Recent Advances in Low-Level Nuclear Measurements at the CEA– 9212

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ABSTRACT

For several years the CEA has been performing nuclear measurements at different stages of decommissioning projects. The characterization tools initially developed for high-level radioactive waste analysis must be adapted to a new area of application: low-level measurements. Recent technical improvements in gamma imaging, gamma spectrometry detectors, and data analysis make it possible to provide relevant radiological data for waste management, as well as to develop robust and optimized decommissioning scenarios from the initial dose rate mapping to the final declared activity.

These techniques have been implemented at various nuclear sites for both trial measurement campaigns and expert investigations to localize residual contamination, to identify the radioelements and to provide an accurate estimate of the declarable activity. Different types of measurements and devices have been used: gamma cameras [1][2], coded aperture techniques, alpha imaging prototypes, gamma spectrometry detectors (CdZnTe, HPGe, NaI, and LaBr$_3$) [3], dose rate cartography, and calculation codes (Mercure, MCNP, etc.), all of which provide complementary data for radioactive waste categorization [4].

This paper describes the latest developments and methods deployed on decommissioning projects focusing on low-level in situ applications. Waste drum characterization and in situ glove box measurements are discussed and the technologies and performance of gamma imaging systems, gamma spectrometry detectors, calculation codes and software are described. The paper concludes with a review of future developments and tests necessary for these applications.

INTRODUCTION

The CEA has for many years been developing and qualifying nuclear measurement systems for dismantling projects. Some of these systems are now commercially available and are used by operators in decommissioning nuclear facilities. They are applicable at several stages of dismantling a facility or decommissioning a site: for the initial characterization, during the decontamination phases, and up to the final surface inspections for decommissioning.

The CEA Techniques and Methods Laboratory at Marcoule has developed nondestructive measuring instruments for radiological characterization of nuclear facilities. The high irradiation levels initially encountered led to the use of remotely operated equipment and the development of prototypes devices suitable for extreme environments. These developments include gamma imaging, which is widely deployed at dismantling sites and is now commercially available. Compact high-performance CdZnTe gamma spectrometry probes have also been developed and qualified to meet the constraints of characterization in severe irradiation conditions. These systems coupled with other types of measurements (dose rates, surface measurements, etc.) and techniques (calculation codes) are now an integral part of any decommissioning project.

More recent progress in decommissioning projects has identified new requirements for low-level measurements, with the objective not only of detecting the raw signal but also of quantifying the residual activity: characterization of irradiating waste, final inspection (verification of walls and floors), glove box measurements (residual activity, decontamination progress), and miscellaneous radiological characterization. These in situ measurement techniques require increasing sensitivity and resolution. The issue of background noise must also be taken into account. In this context the CEA is improving existing tools (gamma imaging), qualifying new detectors (gamma spectrometry), and developing new methods and techniques for source location and quantification (alpha imaging, high-resolution spectrometry to estimate the contamination depth). Some of this work has reached the qualification stage prior to industrialization, and field tests of measurement systems are now in progress. Others are still under development.

This article describes the techniques currently available or now being developed for low-level detection. Examples are commented to highlight the progress made to date, and the objectives that must be met for future projects.

GAMMA IMAGING

The "Aladin" prototype gamma camera has been developed since the early 1980s. The basic principle is explained in Figure 1; gamma radiation is collected through a scintillator and converted to visible light, but with a very low conversion yield. The scintillator is therefore supplemented with an image intensifier to amplify the signal supplied to a CCD array. The shaped signal is transmitted to acquisition circuitry to form an 8-bit image received by the control unit
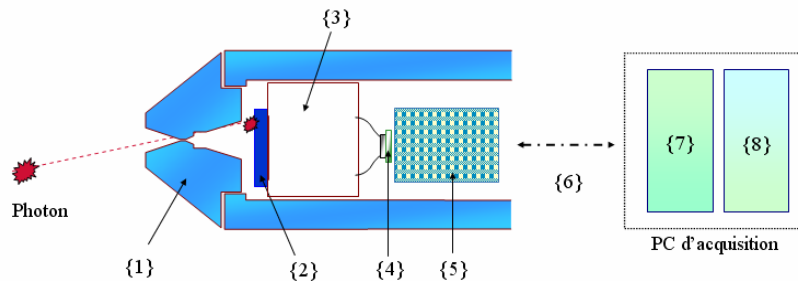

Fig. 1. Gamma camera schematic
{1} Collimator, {2} Scintillator, {3} Image intensifier tube
{4} CCD array, {5} Video circuitry / HT power supply
{6} 30 m cable, {7} Acquisition circuit board, {8} Software

The gamma camera has been widely used for in situ measurements. It is capable of remotely localizing irradiation sources and estimating the dose rate produced by the observed source. This dose rate value is obtained directly by processing the raw gamma image. Nowadays, gamma camera cartography can replace collimated dose rate mapping methods by providing the image of the scene from one measurement point which represents a significant gain of time.

