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R&D APPLIED TO THE NON-DESTRUCTIVE CHARACTERIZATION OF WASTE PACKAGES FOR LONG TERM STORAGE OR DEEP DISPOSAL

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1. INTRODUCTION

To ensure the quality and traceability of waste package management in the long term, knowledge on these packages is necessary so as to confirm their compliance to storage or disposal specifications. Research is focused on the management of the knowledge on these packages (fabrication means, materials contained,...) and on the acquisition, through measurement, of their characteristics, with the aim to :

- being able to propose measurement means to the waste producers, allowing them to better categorize certain types of waste and to fill in the information files on these packages,

- providing the storage and disposal facility designers and operators with qualified measurement means for the achievement of standard controls on these packages, thus checking their compliance with the acceptability criteria of the facility.

Within this context, many studies are underway at the CEA in the field of measurements so as to obtain non-destructive tools to access the parameters which allow the waste packages to be characterized. The two main R&D investigations concern :

- the nuclear measurement methods for the detection and quantification of radionuclides and of chemical elements considered as important for storage or disposal safety,

- the measurement methods for the physical characteristics of the packages by high energy photon imaging, thus allowing pictures of the contents of large, high density and sometimes irradiating packages to be known.

During the last five years, the research at the CEA focused on these two areas and resulted in a significant evolution in the non-destructive characterization means for long lived waste packages.

2. NUCLEAR MEASUREMENT METHODS FOR THE IDENTIFICATION OF THE RADIONUCLIDES AND CHEMCIAL ELEMENTS CONTAINED IN WASTE PACKAGES

The nuclear measurement methods developed or under development at the CEA for the non-destructive characterization of radioactive waste packages for a long term storage or a deep disposal concern, on the one hand, the improvement of classic techniques such as gamma ray spectroscopy, passive and active neutron measurements, and on the other hand, the development of new techniques, such as photon interrogation by means of a Linear Electron Accelerator.

2.2.1. Gamma-ray spectroscopy

The principle of this very widespread technique is to detect and then to classify, according to their energy, the gamma radiations spontaneously emitted by many radioactive elements after their disintegration. The energy of the gamma rays which appear on an acquisition spectrum are characteristic of each radioisotope which has disintegrated and allows its identification. The number of pulses recorded in these rays also allows its activity to be quantified, if the detection efficiency is known.

One of the greatest advantages of gamma spectrometry, particularly compared to non-destructive measurements based on neutron detection, is to provide specific information on each radio-isotope detected, and to cover a very wide range of radioactive elements. In addition to its use for identifying radioisotopes and quantifying their activity, it also can be used to establish the isotopic composition of the contaminant, and more precisely of heavy

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nuclei (uranium, plutonium, americium), so as to, for example, help in the interpretation of neutron measurements. The field of application is thus very large for the characterization of radioactive wastes.

Significant developments are underway for the reduction of measurement uncertainties. For example, the development of an « Anti-Compton » detection system, for irradiating bituminized waste packages, has resulted in improving the detection limit by a factor of close to 2 [1]

On the other hand, the coupling made between gamma ray emission tomography measurements and transmission tomography (a technique presented in § 3), enables, beyond the identification of the radioactive elements present in the package, their more accurate localization and quantification [2] (see Figure 1).

