CALIBRATION AND HOT TESTING OF THE ADVANCED NUCLEAR MEASUREMENT SYSTEMS USED FOR WASTE CHARACTERIZATION IN COGEMA'S NEW ACC COMPACTION FACILITY

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Spent nuclear fuel from commercial power reactors is reprocessed at the COGEMA plant in La Hague. After shearing and dissolution of the fuel assemblies, the hulls and nozzles are sent to COGEMA's new compaction facility (ACC) to reduce the final volume of waste. Technological waste generated in the reprocessing plant is also sent to the ACC facility.

Compacted waste is characterized by two measurement stations:

- a gamma spectrometry station,
- an active and passive neutron measurement station.

The main purpose of these measurement stations is to determine the guaranteed nuclear parameters of the compacted waste and their associated uncertainties:

- total U and Pu masses,
- Pu, Cm, and total alpha activities,
- ^{137}Cs, ^{90}Sr-^{90}Y, ^{241}Pu beta activities,
- decay heat.

After giving a description of the measurement stations, this paper will describe the qualification tests performed in the context of the ACC project.

The extensive calibration tests performed on site with different sources and different waste matrices will be described (approximately 500 neutron and gamma experiments).

Hot tests that were conducted after hot start-up at the end of 2001 and prior to the start of commercial operation will be also presented. A number of drums produced by the upstream facilities were introduced one by one into the ACC facility in order to avoid mixing of different fuel assemblies. This procedure allows comparison between characterization performed in the upstream facilities on the basis of fuel data available before processing and the measurements performed on the new ACC stations. These comparisons showed good agreement between the different methods of characterization and thus validated the innovative technologies and methods used by COGEMA for compacted waste generated by the ACC facility.

1 BACKGROUND

COGEMA's facilities in La Hague, France reprocess irradiated fuel assemblies from approximately 100 nuclear reactors around the world. After shearing and dissolution of the fuel assemblies, hulls and nozzles are compacted in COGEMA's new compaction facility which was commissioned in 2001. Compacted waste is packaged in standard-design waste drums, known as CSD-C canisters, for disposal. The radiological characteristics of the CSD-Cs are determined by extensive assessment with two non-destructive measurement stations: a gamma spectrometry unit with five germanium diode detectors and an active and passive neutron measurement station with 249 neutron counters and two new-generation neutron generators.

The extensive calibration tests performed on site with different sources and different waste matrices and hot tests which were carried out after hot start-up at the end of 2001 and prior to the start of commercial operation are described here.

2 DESCRIPTION OF COMPACTING PROCESS

After shearing and dissolution, the fuel assembly hulls and nozzles fall into drums measuring approximately 1 m in diameter and 1.5 m high. The mean bulk specific density of the structural waste in the drums is about 0.8 kg/l. The drums are taken through the compacting facility entrance and checked by gamma spectrometry and by active and passive neutron measurement identical to that performed at the exit from the ACC facility. The hulls and nozzles are then emptied into a metering separator unit that fills 80 l metal containers. After drying, the waste is compacted in a 2500 ton press and the compacted containers, known as disks, are inserted into standard-design CSD-C canisters which are closed with weld-sealed lids. The CSD-Cs may also contain disks of technological waste that are not suitable for surface storage. The radiological characteristics of the CSD-Cs are checked at the facility exit by gamma spectrometry and by active and passive neutron measurement identical to that performed at the entrance to the ACC facility.

3 MEASUREMENT OBJECTIVES

The measurement cells have already been described in the document in Reference 1, but are mentioned here to improve understanding of the qualification and calibration tests which are the subject of this document.

3.1 INCOMING MEASUREMENT STATION

This measurement station located at the compacting facility entrance is used to:

- determine the fissile mass in the drums and compare it with a threshold to ensure that all areas remain subcritical up to the intermediate storage area at the facility exit,
- ensure similar determination of characteristics for all types of drums produced in the reprocessing facilities,
- determine the structural waste residue unit (URSD) inventory for each COGEMA La Hague customer.

The URSD inventory, which depends on the alpha and beta activity of the waste package, enables balanced distribution of CSD-Cs for the return of waste to its country of origin.