The performance of the gamma camera has been enhanced by several upgrades of the prototype system that are now available depending on the application requirements [5][6][7]:

- optical system using coded aperture masks,
- antipinhole collimator,

- improved image acquisition and processing circuitry.

Coded aperture mask optical system

Coded aperture masks are used in the aerospace industry and have been adapted to gamma imaging. The technique is based on theoretical work by Fennimore and Cannon [6].

The standard objective of the Aladin gamma camera is a pinhole aperture 1.2 mm in diameter. In this implementation it was replaced by an objective with a coded aperture mask forming a Hexagonal Uniformly Redundant Array (HURA) [6][8].

The Figure 2 shows the HURA coded aperture mounted on the Aladin II gamma camera system.

Fig. 2. Aladin II in coded aperture mask configuration

Replacing the pinhole significantly increases the signal level at the scintillator and thus improves the gamma camera sensitivity. Laboratory tests showed that in 20 minutes it can localize a ^{137}Cs point source generating a dose rate of 35 nGy·h^{-1} at the detection head.

Field tests also yielded promising results. Comparative measurements between pinhole and coded aperture mask systems were carried out on waste drums and glove boxes, confirming the significantly higher gain, sensitivity and resolution obtained using coded aperture masks. Figure 3 represents one measurement involving a test drum with Pu pellets randomly distributed inside; each test pellet contained 1 g of Pu.
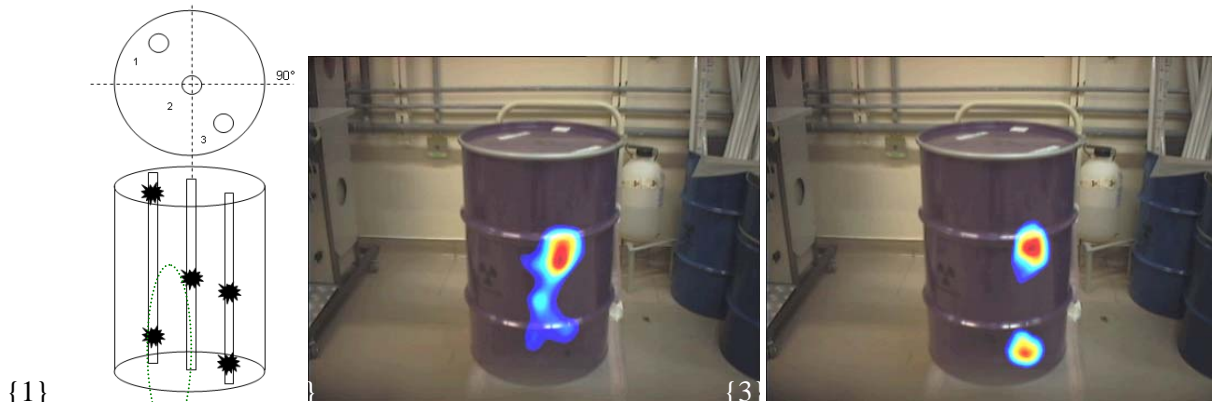
{1}

{3}

Fig. 3. {1} Positions of 2 Pu sources (the 3 others were "masked" from this position of the gamma camera); each granulate is a 1 g Pu source,
{2} Pinhole image, {3} Coded aperture image

Imaging was carried out in the same measurement configuration using a gamma camera equipped alternately with a coded aperture mask and a pinhole aperture. For a given acquisition time, the coded aperture mask increased the gamma camera performance by a factor of 4.

