How Digital Autoradiography Technique can be useful for D&D projects ?

Pascal FICHET*, Raphaël HAUDEBOURG

CEA SACLAY, DEN/DANS/DPC/SEARS/LASE PC
171, 91191 Gif Sur Yvette, *France*

**corresponding author: pascal.fichet@cea.fr*

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ABSTRACT

For many stakeholders, it is obvious that radiological characterization is essential for D&D projects and all of them have its own specificities. Different types of techniques have been developed for characterizations not only by destructive methods but also by in situ investigations. Since 2010, the LASE laboratory, in charge of characterizations of different radionuclides that can be found in nuclear wastes, has developed the Digital Autoradiography (DA) technique [1]. Today, DA technique is in routine use commercially for biological researches to obtain images of labelled molecules containing for example H-3, C-14, I-129, … i.e. radionuclides difficult to measure for D&D projects. The needs to investigate these hard to observe radionuclides are one of the R&D subjects for D&D proposed recently and highlighted by OECD [2]. Up to now the R&D projects concerning Digital Autoradiography has been focused on mapping process in buildings under dismantling process and also on sampling procedures being essential before any destructive analysis. New developments are still going on to develop the technique and to modify technologies coming from biological researches [3].

I. Introduction

Measurements are essential for decommissioning nuclear facilities. For stakeholders in charge of D&D, analysis must be reliable fast and if possible cost effective. In that purpose Digital Autoradiography has been proposed as an alternative to investigate materials. Although alpha [4] and gamma camera [5] were largely described in the literature, the in situ beta emission analysis remains a challenge.

Coming from medical researches, the Digital Autoradiography technique was completely reviewed to fit different problems of investigation for D&D projects. Different applications have been developed for (i) tritium (and more generally hard to detect radionuclides mapping) analysis in facilities (ii) for in situ characterization of nuclear wastes.

Researches coming from oil explorations have been developed from several years. These skills were optimized recently to produce a GIS (Geographical Information System) that can be connected to data coming from gamma, alpha dose level, and here autoradiography to produce a contamination mapping (Kartotrak software, Geovariances company).

For characterization of nuclear wastes with destructive techniques, the requirement of solid sample before digestion process is typically around 1 g. For sampling optimization of quite large nuclear wastes that must be characterized, digital autoradiography can be very useful.

II. Digital Autoradiography technique

The DA technique enables visual and 2 dimensional detection of high and low energy radiations including hard detecting radionuclides such as H-3, C-14 and alphas. The radiation data is collected on specially manufactured screens (known as phosphor screen, photo imaging plate) which have photostimulable crystals, for example BaFBr:Eu^{2+} phosphor crystals, on a support layer. The crystals may be covered with a protecting layer, such as in MS screens (PerkinElmer, GE Healthcare), or the crystals may be uncovered, such as in TR (**Figure 1**) screens (PerkinElmer, GE Healthcare).

Figure 1: Phosphor screen used in the study; left: sensitive side, right: back side; type: TR, commercialized by Perkin Elmer; size: 12.5 x 25 cm²

The protecting layer protects the MS screens from scratches and moistures and they can be cleaned with a commercial cleaning solution. Since the TR screens does not have protection layer on the crystals and water destroys the crystals, these screens require especially careful handling and dry measurement conditions. But it was observed that for characterization for dismantling facilities the screens are strong enough. Thus, due to the lack of protection layer, TR screens can detect low beta energy such as H-3 (**Figure 2**, example of an image representing some H-3 fixed in a wood piece).

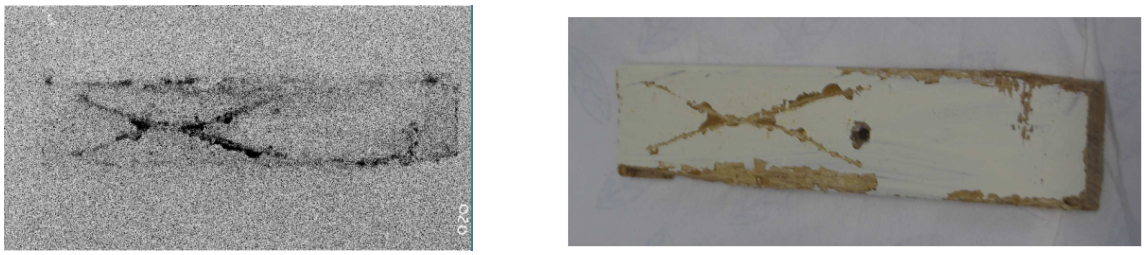

Figure 2: Tritium measured on a piece of wood

The screen for digital autoradiography is placed in close contact with the radiation source and different exposure times are used depending on the source of radiation and its activity. The mechanism for the collection of the radiation data by the DA screens is based on the ionization of Eu^{2+} to metastable Eu^{3+} in the BaFBr:Eu^{2+} phosphor crystals. The radiation energy is stored in bromine vacancies of the phosphor crystals until they are exposed to stimulating laser light at 633 nm causing production of unstable Eu^{3+}. The unstable Eu^{3+} goes back to the initial Eu^{2+} state by releasing photons at 390 nm which are collected by a photomultiplier tube (PMT). The stimulating laser in Cyclone Plus Scanner (PerkinElmer) can scan the screens with different resolutions varying from 150 dpi to 600 dpi. The corresponding pixel sizes are 170 microns and 42 microns, respectively which are of course very low values for the decommissioning requirements but which corresponds to the actual developments of the technique. The Cyclone Plus Scanner converts the scanned information into Digital Light Unit (DLU) in each pixel and results are