Gamma ray emission tomography of 661,7 keV ray of ^{137}Cs

Superimposition of attenuation and activity of ^{137}Cs

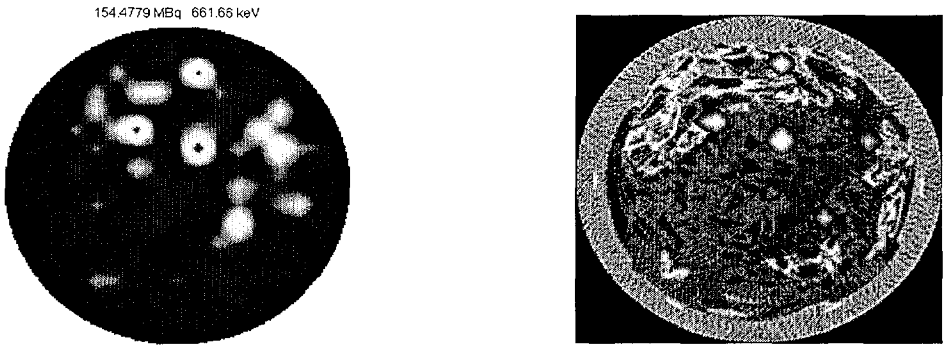


***Figure 1** : Coupling of gamma ray emission tomography and of transmission tomography*

2.2.2. Passive neutron measurement

The principle is based on the detection of the neutrons spontaneously emitted by the heavy nuclei during the reactions (α,n) and/or spontaneous fissions reactions. If one counts all of these neutrons indifferently, this is called total counting. However, a time analysis technique of the signal has been developed to distinguish the contributions of the neutrons issued from the reactions (α,n) on the light nuclei and the spontaneous fission neutrons. This is based on the fact that the first are emitted singly at each reaction whereas the second ones are emitted by 2 or 4 on average during each fission. This technique is called « neutron coincidence analysis ». Total counting provides access to a global response of all the isotopes with a high neutron emission rate ($^{242 \text{ and } 244}Cm$, $^{238,\ 240 \text{ and } 242}Pu$, ^{241}Am), and sometimes of those present in large quantities (^{239}Pu, ^{238}U). The measurement of neutron coincidences is only sensitive to heavy nuclei with a high spontaneous fission rate ($^{242 \text{ and } 244}Cm$, $^{238,\ 240 \text{ and } 242}Pu$), and sometimes to some radioelements present in large quantities (^{238}U).

The passive neutron measurements are today mastered and widespread in the industrial world, in total counting and in coincidence analysis. Many applications can be found in nuclear fuel cycle plants, both for the measurement of low activity wastes (plutonium activity measurement in technological wastes) and of middle level activity (curium measurement in hull and nozzle waste resulting from irradiated fuel assemblies reprocessing). The measurement of high activity wastes (such as vitrified fission products) does not seem to raise any major problems.

2.2.3. Neutron interrogation

Active neutron interrogation based on fission reaction [3]

The principle is to cause fission reactions in some heavy nuclei (especially in uranium and plutonium isotopes) and then to detect the prompt and/or delayed neutrons emitted after these fissions. The measurement of prompt neutrons provides a global response from all the « fissile » isotopes (^{235}U, ^{239}Pu, ^{241}Pu), i.e. with large fission cross sections in the thermal field. The measurement of delayed neutrons provides access to these isotopes, and also to the contribution of « fertile » isotopes, which can only fission at higher energy (roughly above one MeV), present in large quantities (^{238}U, sometimes ^{232}Th).

This type of measurement is perfectly adapted when the gamma ray spectroscopy does not allow the quantification of heavy nuclei because of the presence of fission and/or activation products. It is also very useful when the passive measurement does not allow the quantity of plutonium to be assessed because of the presence of curium (strong neutron emitter). The interpretation of active neutron measurement requires the knowledge on the relative proportions of the main contributions. The coupling of prompt and delayed neutron measurements, or of other techniques (gamma spectrometry if it allows certain heavy nuclei to be measured) and/or of preliminary information on the contaminant (isotopic composition of plutonium, uranium enrichment, U/Pu ratio) is thus vital.

Recent R&D has been particularly focused on the coupling of prompt and delayed neutron measurement. Simultaneous measurement is possible by means of a pulsed neutron generator. This also allows the contributions of uranium and plutonium in the prompt and delayed signals to be differentiated. The coupled measurement will be applied shortly on the industrial level for compacted hull and nozzle waste and for bituminized waste. At Cadarache, the CEA uses an active neutron measurement cell devoted to controlling the bituminized waste packages and used to demonstrate the characterization of middle level irradiating waste.

