Radiological Mapping of Facilities under Dismantling Process by Digital Autoradiography – 15230

Pascal Fichet*, Florence Goutelard*, Raphael Haudebourg*
*CEA Saclay, DEN/DANS/DPC/SEARS/LASE, PC 171, 91191 Gif Sur Yvette, France,
pascal.fichet@cea.fr

ABSTRACT

In France as well as in other countries, some facilities that hosted nuclear researches must be demolished because safety standards have changed. In that context, radioactive waste management is a challenging task faced by existing nuclear power countries. Therefore, characterization is essential to classify wastes and to reduce costs. In many facilities, after the removal of glove boxes, fume cupboards and so on, the remaining materials on the floor, walls, and ceiling must be characterized. Gamma emitters can be detected remotely by commercial devices. However, analytical techniques to detect alpha and beta emitters in situ are generally not well suited for large area investigations. The Digital Autoradiography (DA) technique, initially dedicated to research in biology, was rerouted for final state characterization before facility demolition. It is based on the exposure of radio-sensitive phosphor screens on the materials to characterize. Phosphor screens, after a scanning process, provide a picture of the radioactivity, and are reusable several tens of times. This analytical technique is sensitive to all types of radioactivity, requires neither operators nor electricity during acquisition process, and produces only very few wastes. DA has already been tested in different facilities to obtain semi-quantitative values of C-14 and H-3 activities. Large amount of data gathered to cover the whole surface to be characterized. A Geographic Information System (GIS) associated with geostatistical method has been used to provide Radiological Mapping. This method to interpret Digital Autoradiography results has been proved very efficient to prepare remediation project.

INTRODUCTION

In French nuclear facilities under dismantlement, there is no threshold value under which wastes are considered as “non-nuclear”: all wastes are classified as “nuclear” and their activity must be measured for management purposes, which dramatically increases the amount of materials to characterize.

Whereas gamma emissions can be detected remotely by commercial devices, analytical techniques to detect alpha and beta emitters in situ are generally not well suited for large area investigations. The most common technique to investigate the presence low-energy beta emitters, such as H-3 and C-14, consists in in liquid scintillation counting of a wipe rubbed on the material to analyze. However, fixed radioactivity cannot be detected by this method, owing to its nature.

Digital Autoradiography (DA) appeared as a promising alternative to evaluate possible alpha and beta contamination in facilities to be dismantled. This technique was initially developed for biological research [1], to investigate localization of radioactively-labeled organic molecules in animals such as mice. Therefore, it has been designed to be sensitive to C-14 and H-3 and to provide high resolutions, but this on relatively small surfaces.

DA technique consists in two steps: first, exposure of radiosensitive screens (Figure 1) to irradiations, by putting the source in contact with the sensitive side of the screen for a given time; and then, scanning of the screen by an appropriate device to obtain a radiological mapping of the radioactive source (2D picture).

A screen consists of a flexible plastic backing coated with a phosphor matrix containing alkaline-earth halide grains (generally BaFBr) doped with radio excitable ions (generally Eu^{2+}). These ions are excited to a metastable state by incident energetic particles (α, β, γ/X) during the exposure step. Energy is thus stored in the screen until scanning step. In the scanner, the surface of the screen is swept by a laser, and metastable Europium ions are excited to a higher energy state (unstable), emitting quite immediately a blue photon, recorded by a Photo Multiplier Tube.

Several types of screens are commercially available, and known as MS (Multi Sensitive, with highest density of grains), SR (Super Resolute, with optimized repartition of grains), and TR (TRitium sensitive, without usual protective top layer, to allow penetration of low-energy β particles in the phosphor matrix). Screen sizes usually range around several hundred cm² (for instance size of TR film: 12.5 cm x 25.2 cm).

