Digital Autoradiography technique an efficient tool for sampling procedure

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INTRODUCTION

Issues in the radioactive waste management are challenging tasks faced by nuclear power countries. These issues are a prime concern of the public and therefore of the control authorities called in France ANDRA and ASN.

In France, several nuclear facilities (laboratories, NPP) have been shut down few years ago and are now under dismantling. Physical, radiological and non-radiological characterization prior to dismantling is a key element of all decommissioning projects. After the disposal of different equipments (glove boxes etc.) and a cleaning-up step, laboratories for example are subject to decommissioning or demolition. In order to establish the working procedure of dismantling, an inventory on the radioactive level of the various materials and area is essential. Furthermore, correct and cost effective waste classification is crucial to define the appropriate final disposal repository.

Gamma cameras are the instruments commonly used for nondestructive analysis and to investigate the facilities potentially contaminated with gamma radionuclides (1). But, when suspected contamination concerns pure beta emitters such as H-3 or C-14 or alpha emitters and more generally radionuclides difficult to measure, researches are still going on to provide in-situ suitable method. At CEA (French Alternative Energies and Atomic Energy Commission, France), some facilities, where beta or alpha emitters have been handled, are intended to be decommissioned or even demolished. Whatever the end-state defined, the radioactive level inventory of the building must be investigated. In order to fulfil the request for laboratories of several hundreds of square meter, the analytical technique of Digital Autoradiography (DA) well known for biological researches (2) has been developed (3-5). Easy to handle (4, 6), it has been appeared as an efficient technique to localize precisely fixed or non-fixed contamination on various materials (7). For all decommissioning facilities regardless of their size and use, after historical site assessment, targeted characterization are required on different matrices. This requires techniques being able to improve sampling protocol. Sampling is highly important in analytical laboratories performing digestion processes generally requiring less than 1 g of material that must be characterized.

Digital Autoradiography appears as an adequate method to point out the radioactive spots where sample can be collected. Studies on the specificity of the DA technique have also been developed recently to improve the potentialities of this in situ technique.

DESCRIPTION OF THE ACTUAL WORK

Digital Autoradiography (DA) Technique

The DA technique has been developed for in situ analysis of dismantling facilities. This analytical technique uses specific films (Fig 1.), also called screens or photostimulable imaging plates, which are sensitive to all types of radiations (alpha, beta, and gamma and even X). Size of films used in this work is 12.5 x 25 cm². These films contain crystals (blue part of Fig 1) composed with europium traces. Interaction with the ionizing radiation leads the europium in a metastable state. In this way the information is kept in memory in the film material up to several days. A laser scanning (Cyclone® Perkin Elmer or Typhoon GE Healthcare) is then used to locally excite the europium to an upper state which then spontaneously comes back to its stable state releasing a UV photon. The resolution of the resulting image of radioactivity is a combination between laser spot and europium grain and a precision as low as few tens of microns can be typically reached. Thus, the surface investigated is very small (of the order of 100 µm) and even small spots of radioactivity can be observed. Homogeneity of radioactive source currently used in analytical laboratories can be evaluated with this technique. Figure 2 shows the homogeneity of two radioactive sources (Cl-36 and Sr-90).

After direct exposure to intense light, DA screens can be reused several times. This is very interesting for a sampling protocol in a facility under dismantling.

Films are very robust and can also be used on curvy surfaces, as they are relatively flexible.

Fig 1. Typical film used for autoradiography

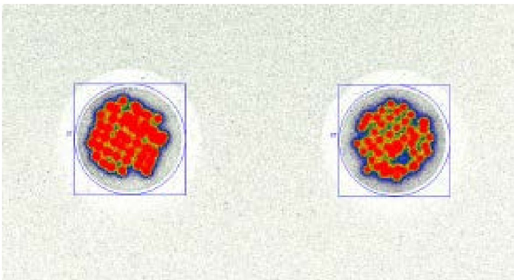

Fig 2: Homogeneity of two sources Cl-36 (left) and Sr-90 (right) showed by an exposure on autoradiographic screen.

Methodology developed for sampling investigation

For H-3 analysis at trace level with no labile radioactivity, wastes must be deposited in close contact to autoradiographic screen called TR. These screen do not possess any plastic cover that could absorb the low energy beta emission of tritium.