Neutron interrogation coupled with γ spectrometry [4]

The principle is to cause, by means of neutrons issued from an interrogating source, nuclear reactions on the elements to quantify, and then to detect the photons emitted simultaneously or in a differed way after these reactions. The two reactions used are neutron capture (n,γ) and neutron activation.

The spectroscopy of the capture gamma rays consists in detecting these photons emitted instantaneously during the captures, the later being essentially induces with thermal neutrons (larger cross section). This requires a neutron source, a neutron diffusion cell (for slowing down and confinement) and a highly efficient gamma spectroscopy detector since the capture radiations are generally emitted with an energy of several MeV.

The neutron activation measurement followed by disactivation gamma radiation spectroscopy consists in transmuting, by means of a neutron source, the isotopes to be characterized into unstable isotopes, which de-excite by emitting, among other things, gamma radiations. Those whose radioactive period is sufficiently long to allow the counting phase to start after the disappearance of the background noise due to the capture radiations used in the preceding technique are measured. Nevertheless, sufficiently short periods (from a few minutes to a few hours) are of interest since they allow a sufficient activation level. The features of the device are the same as for neutron capture. The gamma ray detector is devoted to average and high energy radiations (a few hundred keV to a few MeV). The neutron cell must be optimized (more or less moderator) depending on the isotope to be measured, because the activation reaction aimed at is either favored with fast neutrons (reactions (n,p), (n,α) and (n,2n)) or with thermal neutrons (reactions (n,γ)).

The elements who can possibly be quantified by these techniques have been studied and are long-lived radionuclides and toxic chemicals. For example, for the latter, the detection limits in small samples of roughly 1 kilogram have been established using numerical simulation. These are of a few ppm of Cd, a few dozen ppm of Hg and Cl and a few hundred ppm of Cr, Ni, As, Sb and Se.

2.2.4. Photon interrogation

Interrogation by induced photofissions [5]

The principle is to cause fission reactions on heavy nuclei (such as uranium and plutonium) by means of high energy photons (photofission threshold close to 6 MeV), then to detect the prompt and/or delayed neutrons emitted after these fissions. Only the delayed neutron measurement can be used in this technique because the measurement of prompt neutrons is too sensitive to the active background noise. The high energy photons are produced by a *LINAC* (LINear electron ACcelerator) and an electrons to photons conversion target based on bremmsstrahlung radiation. The photons used to cause the fission create an extremely intense background noise which blinds the neutron detection system. Therefore, the photon emission must be pulsed and the delayed neutrons must be measured between the accelerator pulses. The measurement of photofission delayed neutrons provides access to a global response of the heavy nuclei present in large quantities and mainly of ^{238}U.

Studies have been carried out on obtaining a photofission tomography whose aim is to localize the position of the heavy nuclei in the waste package (with the aim to correcting matrix and localization effects). This system is based on the use of the directivity of the photon beam and the response of the different counting blocks placed around the package. It reconstructs a 2D image of the heavy nuclei distribution in an axial section of the package, using measurements made in different irradiation positions and under different angles.

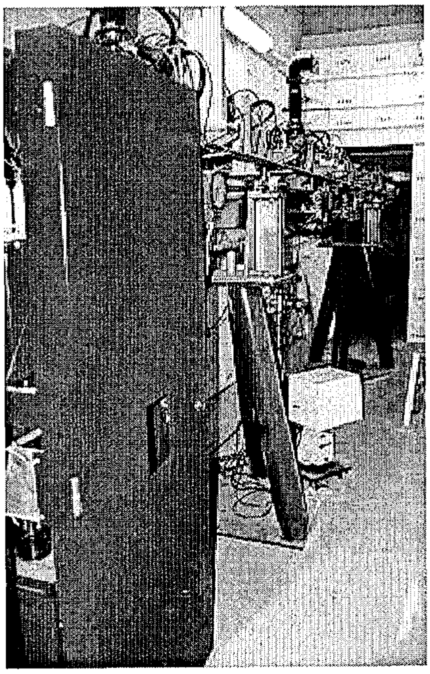
For the quantification and localization of heavy nuclei (U, Pu...), photofission can improve the detection limits by one order of magnitude compared to active neutron interrogation ; the detection limit in a concrete drum of roughly one m^3 was thus lowered under the alpha activity acceptability threshold of the Aube surface storage Center (CA) where only short-lived radioactive packages are stored.