3.2 OUTGOING CSD-C MEASUREMENT STATION

This measurement station located at the compacting facility exit is used to:

- determine the fissile mass in the CSD-Cs and compare it with a threshold to ensure that subcritical conditions are maintained in the downstream facility intermediate storage area,
- characterize the final CSD-Cs by determining guaranteed parameters (activity and mass with associated threshold for each parameter) and complementary parameters expressing the specifications for the waste repository contractors (ANDRA in France),
- determine the URSD rate, which enables balanced distribution of CSD-Cs for each COGEMA La Hague customer.

The threshold which have to be guaranteed are as follows :

Guaranteed Parameters	**Threshold acceptance**
Beta activity of ^{137}Cs	65 TBq
Beta activity of ^{90}Sr-^{90}Y	115 TBq
Beta activity of ^{241}Pu	75 TBq
Alpha activity of Pu	3,3 TBq
Alpha activity du Cm	2 TBq
Alpha activity of > 50 years half life emitters	4,2 TBq
Decay Heat	90 W

4 THE MEASUREMENT STATIONS

Figure 1 shows the general layout of the CSD-C outgoing measurement station.

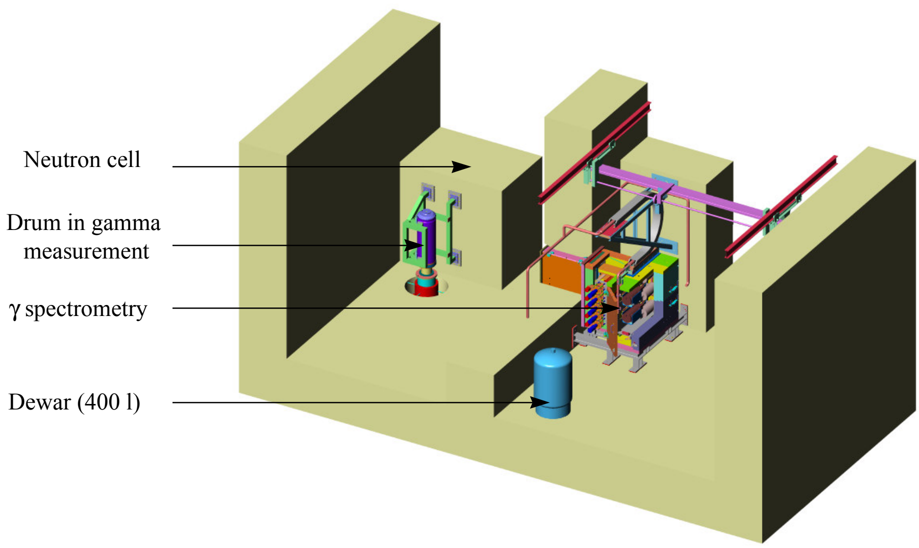

Figure 1

4.1 GAMMA SPECTROMETRY

The gamma spectrometry station is of the same design as the incoming measurement station. However, because of the amount of CSD-Cs to be handled, five detectors have been installed to perform all CSD-C measurements in a single operation. A 2-minute preliminary measurement determines the total duration of drum measurement and the detection limits for the specified peaks. Total measurement time ranges from 15 to 45 minutes.

4.2 NEUTRON MEASUREMENT

The neutron measurement station uses the same instrumentation as the station at the facility entrance. Since the CSD-Cs have a smaller diameter than the incoming containers, the detection blocks have been installed closer together, which provides a cell detection efficiency of more than 20%.

Figures 2 and 3 show the vertical and horizontal views of the measurement station.