DA was therefore rerouted from biological research to nuclear facilities dismantling, expecting the following advantages:
- high sensitivity to H-3 and C-14 (and all the more so for higher-energy β and α)
- detection of non-labile radioactivity (contrary to wipe tests) [2, 3]
- neither electric supply nor operator required during exposure, which is crucial in dismantling context
- low costs and constraints of development and utilization (screens and detectors are commercially available [4, 5], screens are reusable several tens of times, no radioactive material has to be taken out and transported, very few waste production).
- possibility, subject to re-engineering, to decrease scan time (limiting step, especially with large amounts of screens) by willingly decreasing resolution (a typical resolution of 1 cm² would be more than enough for this application, while resolution for biological application is typically 1000 µm²).

DA was tested on particles collected after Fukushima accident to measure Cs-137 with activity lower than 1 Bq [6].

In this paper, attempts to perform quantitative measurements of H-3 and C-14 contamination on the concrete floors and walls of a facility under dismantlement, and corresponding results, will be presented. Efforts made since 2011 to develop a software tool to manage the very high amount of data produced by such a measurement campaign will be highlighted.

Fig.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

DESCRIPTION

In the development of Digital Autoradiography on dismantling sites, non-fixed radioactivity was first removed before any investigation by Digital Autoradiography in order to avoid potential contamination of screens, which were then deployed in batches of several tens. H-3 and C-14 investigation in this example was logically performed with TR screens, whose response was evaluated thanks to a preliminary calibration.

To characterize the radioactivity on floors and walls, screens were put down directly in close contact with the material, as shown on Figure 2 (screens on concrete). A statistical approach was chosen to investigate large surfaces (floors and walls): instead of fully covering the area with the corresponding number of screens (75 screens for a surface of approximately 2.5 m² in this example), a batch of only 20 screens was set within a specified grid drawn on the floor. Thus, overall scanning time was divided by 4, but large spots of contamination could not be missed.

Once deposited, screens were exposed for a chosen period of time (order of magnitude 24 hours), depending on the expected radionuclide(s) and activities, namely H-3 and C-14 in this example. No operator presence is required during the exposure time which can be a great advantage in terms of potential dose rate received.

At the end of the exposure, screens were withdrawn, and screen scanning was performed by the Cyclone Plus Device (Perkin Elmer), displaying the image of the radioactivity of the material underneath the screen.

Fig.2. Batch of films (20) deposited on the floor of a facility

DISCUSSION

The resulting signal, expressed in DLU (Digital Light Units) for each pixel of the image, is linear with activity and exposure time. The image corresponds in real size to the zone studied (Figure 2). In the basic software setup (greyscale, darker pixel for higher DLU), radioactive areas appear as black spots and background as homogenous grey (Figure 3).

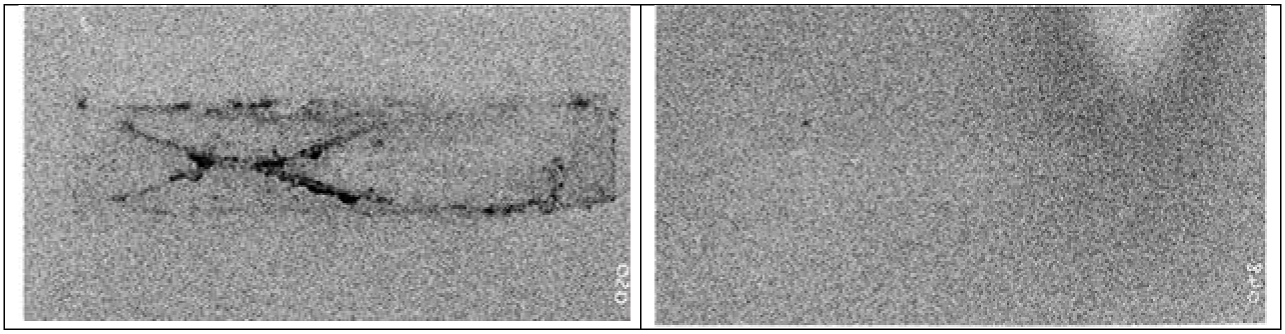

Fig. 3. Scan-of a screen exposed to a piece of wood containing 3000 Bq/g of H-3 (left), compared to background level (right)