Photon activation

The principle is to cause, by using photons issued from an interrogating source, nuclear reactions of activation on the elements to be quantified, and then to detect the gamma rays emitted during disactivation. This technique can be applied to the quantification of long-lived radionuclides and to matrix characterization (quantification of absorbing elements for neutron and/or photon radiations, in order to correct the matrix effects) and, as in the case of neutron excitation, the quantification of toxic chemical elements could also be a possibility. The feasibility studies underway also make use of a LINAC with an electrons to photons conversion target. The counting phase starts after irradiation is completely stopped, so as to not saturate the gamma ray spectroscopy system (and not between the accelerator pulses as when measuring neutrons issued from photofission). The gamma ray spectroscopy device is efficient at high energy, some delayed photofission gamma radiations being emitted with an energy of several MeV.

The expected detection limits, established by numerical simulation, for the measurement of long-lived radionuclides by photon activation in packages or samples of up to 220 liters, are lower than the acceptability thresholds of the Surface storage center (CA) for ^{59}Ni, ^{63}Ni, ^{79}Se,^{93}Zr,^{93}Mo, ^{99}Tc, ^{129}I, ^{135}Cs ; for example, for Iodine 129, the detection limit is less than 1Bq/g for a 220 liter cemented package and less than 50Bq/g for a compacted hull and nozzle package.

In order to carry out the R&D on photon interrogation (interrogation by induced photofission and photon activation), the CEA uses a test hall including a LINAC devoted to the control and characterization of radioactive waste packages. This is the SAPHIR demonstrator started end of 2000 on the center of Saclay (Figure 2).



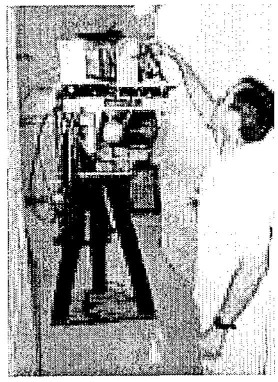


Figure 2 : The SAPHIR facility devoted to photon interrogation

3. MEASUREMENT METHODS FOR PHYSICAL PROPERTIES OF PACKAGES BY HIGH ENERGY PHOTON IMAGING

High energy photon imaging is different from X-ray tube imaging (medical and non-destructive control of materials) by its use, in the same way as photon interrogation, of a LINAC (LINear electron ACcelerator) coupled to an electrons to photons conversion target. An intense and very energetic radiation adapted to measurements by transmission in dense and very thick materials is thus obtained. The attenuation measurements of this radiation associated to the coordinates of the object in the acquisition space are the basis of imaging techniques. The associated detection devices must be adapted to the time, space and energy characteristics of the transmitted beam. The integration on a mechanical bench of these components and the rotation of the package under the beam allow measurements to be acquired under different incidence angles.