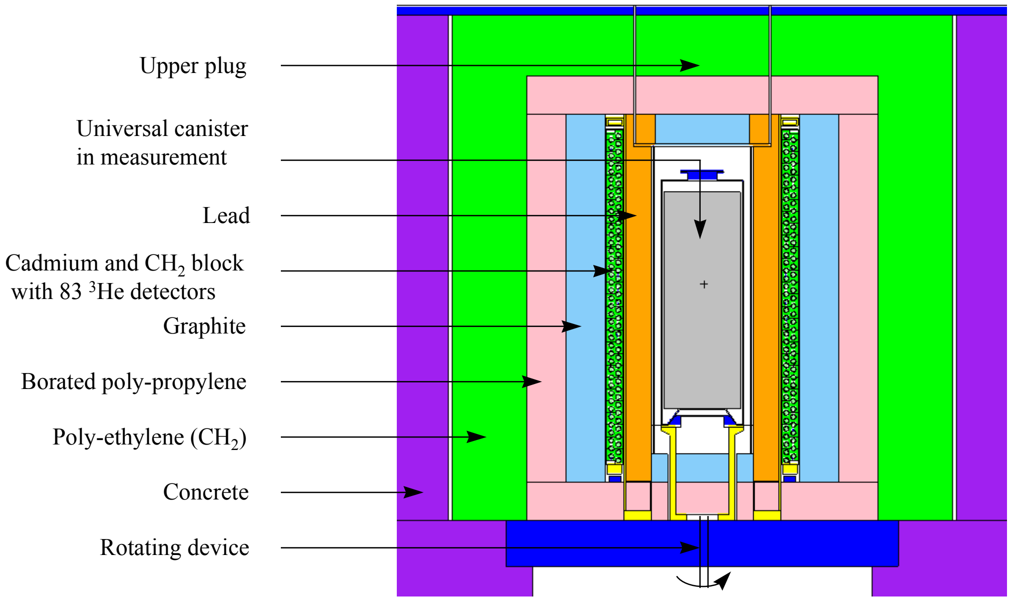

Figure 2

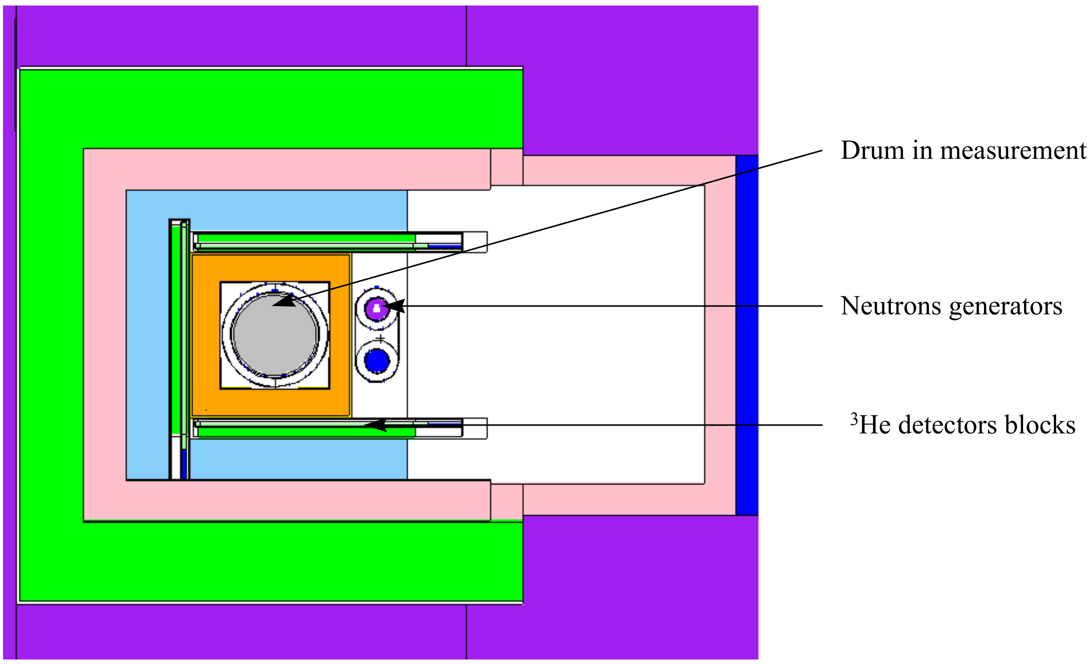

Figure 3

The outer layers of the cell protect the neutron measurement section from gamma rays and neutrons emitted by CSD-Cs outside the measurement cell. A layer of concrete attenuates the gamma rays, a polyethylene layer slows down the neutrons and a borated polypropylene layer absorbs the thermalized neutrons. The measurement cell is therefore insensitive to outside radiation after the borated polypropylene layer.