Figure 4 represents another series of measurements in glove boxes also containing residual Pu traces. The contamination traces inside glove box were identified more rapidly and with greater accuracy with the coded aperture system than in pinhole mode.
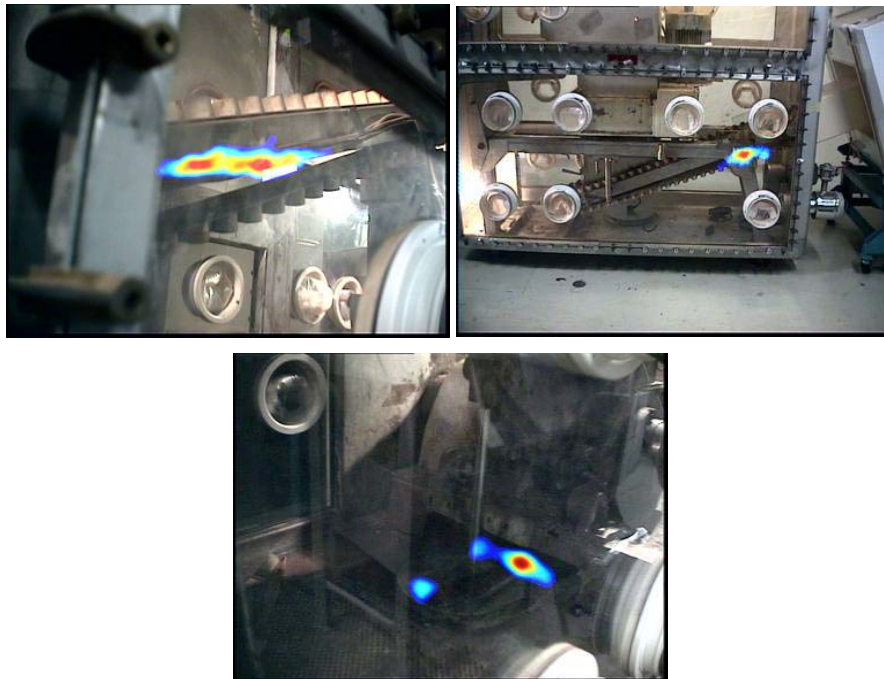

Fig. 4. Coded aperture images in glove box application

One limitation of this technique is the appearance of phantom sources or flare when an irradiating source is situated outside the field of view of the gamma camera. Figure 5 shows the results from an image processing module developed by the CEA to facilitate the identification of true sources in the image. In this example, when an extended source (1) is created along the conveyor, the simulator shows

that zone (2) in the image is a decoding artifact generated by the signal situated in the partially coded field (3). This was subsequently confirmed by the pinhole image.
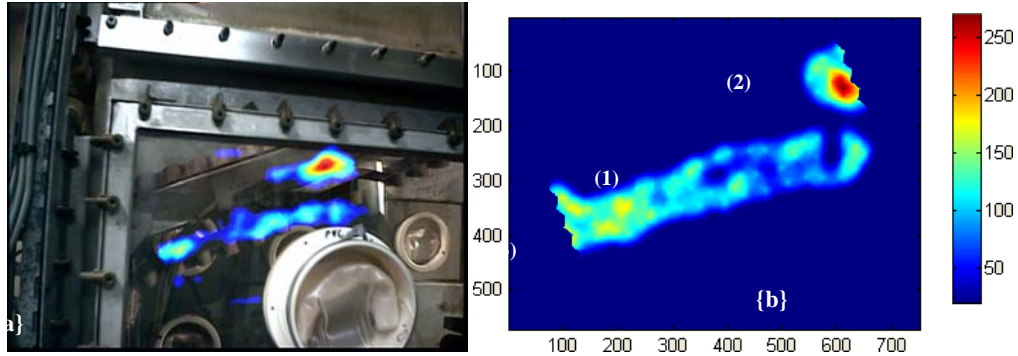

Fig. 5. Artifact simulator software
{a} in situ measurement, {b} simulator result

Moreover, the use of coded aperture masks is also an advantage at a fixed measurement station (waste characterization, glove box measurements) when the gamma camera field of view is controlled.

Antipinhole Collimator

Adding a prototype objective with a dense ("antipinhole") shutter also enhanced the performance of the basic Aladin pinhole gamma camera. The principle is simple: block spurious environmental noise and electronic background noise by systematic noise measurement. The noise image is obtained by blanking the pinhole aperture with a Denal screen (Tungsten alloy). Subtracting the noise level in the blank image from each pixel in the measurement image significantly improves the signal/noise ratio and enhances the system performance. Figure 6 represents this "antipinhole" system mounted on the Aladin II prototype.