Otherwise for higher amount of radioactivity plastic covers can be an effective solution to avoid any contamination on screen. One of the drawbacks of the method, at that research level, remains in the separation between all types of radioactivity. The exact identification of the radionuclides investigated can be difficult although some researches are developed to try to identify more clearly the different radionuclides investigated. It has been shown that the superposition of several films one on top of each other can allow differentiation between different radionuclides (5).

For different investigations on sampling investigation using DA technique screen stacking technique (5) has been used directly on wastes (Figure 3). This technique was used to improve the specificity of the DA screens. By using a single screen it is totally impossible to identify particular radionuclide, however with ten DA screens and by comparing the absorption of different radionuclide in different screen it was proven that it can represent a sort of fingerprint. As an example the Cs-137 has its fingerprint shown in Figure 4. This ratios Sn+1/Sn, where Sn (Sn+1) is the signal recorded in screens n (n+1), is plotted as function of n. n is the number of the screen used in the stacking procedure.

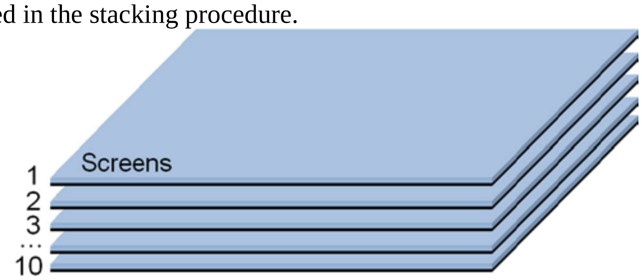

Fig 3: stacking technique used directly on wastes deposited on screen 1

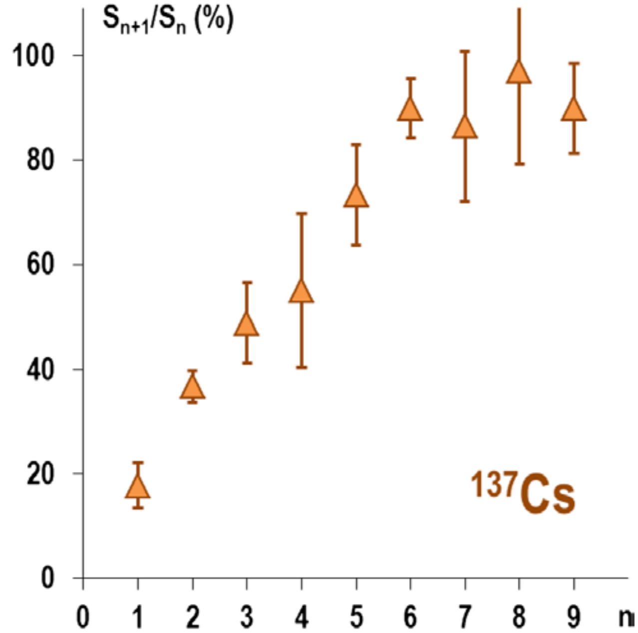

Fig 4: Fingerprint of Cs-137 by the stacking technique method

RESULTS FOR SAMPLING

Sampling for different wastes.

In order to investigate different wastes collected in a drum containing solids potentially contaminated by tritium the DA technique was particularly interesting to separate contaminated and not contaminated wastes. In Figure 5 (top), the very low level waste was deposited on the DA screen and the scan obtained shows at real scale the H-3 effect on screen (bottom of Figure 5). The sampling protocol by digital autoradiography is straight forward.

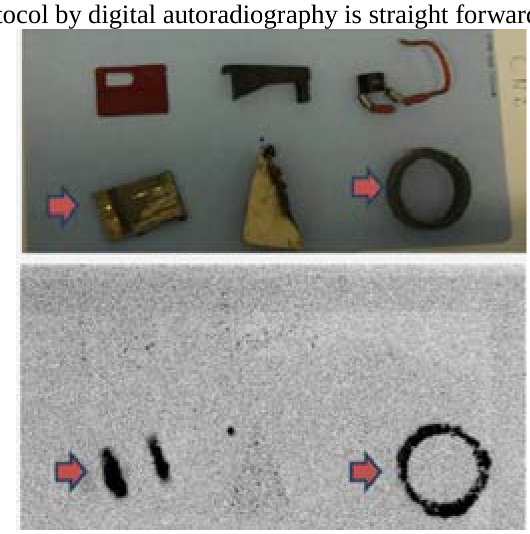

Fig. 5. Example of potential tritiated materials containing (red arrows) or not H-3 (black image on autoradiographic image).