The inner layers of the cell perform the following functions. The lead shielding is dimensioned to maintain the gamma dose rate at the ^{3}He neutron detectors below 2 rad/h. The graphite layer slows down the neutrons and was selected because it has a lower capture capacity than polyethylene. Once the epithermal neutrons have been thermalized, after going through a thin layer of cadmium, they enter the polyethylene detection blocks. Then the neutron produces the following reaction:

$$^{3}He + n => {}^{1}H + p + Q \qquad \text{where } Q = 765 \text{ keV}$$

The proton is detected. An electronic circuit with current amplifiers and a very fast acquisition card developed by CANBERRA-EURISYS are used to count each signal coming from one neutron extremely rapidly.

5 COMBINED INTERPRETATION OF NEUTRON AND GAMMA MEASUREMENTS

A combined interpretation of neutron and gamma measurements is used to determine CSD-C characteristics at the compacting facility's outgoing measurement station. The measurements are interpreted in three stages:

1. interpretation of gamma measurements,
2. interpretation of neutron measurements,
3. combined interpretation of neutron and gamma measurements to determine CSD-C activity in terms of mass and decay heat.

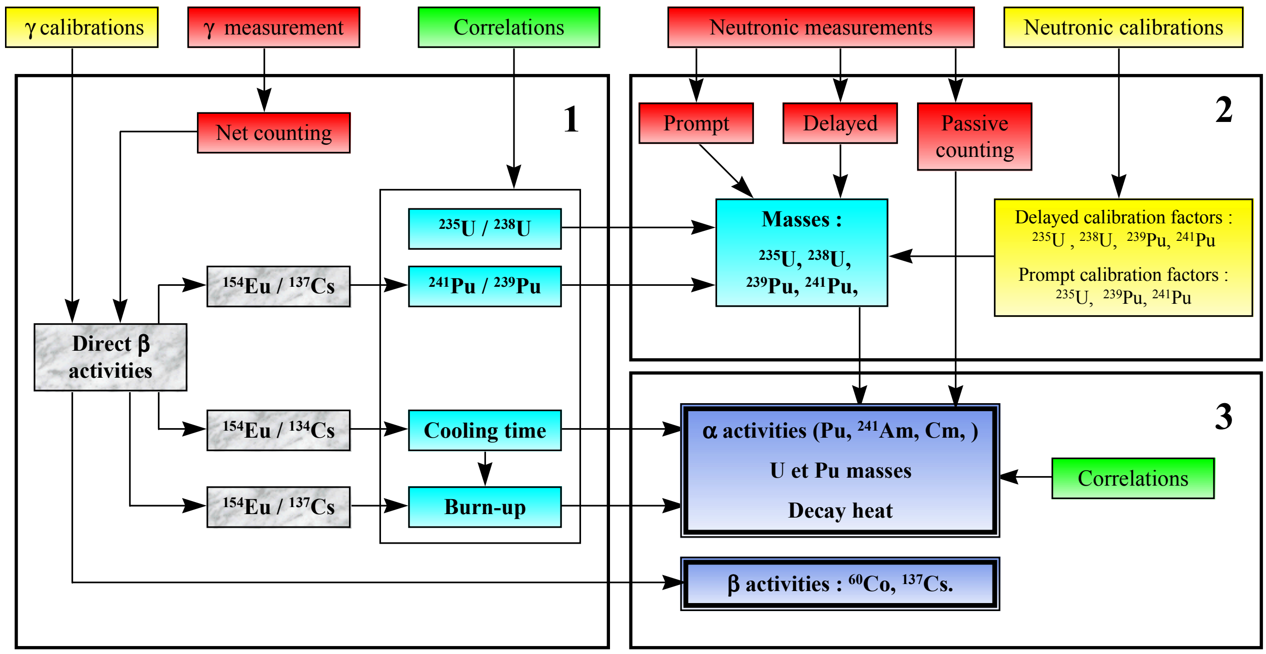

Figure 4

All the necessary correlations were established using the CESAR depletion code (see Reference 2).