Fig. 6. Antipinhole collimator on a gamma camera

The dense shutter technique eliminated the noise by generating a homogeneous background, and identified the most activated zones. An example is given on the following figure 7.
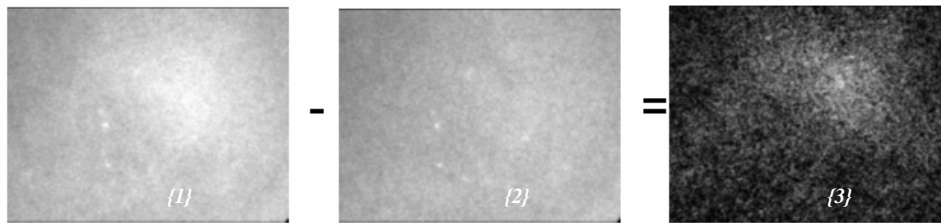
Fig. 7. Using the antipinhole collimator
{1} Raw image, {2} noise image, {3} sharp image

Figure 8 represents the results of the full gamma camera characterization of the inner walls of the SILOE reactor vessel (Grenoble, France). Activation of all the walls generated a strong ambient signal around the gamma camera placed at the center of the cylinder. This noise signal was completely removed using the antipinhole collimator technique and the remaining hot spots were located on the final image.
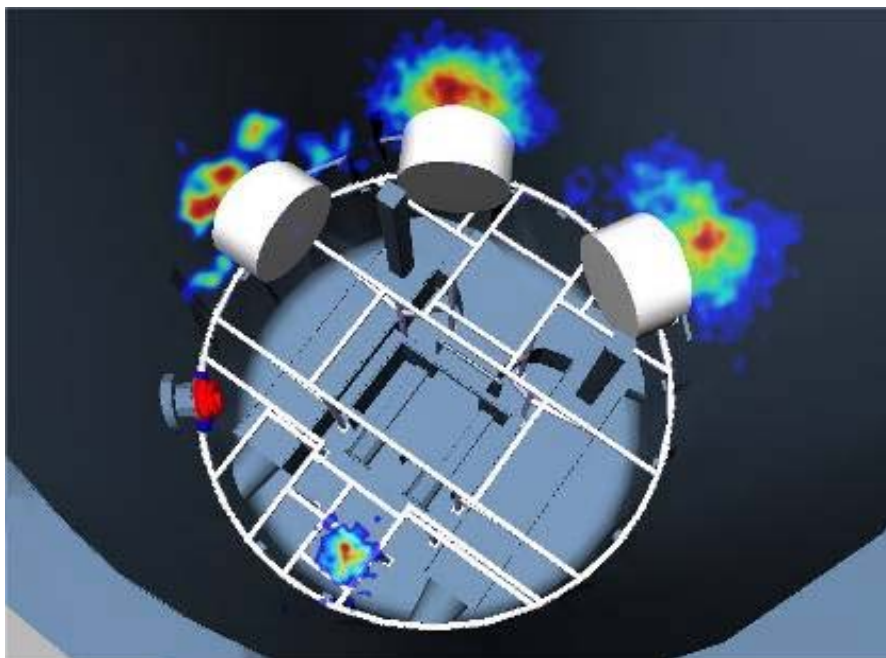

Fig. 8. Inspection of the SILOE reactor vessel (CEA Grenoble)

Improved image acquisition and processing circuitry

The most recent developments of the basic prototype (Aladin 3 v2 pinhole) did not concern the hardware but rather improvements to the raw gamma image acquisition and processing software.

In gamma image acquisition the CCD discharge rate generally varies between 0.2 s (5 frames/second) and 1 s (1 frame/second). The final image is obtained through accumulation of all the images acquired with a fixed exposure time. The images transmitted by the CCD are encoded with 8 bits (256 levels) per pixel; the frame buffer uses variable storage from 8 bits at the beginning of the accumulation process to 16 bits if high signal levels are accumulated (to prevent image saturation).

The development of an optimized image processing algorithm allows the images accumulated in the buffer to be analyzed in real time to detect the appearance of high-density zones in the image. In other words, an irradiating zone can be identified without waiting until the end of the image acquisition period.

If the detected signal is strong enough it can even be identified after the initial CCD discharges. Consequently, with the same exposure time areas of weaker signals can also be identified.

