New developments of autoradiography technique to improve alpha and beta measurements for decommissioning facilities

P Fichet (1), C Mougel (1), I Desnoyers (2), P Sardini (3), H Word (4)

(1) Den – Service d'Etudes Analytiques et de Réactivité des Surfaces (SEARS), CEA (Alternative Energies and Atomic Energy Commission, France), Université Paris-Saclay, F-91191, Gif sur Yvette, France
(2) Geovariances 49bis avenue Franklin Roosevelt - 77215 Avon cedex – France
(3) IC2MP- HydrASA / UMR 7285 CNRS, 86073 Poitiers cedex 9, France
(4) ARL/ Biospace Lab, 95960 Nesles-la-Vallée, France

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Abstract

In the framework of dismantling of nuclear facilities, there is a strong need to obtain accurate analytical results on residual radioactivity on different solid materials (concrete, metals, …). Based on lots of developments of autoradiography technique for biological investigations, the technique has been used for different characterizations on radioactive materials because autoradiography is particularly dedicated to radionuclides difficult to measure i.e. alpha and beta emitters. The different results obtained has proved that autoradiography technique is applicable to many characterizations of materials from decommissioning facilities.

Introduction

For any radioactive waste generated during operational and post operational activities, there is a strong necessity to characterize as much as possible the residual radioactivity. The initial characterization informs the requirements for safety and the importance of accurate characterization is essential for decommissioning and moreover to nuclear waste management. The final characterization is very important for final release of nuclear facility, depending on each country regulation.

Gamma contamination can be easily detected using different commercial detectors (Gamma Camera, Portable Gamma, etc..). To investigate non gamma emitters in situ directly on solid radioactive wastes, robust, sensitive and cost effective analytical technique are rather scarce. That is the reason why a new technology called autoradiography has been developed at CEA for decommissioning and Research and Developments are ongoing on this technique. Initially developed for biological researches, the autoradiography technique [1] has been found to be very promising to investigate alpha and beta emitters [2-4] for decommissioning. This

technique is a nuclear measurements that provides an image of radioactivity at the surface of the sample.

For different dismantling facilities, autoradiography has been described [2; 3] as a very powerful technique to investigate C-14 and H-3. Some applications concerning the quantitative measurements obtained was described [2] and new techniques have been developed to improve the potentialities of autoradiography to identify different radionuclides [4].

Autoradiography using reusable screens has been developed at CEA (Atomic Energy Commission, France) for different applications (mainly for beta (even tritium) and alpha emitters) relating to qualitative mapping, sampling procedures and in-depth investigations (by different core studies). For qualitative mapping and more generally for results presentations, a GIS (Geographic Information System) connected with geostatistics calculations is a powerful system to optimize measurement time, costs and stakeholder decisions.

Apart from systems based on reusable screens (post-treatment required), commercial systems using CCD cameras (imaging in real time) already exist for biological applications. For these applications, the spatial resolution required for the images produced is very high (of the order tens of μm). For in-situ measurement of nuclear wastes, developments of these systems can be required to provide images of radioactivity in real time. These developments will lead to the necessary efficiency for sampling procedures always required for analysis in expert laboratories. The spatial resolution required for such measurements on radioactive wastes is lower than for biological researches and must be studied. Autoradiography is a new and innovative system that could be very useful for in-situ measurements.

Results and Discussion

Use of Autoradiography to obtain radionuclides mapping

The first technique developed for autoradiography for decommissioning was based on the use of reusable screen (TR (for Tritium) or MS (Multi Sensitive), **Figure 1**) that are sensitive to all types of radiation. These films contain photostimulable crystals which accumulate and store radiation effect when placed in close contact with a radiation source.

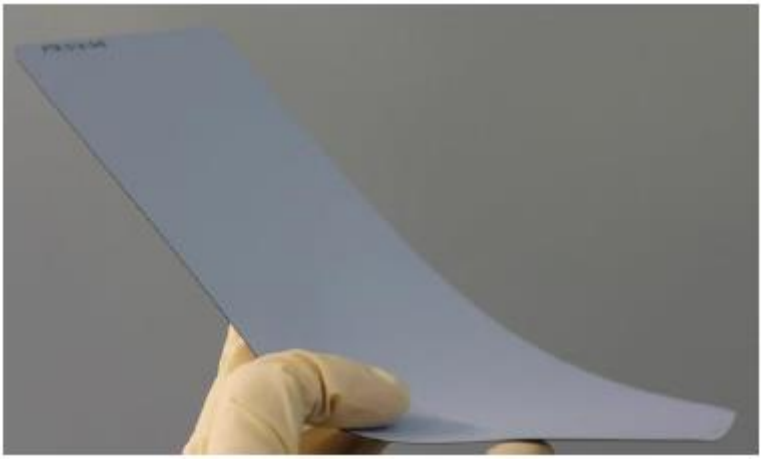

Figure 1: Autoradiographic screen (TR screen from Perkin Elmer)

The screen size used is 12.5 cm * 25.2 cm which induces around twenty screens at the same time to be able to obtain radionuclides mapping. The stakeholder requirement for the mapping has been to localize precisely the remaining traces of radioactivity on the floor of a laboratory to be dismantled.