visually expressed in OptiQuant software as a 2D image. Contrasts obtained in the image correspond to different values of radioactivity. The file obtained is in TIFF format. The scanned images can be expressed in color or in intensifying gray color scaling from white to black. One of the main advantages of the DA screens is their re-usability which decreases the cost of the studies and production of wastes. After the DA screen has been scanned, it is exposed to strong white light which erases the remaining radiation information from the screen after which the initialized screen can be re-used. Only a couple of minutes are required to initialize the screens. The actual erasure time will vary with parameters such as the energy of the original isotope, the original exposure time, and the intensity of the white light source. The screens can be re-used hundreds of times if they are handled with care.

III. Digital Autoradiography for radionuclides mapping

By using batches (typically 20 screens) of reusable screens (**Figure 1**) several times on different locations, it is possible after data processing and Kartotrak software use, to obtain reliable two dimensional mappings of radioactivity traces for decommissioning nuclear laboratories. The batches of reusable screens were used in different locations providing up to several thousands of results in the ground of a laboratory with around 250 m^2 area. **Figure 3** shows the raw image of the results of the autoradiography measurements. The black rectangle on the border represents the boarders of the laboratory.

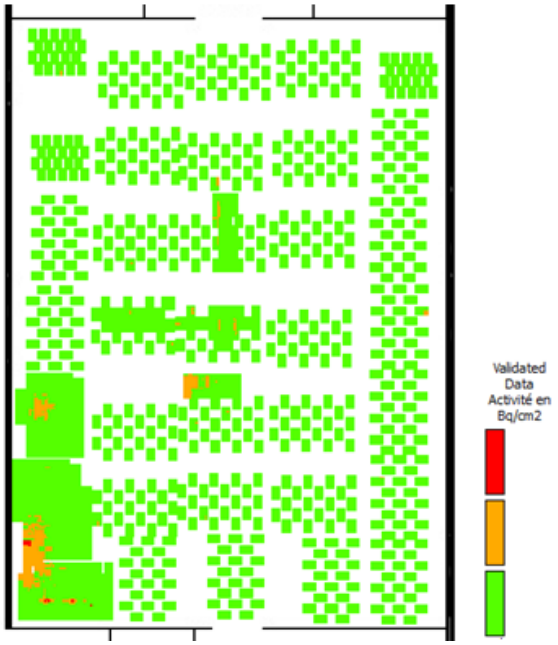

Figure 3: Measurements obtained with thousand autoradiography images on the ground of a laboratory (green color represents no radioactivity, orange and red shows locations where tritium is detected by autoradiography)

The Kartotrak software is able to calculate, by using a geostatistic method, with the raw data a variogram function which represents the effects of all the neighbors to calculate the value of the radioactivity of a particular point. With the variogram function, Kartotrak can calculate the kriging image (**Figure 4** shows the traces of tritium on the concrete surface). Moreover the variance of the kriging can also be calculated to confirm the reliability of the results. By assuming that contamination is rather large in decommissioning facilities, only 20% of the measurements on the ground are necessary to obtain the contamination of the whole surface with a minimum of risk. This allows calculating contaminated surfaces in order to provide accurate results to work on D&D scenario to minimize risks and costs.

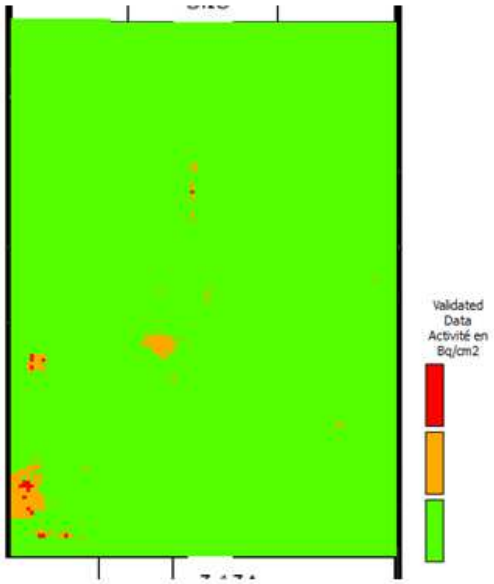

Figure 4: Mapping of the whole surface of the laboratory after kriging calculations.

The combination of digital autoradiography for beta measurements and geostatistical calculation can be a very efficient way to evaluate remaining radioactivity for decommissioning nuclear facilities.