The Digital Autoradiography technique developed herein provides numerous images of radioactivity that must be accurately linked to the geographical reality of the site, to locate radioactive spots. In the earliest developments of the method, radioactivity localization was enabled by printing the scans and positioning them in close contact from one to another in the facility, at the same place where corresponding screens were exposed. Figure 4 illustrates this attempt to represent a spot of C-14 at a real scale on a floor made of concrete. Radioactivity could be seen on the black parts on white background (i.e. great contrast, while low-contrasted, homogeneous gray images correspond to background activity). The scanner used for the screens optimizes systematically for each, the contrast between white and black. But it is clear with Figure 4 that some uniform color with black on the right top of the entire image does not represent necessarily radioactivity. To represent the actual radioactivity in a more realistic view, a software treatment process was required.

The different scans obtained with the screens were produced by the laser scanner with a high resolution (approximately 169 µm) to produce the image on Figure 4. Such images are not directly comparable to each other because conversion from raw signal in DLU to final image in grayscale is different for each file, depending on contrast optimization performed by the embedded software. To enable comprehensive radiological map with a consistent scale, it is therefore essential to process raw files and display all results with a dedicated software. Figure 5 displays the scheme of the calculation developed. Raw data in pixels of 169 x 169 μm^2 are merged (by summation) into elemental squares of 5 x 5 mm^2; this drastically decreases amount of data to process (divided by 30) and speeds up calculations, while keeping a satisfying resolution for the application. These squares are also associated with coordinates (X, Y, Z), to enable localization by the Geographical Information System.

Fig. 4. Image of C-14 (50 000 Bq/g) on the floor of a facility

LISA is a homemade software designed to automatically compare images of radioactivity obtained by the screens. The GIS connected with LISA, known as Kartotrak® software, was purchased from Geovariances implements geomatics optimization (see Figure 6) and optional geostatiscal methods.

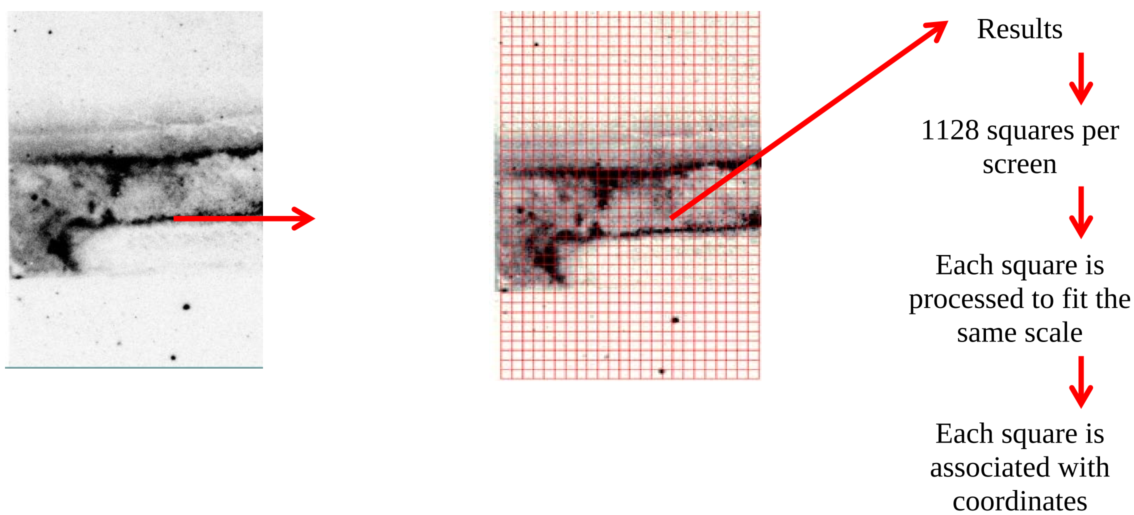

Fig. 5. Scheme to obtain a radiological mapping.