Each of the twenty screens deposited in different well localized zones of the floor were systematically scanned by the Cyclone Plus (Perkin Elmer) able to extract the effect of the radioactivity [1] on the screen in order to obtain an image of radioactivity. For each screen deposited an image was produced. Each screen is reusable tens of times and thus can be used several times.

To obtain the residual of radioactivity on the floor (250 m^2 area), a total of thousand screens was deposited on different locations producing thousand images on different locations of the floor. The choice of the different locations was done in order to provide results coming from quite all the different areas of the floor. All the measurement obtained were uniformely distributed.

All the different images provides real measurements of approximately 20% of the whole area distributed uniformly. Once the autoradiographic images were obtained the GIS (Geographic Information System) of Kartotrak software [5] were used to draw at scale the measurements. Kartotrak is a software using geostatistcs methods in order to provide the whole measurements assuming that traces of radioactivity are well structured. Once geostatistics methods are used all the results can be drawn on the facility mapping. **Figure 2** shows the results of tritium traces measured partly by autoradiography and calculated by geostatistics.

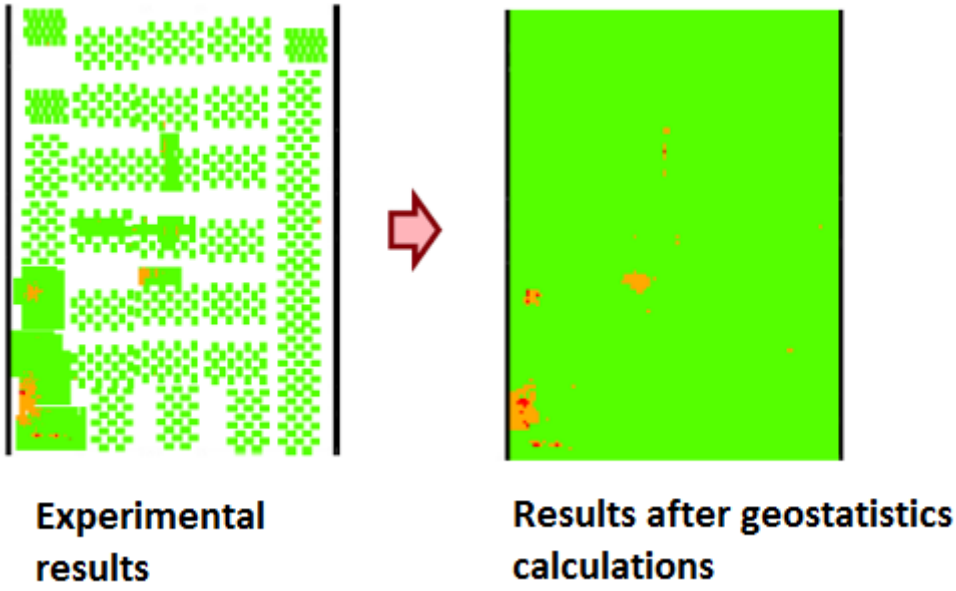

Figure 2: Tritium mapping on the floor (concrete) of a facility (250 m^2). Left: analysis results by autoradiography Right: Results after geostatistics calculations.

Autoradiography measurement for sampling procedures

The ratio methods using easy to measure radionuclides (gamma emitters) are widely used for nuclear waste management. But the ratio determination can be quite hard. To address this issue destructive analysis are very efficient but sampling procedure is a real problem. Autoradiography technique can be very efficient to provide results particularly for alpha and beta emitters to improve sampling procedure. **Figure 3** shows real tritiated wastes that were deposited on a TR autoradiographic screen. After 24 hours of exposure time, an image in black appeared when waste contains tritium whereas no image appears for free of radioactivity sample. This way of investigation by nondestructive technique is very efficient to improve sampling procedure required for destructive analysis.