Until now, raw gamma images were processed after the end of the accumulation phase. The new algorithms combined with an optimized gamma camera design (CsI scintillator, pinhole aperture) now allow real-time image processing and computation, and significantly improve the hot spot detection time. The next figure (Fig. 9) shows some of the specific software developed by the CEA for gamma imaging.
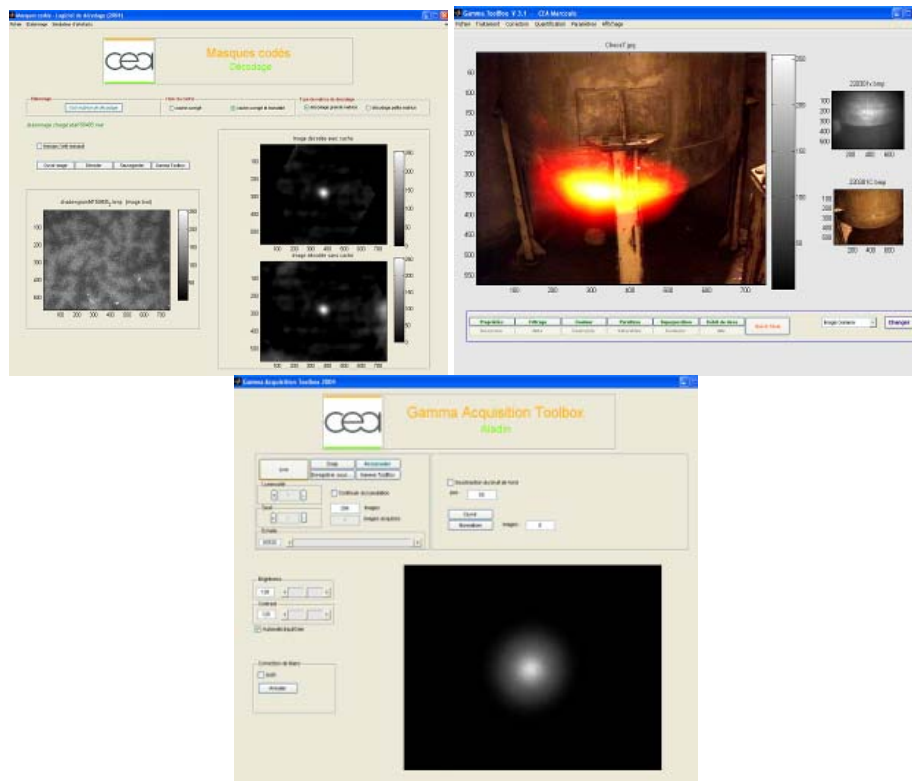

Fig. 9. Acquisition and processing software

With these developments, gamma imaging has become suitable for low-level measurement. Installed measuring stations for waste packages or glove boxes are easy to set up since the field of view of the gamma camera is known and controlled. The imaging results are very useful for developing robust radiological models: the hot spot location and shape constitute valuable input data for minimizing the calculated uncertainties in numerical simulations.

The drawbacks of gamma imaging for both low- and high-level measurements are:

• high equipment cost,

• in situ gamma imaging requires the presence of qualified technicians,

• coded aperture masks and pinhole/antipinhole collimators are only available on the Aladin prototype at this time. Tests have also been carried out with the Cartogam gamma camera [8][9] marketed by Canberra France SAS (CEA license), but no industrial version is yet available.

GAMMA SPECTROMETRY

Gamma spectrometry is still the only means of quickly identifying the radioelements detected and of obtaining quantitative data on each of them. It is frequently coupled with gamma imaging for radiological characterization [CEA patent]; the additional information provided in this way (location, identification and quantification) is then used to develop robust and reliable models. The CEA qualifies gamma spectrometry detectors to provide technical support for decommissioning projects. As with gamma imaging, the constraints of strong irradiation require the use of suitable detectors, especially compact CdZnTe semiconductor detectors capable of room-temperature operation. Crystals are available in sizes ranging from 0.5 mm^3 to 1500 mm^3; the largest are capable of low-level measurements of about 1 $\mu Gy \cdot h^{-1}$ for ^{137}Cs. Other detectors (scintillators) using more recent technology have also been evaluated (LaBr$_3$, LaCl$_3$) and have been found advantageous for low-level measurements and VLLW characterization despite their intrinsic radioactivity. Better known and inexpensive NaI detectors are suitable for low signal levels by virtue of their high sensitivity but are capable of only very limited resolution. These kind of detectors are presented in Figure 10.
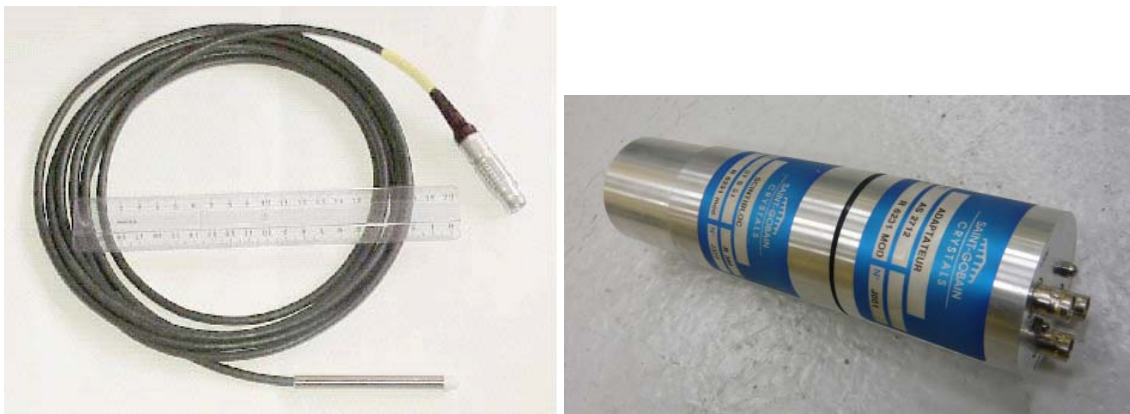