High energy photon imaging techniques, initially developed by the CEA for the air-space industry [6], are now being applied to the characterization of waste packages in order to allow large, high density and/or irradiating packages to be examined. The most attractive imaging techniques which have been identified with this objective in mind are radiography, tomosynthesis and high energy tomography. These non-destructive control methods by photon transmission allow the fine examination of the internal structure of the waste packages without any alteration to their physical integrity. Thanks to their flexibility, these methods allow the detection, localization and in some cases, characterization of internal defects, as well as the material density distribution to be established, the geometric conformity to be checked and dimensional measurements to be made. These three techniques are to be used together and each has specific characteristics :

- Radiography is operated using a « large field » detector which allows within a few seconds the acquisition of information on the whole package.
- Tomosynthesis uses the same « large field » detector as above but this technique focuses on the peripheral layers of the cylindrical packages (a few dozen cms). It can rather accurately localize the structures and/or defects within only a few minutes of examination.
- Tomography is the most efficient of all these imaging techniques : it provides a better analysis and this on the whole object under examination. The acquisition time is however much longer than for the two preceding techniques since the acquisition is very accurately collimated. Tomography imaging also provides a mapping of the linear attenuation necessary to global or segmented gamma ray spectroscopy. Active imaging thus, when coupled to nuclear measurement methods, provides a better quantification of the β and α activities downstream from the gamma ray spectrometric techniques and from neutron measurements.

Studies on high energy photon imaging include specific technological development, numerical simulation to dimension future facilities and evaluate their performances, and the achievement of a prototype facility.

Technological developments

Detection systems adapted to the examination of large packages have been developed :

- For radiography and tomosynthesis, a large field 800 mm × 600 mm detector coupled with a CCD camera [7] (Figure 3) : this system is very sensitive and the acquisition very fast. A treatment in real time of the measurements is made, particularly for tomosynthesis in order to increase the signal to noise ratio and provide a spatial localization of defects.

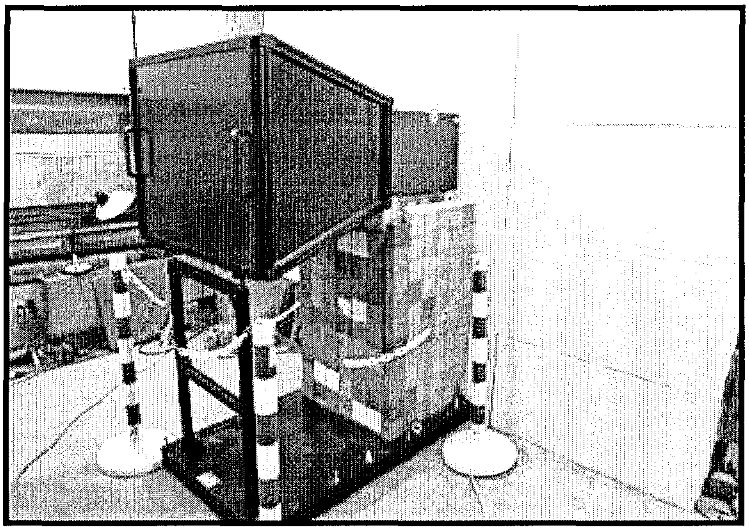


Figure 3 *: « Large field » detection device*

- For tomography, the detector is made of a multi-element detection block consisting of 25 CdTe crystals.

These detection systems have been characterized and their performances assesses by means of mock-ups.

Numerical simulation

A numerical simulation tool has been developed, allowing the modeling of LINAC photon production, attenuation in the measured object and then detection. It also can reconstruct the images obtained. This tool, called MODHERATO, was validated using experimental data . Figure 4 presents a simulation of the expected performances with an interrogating beam of 15 MeV on a concrete hull of 110 cm in diameter containing a concrete-steel-vinyl matrix with a density of 3.5 and the presence of defects in the steel of 10, 4, 2 and 0,5 cm in diameter.