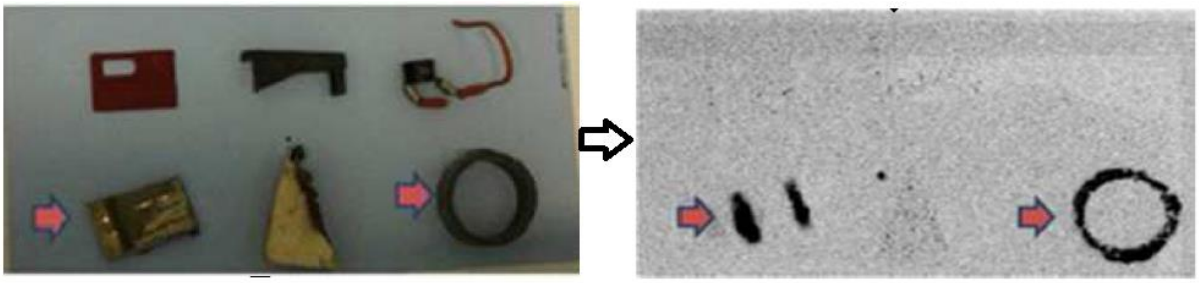

Figure 3: Investigation (left: real wastes on a screen, right: autoradiography scan) of different nuclear wastes by autoradiography, some containing tritium (black image obtained by autoradiography), some without tritium on surface.

Development of a new detector for autoradiography measurements

Following different requirements of researches for biology, commercial systems based on CCD detector exist. A first result obtained in 4 hours exposure time was obtained (see Figure 4) on a natural sample containing approximately 1 Bq/cm^2 of Uranium. For residual activity control this system could be of high interest to evaluate potential radioactive spots of radionuclide hard to be measured. The experimental spot resolution has a surface of around 10 µm.

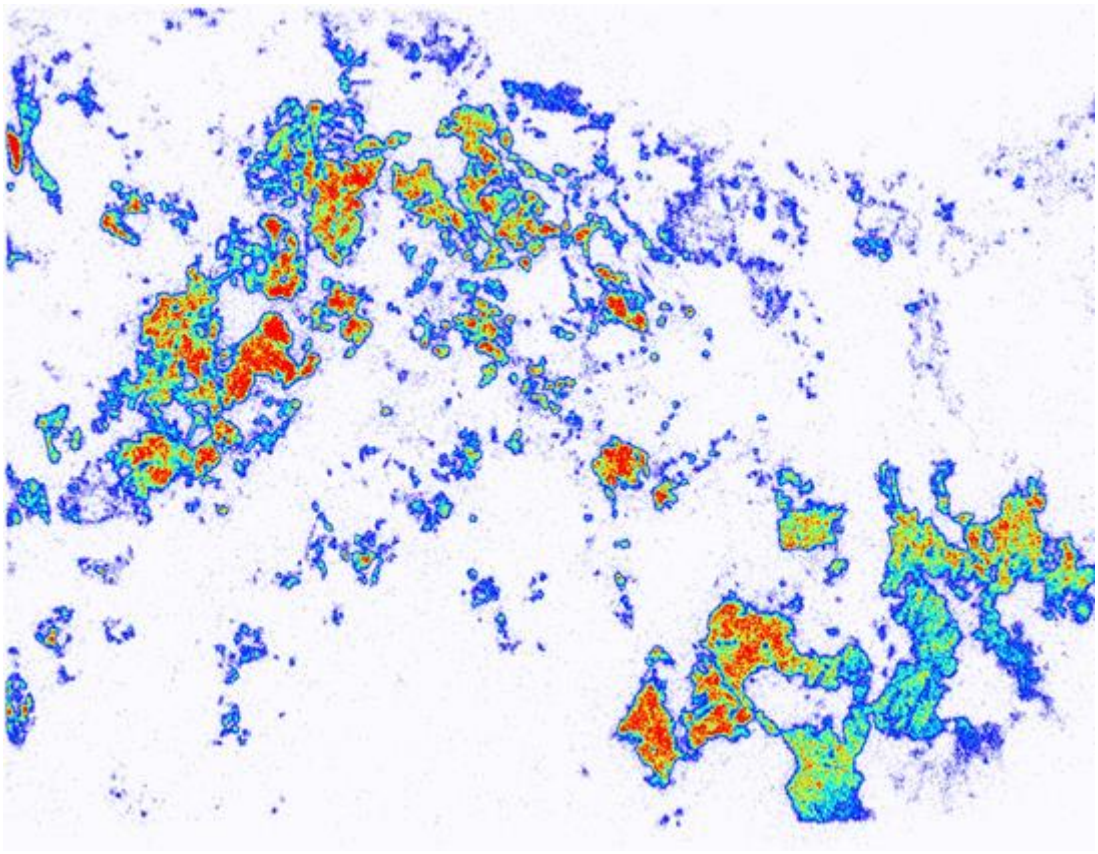

Figure 4: Measurements by Autoradiography of a sample containing Uranium.

Conclusions

Autoradiography method is under development to evaluate its potentialities to measure beta and alpha radionuclides that must be characterized in dismantling facilities. Geostatistics methods applied to facility characterizations have been very efficient to interpret experimental results obtained by autoradiography.

Autoradiography technique has high potentialities to improve sampling procedures required for destructive methods.

Finally new developments are underway to develop new autoradiography technics using CCD detectors.

Acknowledgements

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