Fig. 10. Examples of CdZnTe and LaBr$_3$ detectors

Low-resolution detectors all share the same problem when processing low signal levels or seriously perturbed measurements: it is difficult to determine the peak area reliably and reproducibly. In this context a software solution, SIGALE, is now being qualified in collaboration with CEA/DETECS after two years of development work. SIGALE is designed for automatic processing of "degraded" spectra, i.e. spectra that are seriously perturbed (strong Compton front) or obtained from low-resolution detectors. Under certain conditions, manual processing is a significant factor in the quantification uncertainty for isotopes in the spectrum. SIGALE is designed to mitigate this problem through software that is easy to use in the field. An automatic peak detection algorithm and automated surface processing routines are implemented in the program. This software illustrated in Figure 11 is particularly useful for dealing with spectra obtained using CdZnTe probes, which characteristically generate spectra with skewed peaks despite their satisfactory resolution.
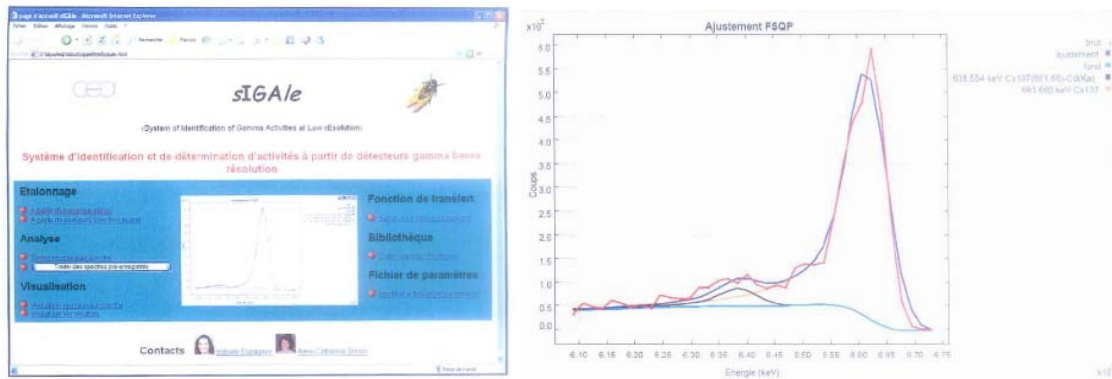
Fig. 11. SIGALE: System of Identification of Gamma Activities at Low rEsolution

Germanium detectors are used for high-resolution surface measurements (final inspection) and to estimate the contamination depth in concrete structures.

The objective of the final inspection is to confirm that a surface, a room or an entire nuclear facility can be classified as nonradioactive [10]. This implies measuring very large surface areas: several thousand square meters in the case of a French nuclear power plant. The inspection method uses a HPGe detector (40%) with a collimator specifically designed to observe an area of about 9 m^2 from a distance of 1.5 m (Figure 12). The detection limit of 0.1 Bq·g^{-1} (for ^{137}Cs and ^{60}Co) is quickly reached in three minutes. The measured data are then converted using a straight-line attenuation calculation code; the main hypothesis is the distribution of the source term on the surface. This technique is available today and was recently used in the program to decommission CEA facilities in Grenoble (France).
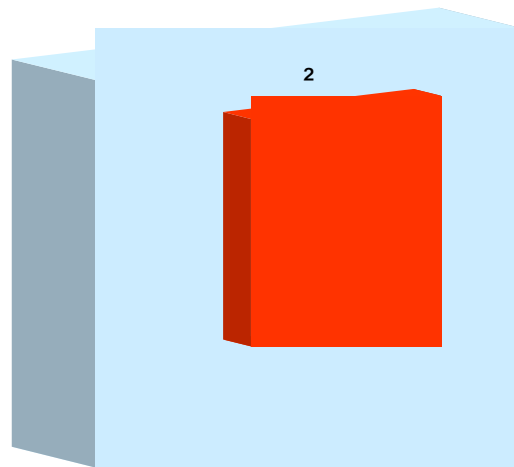