6 CALIBRATION OF THE OUTGOING MEASUREMENT STATIONS

6.1 METHODOLOGY USED FOR CALIBRATION AND PARAMETERIZATION

The stations are calibrated to adapt the calculation diagrams used to the MCNP code (see Reference 3). For while it is true that the MCNP code is widely used and implements qualified basic data, the incoming and outgoing Stations have a number of special features requiring numerous experimental calibration operations. Concerning the outgoing Station, this involves gamma spectrometry measurement of a high density package. A slight difference in procedure has a considerable effect on the calibration coefficients. Interpretation of the neutron measurement stations involves extracting background noise values that cannot be determined by calculations. Furthermore, some of the matrices making up the CSD-C can have a serious effect on measurement (factor of 10) and require that the code be adjusted. In addition, the reaction of the acquisition electronics cannot be modeled. Thus, after numerous experimental adjustment operations, the results obtained using the code can be used to determine the appropriate calibration coefficient values for all the foreseeable operating configurations.

The calibration operations carried out for the outgoing station are described below. The same operations are carried out for the incoming station.

6.2 CALIBRATION OPERATIONS CARRIED OUT FOR THE GAMMA SPECTROMETRY STATION

The activity of a radionuclide X can be expressed as follows:

$$\mathbf{A_i\,(X) = \frac{S_i(E)}{\varepsilon_i(E).FT_i(E).I(E)}}$$

In this formula, $\varepsilon_i(E)$ is the intrinsic efficiency of the zone i detector at energy E and $FT_i(E)$ is the zone i transfer function, also known as the geometric efficiency. These two quantities have to be determined by calibration.

Thus, the first stage of the calibration operations consists in determining the intrinsic efficiency of each detector using a multi-ray source. ^{152}Eu is usually used for the energy range involved here (0.5 MeV to 1.5 MeV).

The second stage consists in determining the geometrical efficiency. A titanium disk was specially created for the purpose (since it has a relative density of approximately 4.5, see Figure 5). Five holes were drilled in it along five different radii in order to validate the attenuation function for each radial position of the source. Likewise, all the collimators were calibrated to ensure that they were suitable for the count rates that would occur.

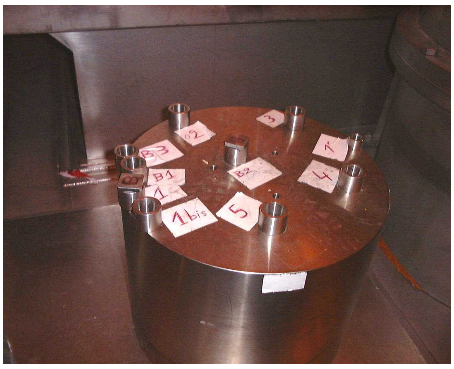

Figure 5 -
Titanium disk with holes for inserting sources along different radii

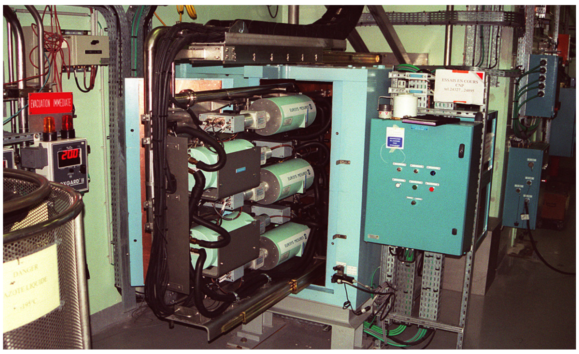

Figure 6 -
Layout of the five Ge detectors being calibrated

Various sources covering the energy ranges likely to occur were used for the calibration operations (^{60}Co, ^{137}Cs and ^{152}Eu with different activities).

The sources used had activities of between 100 and 500 MBq, producing a variety of count rates depending on their positions in the disk.

A testing ^{134}Cs source with an activity of a few tens of kBq was placed against the detector to check the linearity of the acquisition system.