IV. Digital Autoradiography for sampling processes

Apart from these developments on mapping results, Digital Autoradiography has also been developed for optimization of samplings required for destructive analysis. Destructive analyses are mandatory to provide activity of nuclear wastes in Bq/g with high accuracy linked with a reliable uncertainty. These values in Bq/g are essential to obtain an optimal classification of wastes needed for D&D projects to ensure a good safety of the nuclear wastes repository sites.

High accurate techniques providing analysis of radionuclides requires the digestion of very small amount of solids, muds, … (typically less than one g) that must be chosen and Digital Autoradiography can be very efficient for sampling processes. Hot spots can be observed on DA images providing very significant samples (see **Figure 2** for H-3). The possibility to provide images of radioactivity is a great advantage in many applications that have been developed by LASE laboratory: wipes sorts, core investigations, contaminated and non-contaminated wastes….
In D&D projects, control of labile contamination by wipes is very common. Wipes can be deposit on a screen and images obtained can reveal traces of radioactivity (**Figure 5**). The Autoradiography is a rapid and cost effective technique that can be applied in situ to sort wipes.

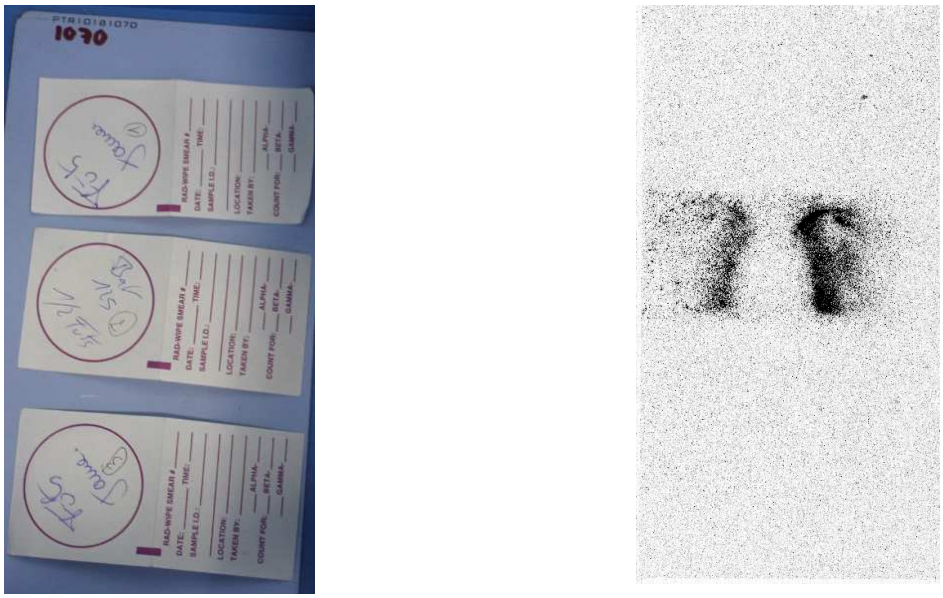

Figure 5: (a) several wipes on a film (b) only one contains a radioactive emitter

The efficiency of this sampling method can also be used to choose particular wastes. As an example, for trace of H-3 some different wastes can be studied on a screen. By comparing the real image of wastes in **Figure 6(a)** and the autoradiography image in **Figure 6b** it appears clearly that some pieces of wastes are contaminated but not all. Red arrows show the two contaminated pieces.

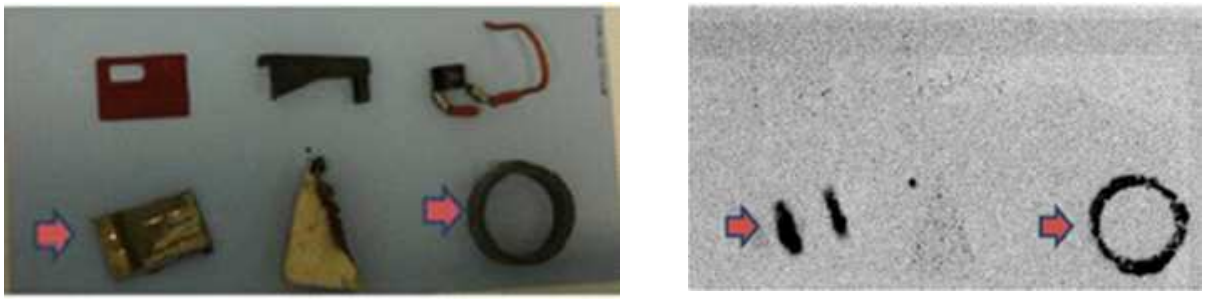

Figure 6: (a) different wastes (b) autoradiography result showing radioactivity for some only 2 pieces among six

V. Conclusion

The combination of Digital Autoradiography results and Kartotrak software providing geostatistical technique, was used to obtain characterization of the ground of a laboratory that hosted nuclear researches and that must be dismantled.

Autoradiography can be also a very efficient technique to sort different wastes and to provide an efficient and easy way for sampling. The sampling is essential for destructive methods requiring only 1g for digestion processes.

References

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