Fig. 12. Wall surface inspection system (collimator + HPGe detector)

Nondestructive measurements remain the least costly means of obtaining radiological data concerning a facility, resolving uncertainties about the presence of contamination, supporting a project study, or directing a decontamination project. In collaboration with the Kurchatov Institute of Moscow (Russia) the CEA is currently developing a nondestructive method for estimating the contamination depth in concrete structures based on gamma spectrometry measurements [11].

The objective is to develop a device that is easy to set up opposite a concrete surface (wall or floor) and capable of determining the concrete surface area contaminated by ^{137}Cs or ^{60}Co and the total activity of the observed sample. The core sampling techniques currently in use are very costly and have therefore

been optimized to limit the expense. The representativeness of the measured samples is thus often limited by the measurement cost.

The proposed method is based on collimated gamma spectrometry measurements of the work zone and requires a high-resolution (hyperpure germanium) detector. Prior Monte Carlo modeling of the detector allows the expected spectra to be simulated numerically according to the radionuclide migration configurations (linear, exponential, or polynomial profiles, nature of the radioelements.

Some specific software developments allow the spectrum analysis to be performed online and is capable of determining the contamination depth comprising 80% of the total sample activity. The method is currently being developed and qualified for ^{137}Cs and ^{60}Co. The low detection levels (1 Bq·g^{-1}) require ambient noise subtraction (using a blanked collimator) for each measurement. Figure 13 represents the full system in use on CEA Grenoble and EDF sites.

Fig. 13. System in use

The system is now being qualified and is currently being field tested. With the support of the R&D department of EDF, the system was implemented on a decommissioning site and the results were compared with those given by core sampling of the same surfaces; the results are currently being analyzed. These tests will continue in 2009 to validate the system in various configurations. The system capabilities may subsequently be extended to other radioelements and other types of gamma spectrometry detectors.

The use of gamma spectrometry detectors for low-level measurements is not new, but new development work involving the qualification of new detectors is now required in view of the very high stakes arising from future decommissioning needs. Some of these developments (SIGALE software, contamination depth estimation, final inspection methodology) are now sufficiently advanced to consider industrial deployment.

ALPHA IMAGING

Like gamma imaging, remote localization of hot spots and surfaces contaminated by alpha-emitters is a decisive factor in developing a radiological model or monitoring the decontamination of process equipment in glove boxes, for example. A few years ago the CEA developed the first prototype alpha imaging system capable of remotely localizing alpha-emitters [12]. The detection uses the UV radiation emitted when nitrogen in the air is excited by the passage of alpha particles. The observable UV spectrum

is situated in the near-visible wavelengths (between 280 nm and 500 nm for the main peaks); the main issue is thus spurious detection when the measurements are perturbed by visible light.

The first prototype was a cryogenic UV camera cooled by liquid nitrogen with a wide-angle lens. Signal collection to obtain a good quality image is long but interesting results have been obtained and demonstrate the feasibility of the process. The system has even been field tested in complete darkness. Figure 14 shows the first prototype camera {1} and the results of measuring a 30kBq [241]Am point-like source, superimposed on a visible image of the source {2}.
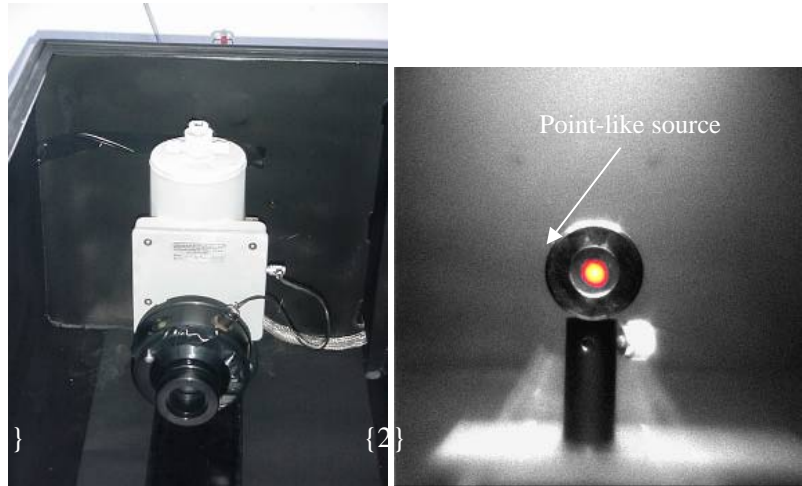

Fig. 14. {1} First prototype alpha camera (cryogenic UV camera)
{2} Detection of a 30 kBq [241]Am point-like source after 1 hour in darkness

The long exposures necessary to obtain images, the need for liquid nitrogen cooling, and the influence of visible light recently prompted new tests with an intensified CCD camera combined with a multi-alkali UV photocathode illustrated on figure 15. The spectrum response ranges from 180 to 800 nm. This camera is particularly useful for detecting very weak UV signals.