Sampling for different wipes.

Twenty wipes were studied by autoradiography. Instead of directly starting the whole digestion of each wipe required by standard analytical technique such as alpha spectrometry, each of them were placed in contact with several screens. Figure 6 shows a part of the experiment performed on three wipes among twenty. Only one wipe marked a screen after the exposure time (Figure 6 bottom). After the digestion of this unique wipe, it was possible to conclude on the presence of Uranium on a single wipe. The DA technique is very sensitive to alpha emitters.

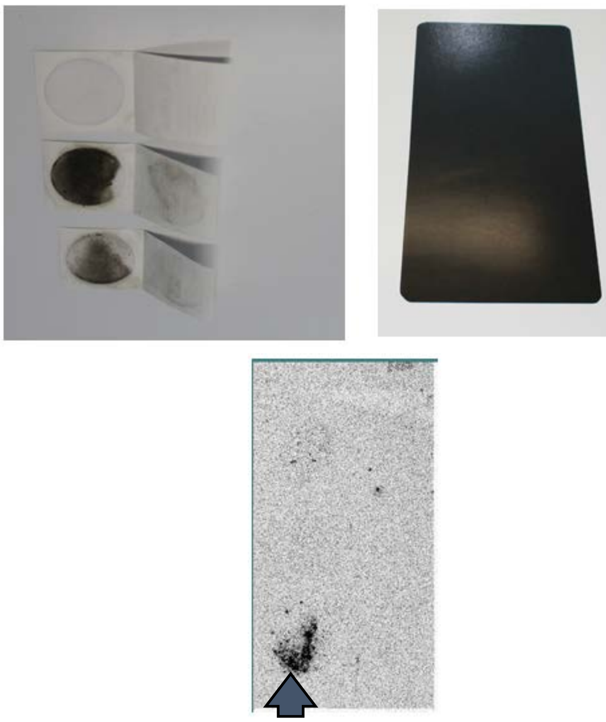

Fig 6: Example of the effect of three wipes on a DA screen (top the measurement with a screen deposited on the wipes, bottom the effect of the presence of uranium)

Sampling on a core

The **UNGG** is an obsolete nuclear power reactor design developed in France. It was graphite moderated, cooled by carbon dioxide, and fueled with natural uranium metal. All NPP are stopped and dismantling has started.

Some graphite cores removed from reactors for characterization must be analyzed and sampled. Thus to study the sampling protocol the autoradiography appeared as a very efficient technique. Once the raw result obtained a grid is drawn in order to provide the homogeneity of the radioactivity at the surface of the core with a resolution of mm^2. Kartotrak software (8) was used for the mapping. Figure 6 (right) shows the relative homogeneity of the radioactivity. Considering the developments on the fingerprints database obtained for DA, it was possible to identify the main radionuclide that impresses the DA screen i.e; C-14.

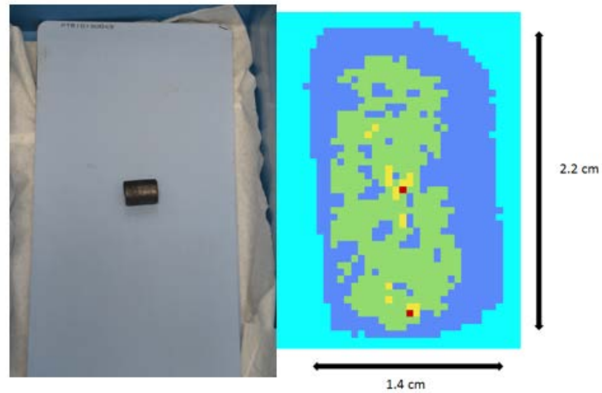

Fig 7: Graphite core deposited on a DA screen (left) and the homogeneity of C-14 (right)

ENDNOTES

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