6.3 CALIBRATION OPERATIONS CARRIED OUT ON THE NEUTRON STATION

As can be seen in Figure 4, the innovative feature of the neutron station is that it can make three different neutron measurements, not only within the one measurement cell but also within the one matrix (this system has been patented by COGEMA-CEA, see Reference 4):

- passive neutron measurement

- active measurement by measuring the prompt neutrons produced during fission caused by the neutron generator

- active measurement by measuring the delayed neutrons produced during fission caused by the neutron generator

These three measurements make it possible to determine the separate contributions of uranium, plutonium and curium.

The calibration operations are therefore designed to determine the following for different quantities of absorbent material (the stainless steel of the nozzles and the Inconel of the grids), technological waste matrices and technological waste depths in the canisters:

Efficiency and linearity of the cell by passive measurement

Calibration coefficients for acquisition of prompt neutrons (in ^{235}U) and delayed neutrons (in ^{235}U et ^{238}U) with verification of response linearity.

Active spatial measurements of prompt and delayed neutron response variation as a function of source position (with enriched or depleted uranium)

Calibration coefficients for the acquisition of prompt neutrons from ^{239}Pu and ^{241}Pu

Experimental calibration was carried out on nine different matrices with various neutron absorber contents (stainless steel and Inconel) and different types of technological waste (aluminum, PVC, polyethylene, carbon steel etc.) (see Figure 8).

The density of around 4.5 g/cm^3 was obtained using various cubes whose sides measured 2.5 cm, as shown in the photos below.

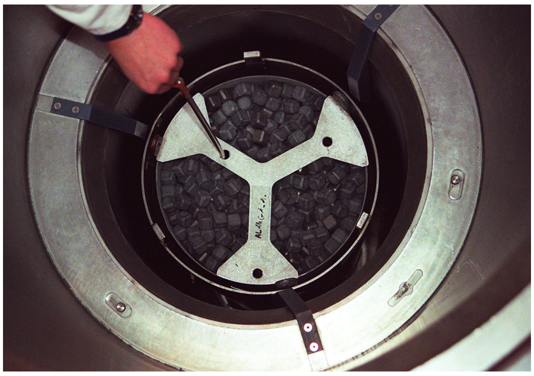

Figure 7 -
Placing a rod in a calibration CSD-C

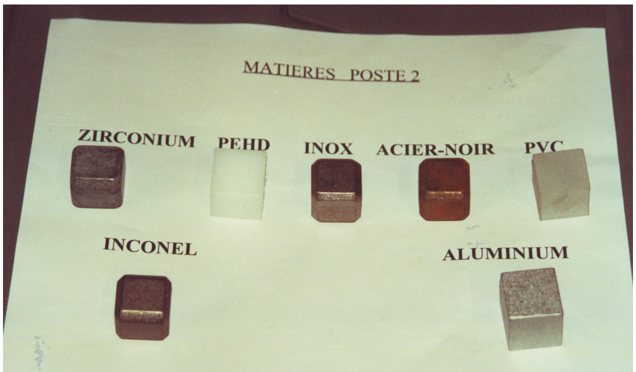

Figure 8 -
The various materials used at Station 2.2

The following sources were required for these calibration operations:

^{252}Cf sources of between 10^4 and 10^6 n/s

rods enriched to 3.5% with ^{235}U, each containing ~10 g of ^{235}U

rods capable of holding up to 9 capsules containing uranium pellets enriched to 3.5%, each capsule containing approximately 1 g of ^{235}U (high mass rods)

rods capable of holding up to 9 capsules containing uranium pellets enriched to 3.5%, each capsule containing approximately 0.1 g of ^{235}U (low mass rods)

rods capable of holding up to 9 capsules containing uranium pellets depleted to 0.25%, each capsule containing approximately 30 g of ^{238}U

rods capable of holding up to 9 capsules containing MOX pellets, each capsule containing approximately 1 g of ^{239}Pu and approximately 30 g of ^{238}U

The rods could be inserted along three different radii (as shown in Figure 7). Since the drum was rotated while the measurements were being made, the rods simulated three radial zones in the CSD-C. Since the capsules could be placed at nine different heights, the CSD-C could be divided into 27 virtual positions (three radial sections and nine heights).

