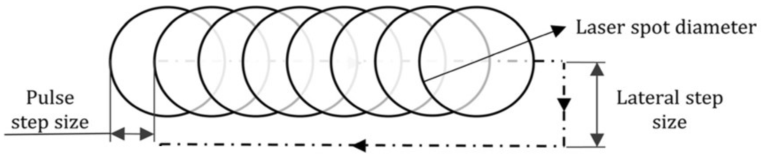

Figure 4: Visualization of laser affected area and lateral step. Figure altered from original created by Piccolo, L., et al. 2022 [5].

3. Results and Discussion

Firstly, the background radiation was measured, then the samples to determine their activity before ablation. Afterwards the samples were ablated (Fig. 5) and slides with transferred material were measured. All measurements were performed by the radiation detector and took 5 minutes. The results are shown in Table I and Table II.

Table I: Table presenting the results of the radiation count of the Ludlum model 2929 dual-channel scaler for background radiation and original sample activity before exposition to laser.

Measured Object	Alfa Count	Beta and Gamma Count
Background Measurement 1	35	607
Background Measurement 2	38	660
Background Measurement 3	32	629
Sample 1	34	4882
Sample 2	46	6574
Sample 3	35	6163

After the ablation process the microscope slides were measured by the radioactive emission counter and, due to the alfa emissions of the samples and the slides being similar to the background radiation, only beta and gamma emissions were considered. Furthermore since the background radiation fluctuated significantly over time, an average value, 632, was calculated and used to estimate the rate of transfer in accordance to the following Eq. 1. The results are shown in Table II.

$$Rate\ of\ Transfer = \frac{Total\ Emission\ of\ Slide - Average\ Background\ Radiation}{Total\ Emissions\ of\ Sample\ Before\ Ablation - Average\ Background\ Radiation} \quad (1)$$

Figure 5: A representative photograph of an irradiated sample. The patterning promoted by the laser raster is visible on the sample, and the brownish spot at the center of the glass slide is also perceivable, indicating the metal deposition.

Table II: Table showing the emission counts of the Ludlum model 2929 dual-channel scaler for the microscope slides and the corresponding estimated rate of transfer of contaminants.

Slide Number	Nominal Beta-Gamma Counts	Rate of Transfer	Z- Test Value
1	901	6,33%	-1,8
2	915	4,76%	0,1
3	826	3,51%	1,7

The average value of the transfer rates is (4.9 ± 0.8) %, with uncertainty calculated through the standard deviation of the mean, and the value for Z-tests were all in the [-2,2] interval, showing that the proposal seems viable but will require further improvement for the development of the technique.

4. Conclusions

In summary, the three Cobalt-60 circular samples were ablated and have transferred part of their contaminants to the microscope sides placed in front of them with distance equivalent to the width of a common microscope slide. The transference rate of contaminants seems to have varied around the average value between 0 and 2 times the standard deviation of the mean, with Z-test value between [-2,2]. Thus, it can be stated that the technique as a sampling tool needs improvement, however the viability of the proposal has been demonstrated.

Acknowledgements

The authors acknowledge the support given by the Brazilian National Council for Scientific and Technological Development – CNPq, grants 465763/2014-6, 407077/2023-5 and Sisfóton 440228/2021-2.

References

[1] R. Burel, J. L. G. Gómez, J. J. Kelly, *Strategy and Methodology for Radioactive Waste Characterization*, International Atomic Energy Agency, Vienna, Austria (2007).
[2] C. P. Lichtenwalner, “Evaluation of wipe sampling procedures and elemental surface contamination”, *American Industrial Hygiene Association Journal*, vol. 53, pp. 657-659 (1992).
[3] A. Castelo, D. Nieto, C. Bao, M. T. Flores-Arias, M.V. Pérez, C. Gómez-Reino, C. López-Gascón, G. F. de la Fuente, “Laser backwriting process on glass via ablation of metal targets”, *Optics Communications*, vol. 273, pp.193-199 (2007).
[4] “Manual for the Detector”, https://ludlums.com/images/product_manuals/M2929.pdf (2020).
[5] L. Piccolo, Z. Wang, G. Lucchetta, M. Shen, D. Masato, “Ultrafast Laser Texturing of Stainless Steel in Water and Air Environment”, *Lasers in Manufacturing and Materials Processing*, vol. 9, pp. 434-453 (2022).