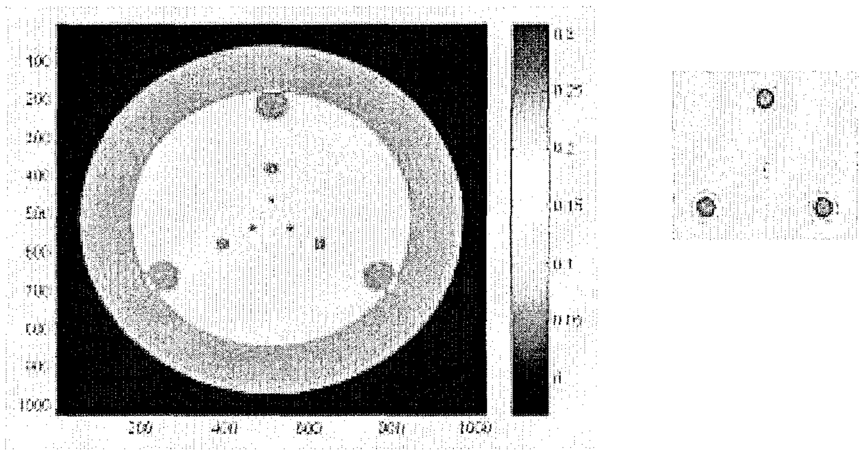


***Figure 4** : Simulation of the performance of high energy photon imaging*

Prototype

The building of a scale 1 prototype aimed at validating on actual waste packages the performances established to this day by simulation is foreseen at the end of 2002 in the Basic Nuclear Facility of CHICADE at the CEA/Cadarache. This measurement bench will integrate the « large field » and CdTe detection devices mentioned above. The association of these devices will provide a more or less rapid and complete imaging for long-lived waste, including irradiating and large volume packages of up to 2.2 m^3.

The MODHERATO simulation tool has enable the expected performances of the measurement bench to be established. The expected spatial resolution, which characterizes the minimum separation distance, is between 2 and 3 mm. Despite this, lower size defects can be detected but not characterized. In a homogeneous environment such as concrete, defects such as cracks of roughly one millimeter could be detected, depending on their direction and shape. The expected typical resolution in density, which characterizes the smallest detectable difference in density, is less or equal to the per cent in tomographic mode.

4. CONCLUSION

The improvements made in the classic nuclear measurement techniques, especially in the method coupling, and the technological breakthrough consisting in using an intense photon interrogating source issued from a linear electron accelerator (LINAC) have provided gains on the performances of non-destructive measurement methods for long-lived radioactive waste packages, in particular :

- for the quantification and localization of heavy nuclei (U, Pu...), photofission can improve the detection limits by one order of magnitude compared to active neutron interrogation ; the detection limit in a concrete drum of roughly one m^3 was thus lowered under the alpha activity acceptability threshold of the Aube surface storage Center (CA),
- for the measurement of long-lived radionuclides and toxic chemical elements, the photon and neutron activation make these measurements accessible in a non-intrusive way for a large number of packages or samples of up to 220 liters and the detection limits evaluated by means of numerical simulation are lower than the acceptability thresholds of the CA for the following radioelements : ^{59}Ni, ^{63}Ni, ^{79}Se,^{93}Zr,^{93}Mo, ^{99}Tc, ^{129}I, ^{135}Cs ; for example, for Iodine 129 the detection limit is less than 1Bq/g for a 220 liter cemented package and less than 50Bq/g for a compacted hull and nozzle package ; the foreseen performances for the chemical elements in small samples of roughly 1 kilogram are of a few ppm for Cd, a few dozen ppm for Hg and Cl and a few hundred ppm for Cr, Ni, As, Sb, and Se,
- for the physical inspection of the waste package structure (density, detection of heterogeneities) and content (detection of undesirable objects), high energy photon imaging should provide spatial resolutions of less than 3 mm and a density resolution of roughly 1% in tomographic mode for large (greater than 2m^3), high density (greater than 3) and/or irradiating packages.

Experimentation on actual packages, particularly for the validation of performances, is underway with existing prototype equipment (SAPHIR facility for photofission at the CEA/Saclay) or is scheduled in facilities under construction (high energy photon imaging at the CEA/Cadarache).

Finally, the integration of all these non-destructive measurement techniques into one and the same facility is under study so as to obtain the most thorough and accurate characterization means of long-lived radioactive waste [8].

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