Passive calibration of an empty cell revealed that cell efficiency was 22%.

Firing into an empty cell made it possible to determine neutron lifetime in the blocks and in the cell (Zones 1 and 2 respectively in Figure 7).

- Lifetime in blocks: 58 µs
- Lifetime in cell: 335 µs

With CSD-C, and depending on the configurations, these lifetimes should be reduced as follows:

- Lifetime in blocks: 30 to 40 µs
- Lifetime in cell: 250 to 300 µs

The absorbent materials in the CSD-C reduce the neutron lifetimes.

The table in Figure 9 below shows the different calibration operations carried out for the nine configurations.

Type of calibration	**Configuration 1**	**Configurations 2 and 3**	**Configurations 4 to 9**
Passive calibration	27	9	9
Effect of matrix on interrogating neutron flux	1	1	1
Spatial, prompt neutrons	27	5	5
Spatial, delayed neutrons	27	5	5
Mass measurements ^{235}U prompt and delayed	9	3	1
^{238}U delayed	9	3	1
Linearity	3		
^{239}Pu and ^{241}Pu prompt and delayed	3	3	1
Total for one configuration	106	29	23
Total for all configurations	106	58	184
Overall total	**approx. 350**		

Figure 9

For each configuration, passive calibration consisted in positioning the ^{252}Cf source along different radii and at different heights, i.e. 3x9=27 positions for Configuration 1 and fewer points for the other configurations.

Spatial calibration for prompt and delayed neutrons was carried out with capsules containing enriched and depleted uranium respectively. These two measurements were used to establish the prompt and delayed calibration factors of Uranium 235 and uranium 238 (CP[U5], CR[U5] and CR[U8]) as shown in figure 4. The measurements were made for the 27 positions of Configuration 1 and to a lesser degree for the other configurations.

Mass measurements comprised calibration operations similar to spatial calibration but with each of the three rods containing just one capsule. Nine calibration operations were carried out (at each of the nine heights) with enriched uranium rods and depleted uranium rods. Less thorough calibration operations involving 1, 2 and 3 whole rods were carried out for Configurations 2 and 3 and with only 3 rods for the other configurations.

Measurements were also made to check linearity of cell response for fissile matter masses of between 1 and 30 g (one rod weighing 1 g to three rods weighing 10 g).

Lastly, measurements were made with 1, 2 and 3 rods containing nine MOX capsules to compare the prompt and delayed signals with the values of the various ^{235}U, ^{238}U, ^{239}Pu and ^{241}Pu contributions calculated by MCNP.

In all, around 350 calibration operations were carried out.

7 QUALIFICATION

Stations 0 and 2 were subjected to stringent qualification. The various topics of qualification are shown in Figure 10.