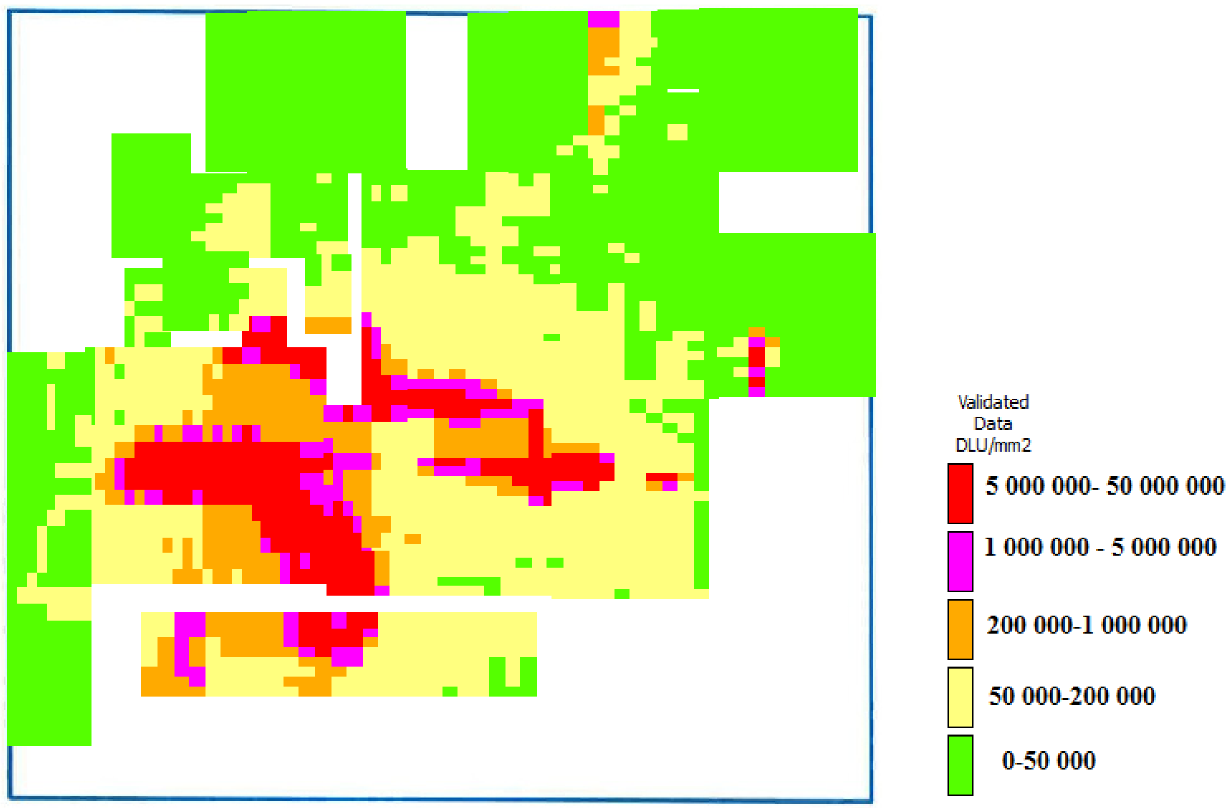

Fig. 6. All data in DLU obtained with the same scale.

Image displayed on Figure 6 shows same experimental results as Figure 4 but in a more realistic scale. All red squares represent the highest radioactivity whereas green color corresponds to area corresponding to background. Small hot spots on the right and at the top of the image can be also observed easily. Kartotrak® is also able to extrapolate by geostatistical methods non measured area. This work is under progress to truly estimate contamination in non-measured locations.

CONCLUSIONS

Digital autoradiography is a quantitative, non-destructive, and inexpensive method for the localization of radioactive contamination on different surfaces of facilities under dismantling. It is compliant with most requirements in such a context (very few constraints, minimal exposure of operators, neither production of wastes nor transport of radioactive materials). While traditional wipe tests for beta emitters characterization can only detect removable contamination, digital autoradiography detects both fixed and removable contamination. In addition, it provides 2D images enabling precise localization of the potential contamination. However, for a large area investigated, the number of images is huge, requiring a mapping method to compare results and also to extrapolate data without direct measurement. The use of a Geographic Information System (GIS) with a geostatistical method is a very efficient way to locate and compare the radioactive spots observed by the Digital Autoradiography Technique.

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