Fig. 15. Intensified UV camera

The same physical phenomenon is detected, but with significantly higher efficiency: 30 and 150 kBq surface [238]Pu and [241]Am sources were observed in less than 10 minutes in darkness. The acquisition process in this configuration resembles gamma imaging, i.e. the accumulation of several frames in a 16-bit buffer at the rate of 25 frames/second. The raw images are processed (filtering, color rendering) then superimposed on the visible-light image also obtained with the UV camera. For example some planar [238]Pu and [241]Am sources were observed with small exposure time during first laboratory trials of the camera (Figure 16).
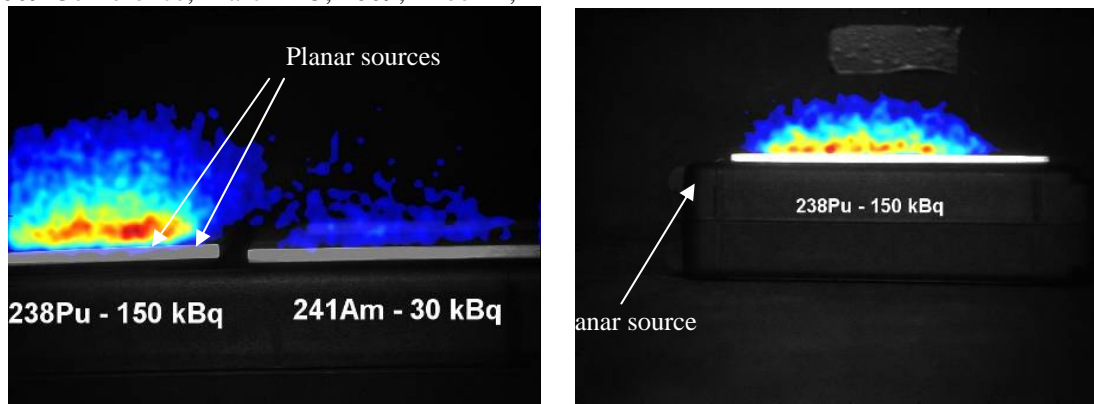
Fig. 16. Observation of planar [241]Am and [238]Pu sources in less than 10 minutes

The intrinsic camera noise is very low, and UV radiation can be detected at wavelengths exceeding 230 nm corresponding to the spectral range of the phenomenon observed. However, the very high sensitivity also makes the camera very sensitive to visible light, and the images must therefore be acquired in darkness.

The new prototype is promising and will soon be tested under actual working conditions. UV filter tests are also scheduled to diminish or eliminate the effects of visible light on the image acquisition and make the system easier to use. Once qualified, this technique will be destined for industrial applications to monitor decontamination (glove boxes) and characterize surface deposits.

CONCLUSION

Nondestructive in situ measurements of low signal levels have not been widely investigated to date. Only gamma spectrometry is in widespread use for low-level measurements because of the wide range of detectors available. Other technological developments and methods, such as contamination depth determination or alpha imaging, are still under development. The lessons learned from experience are also vital for the development of techniques, and it is important to qualify these tools on actual dismantling projects.

The CEA is performing these developments to give more relevant tools to the decommissioning projects and to industrial contractors. A current example of this industrialization process is NARVEOS, a software recently marketed by Euriware under CEA license which can render a 3D scene incorporating all the nuclear measurement data collected in the facility (shape and activity of hot spots, standard spectra, dose-rate distribution maps, etc.) and generate operational scenarios with real-time calculation of the operator integrated dose.

Other issues related to in situ nuclear measurements are also under development such as sampling method that is especially useful when inspecting large surface areas in buildings, and also a statistical sampling process which can orient nuclear measurements and optimize the acquisition of radiological data while cutting costs.

The ongoing development of these methods and techniques will lead to a broad range of technical solutions for industrial projects. The stakes are high, especially at the final decontamination and decommissioning final stages in nuclear facilities.

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