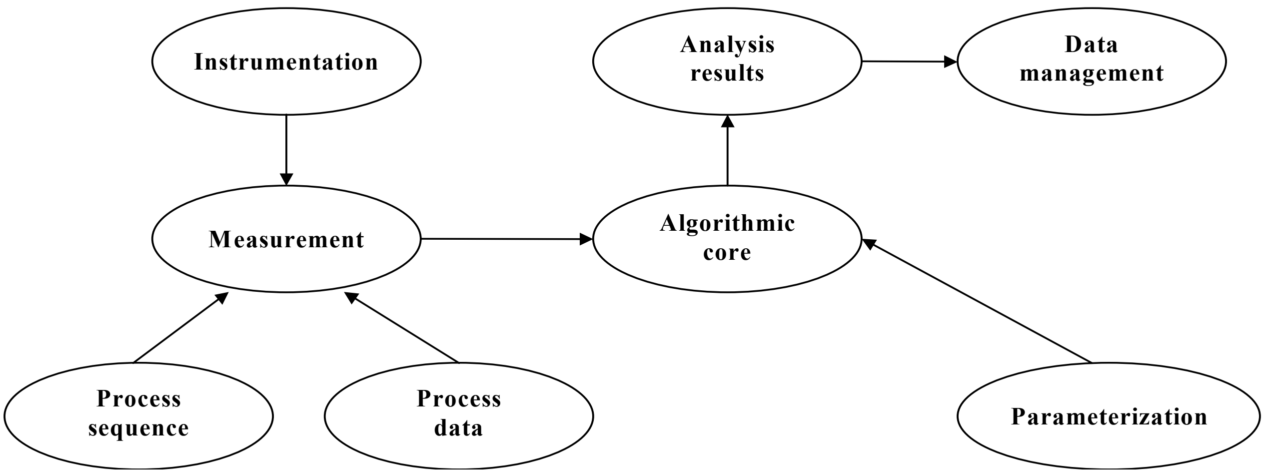

Figure 10

The instrumentation was tested and adjusted several times before calibration began.

Exchanges between machines (for data relating to nuclear monitoring of the process exchanged between PLCs and the control station) were platform tested and then re-tested during hot qualification of the facility. The process sequence and data tests were used to validate the measurement.

Interpretation of the measurement was validation by extensive qualification of the algorithm. Synthetic gamma and neutron measurement spectra were created. These measurement spectra contained a given number of counts in the regions to be interpreted. The counts corresponded to given activities that had to be determined when the measurements were being interpreted. Thus it was possible to validate the entire interpretation system by means of physical and computer tests.

The actual parameters of the measurement stations were tested during hot qualification of the facility. Several tens of thousands of values were involved.

8 HOT QUALIFICATION

8.1 PRINCIPLE OF HOT QUALIFICATION

Hot qualification consisted in selecting 18 actual drums containing hulls and nozzles produced during the last five years of plant production and offering as wide a variety as possible (drums with low and high burn-ups, long and short cooling times, hulls from only one or several types of fuel assemblies etc.).

The drums were measured at the incoming station before being compacted and the hulls in the drums were traced through the facility, meaning that the production rate had to be very slow. The compacted disks were put into CSD-Cs which were measured in the facility outgoing station. The incoming and outgoing values were measured to check the complex measurement analysis system.

8.2 OBJECTIVES

The objectives of hot qualification were as follows:

1. To validate analysis of the measurements made in the incoming and outgoing stations of the compacting facility.
2. To assess the extent to which the radiological contents of the drums were homogeneous before and after compacting, by carrying out more thorough analyses than during normal operation of the facility. This more thorough analysis made it possible to locate radioactive material and fissile material.
3. To check the validity of the hypotheses made at the outgoing station where the results are analyzed with a minimum of information as to the origin of the hulls.

8.3 RESULTS

8.3.1 Criticality safety

The amount of fissile material estimated in the incoming station is perfectly consistent with that obtained for the outgoing station. In addition, the fissile material in the drums in the incoming station appears to be homogeneous. This homogeneousness is also evident for the CSD-Cs in the outgoing station, particularly if they contain disk of compacted hulls taken from the same drum.

8.3.2 Gamma spectrometry

As far as gamma spectrometry was concerned, the results recorded at the incoming and outgoing stations differed by less than 15%, which falls within the measurement uncertainty range for packages with this density (approximately 4.5g/cm^3 in the outgoing station).

8.3.3 Guaranteed activities and parameters

The hypotheses made for measurement analysis were confirmed for the 18 drums used for hot qualification.

Furthermore, the reasonably upper bound nature of the CSD-C package specification was justified.

9 CONCLUSIONS

The non-destructive measurement stations in the hull and nozzle compacting facility at the La Hague plant are highly innovative. They are the most innovative industrial-scale nuclear measurement stations for such waste packages in the world.

The main innovations are :

- the optimization of the neutron measurement cell to separate U and Pu signal contribution;
- the development of a new neutron generator with an emission of $2x10^9$ n/s;
- the development of a combined gamma spectrometry and neutron measurement interpretation;
- the development of an innovative uncertainty propagation technique for the interpretation of the measurement.

Furthermore, thorough qualification of the stations has made it possible to demonstrate the quality of the measurements and their analysis using real packages (hot qualification).

This large development project has also considerably increased the level of expertise in nuclear measurements of the various companies in the AREVA Group (SGN, CANBERRA-EURISYS and COGEMA).

10 REFERENCES

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