EXAMPLE OF A QUALITY CONTROL OPERATION PERFORMED ON A NUCLEAR REACTOR WASTE PACKAGE

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ABSTRACT

A nuclear waste package presents a finished product of such complexity that, until now, the state of the art has failed to permit a complete a posteriori industrial inspection of all the parameters involved in the definition of package quality. The Quality Assurance and Control system set up by ANDRA is hence based on the principle of a priori inspection of all the key components of the 'waste production/packaging/disposal' line.

A specific effort was therefore made concerning detailed knowledge of the raw wastes, inspection of the application of particular specifications for packaging and coating materials, and monitoring of the procedures implemented for the production of immobilization matrices and packages.

However, the need emerged to secure the possibility of the spot sampling and inspection of a package, in the production line or on its arrival at the disposal center. This external inspection (or superinspection) has a threefold objective:

- to check the conformity of the package and of all its components with the mechanical, chemical and radiochemical specifications,
- to ensure, by thorough inspection, that the product is similar to the product which was characterized and qualified, and which secured ANDRA's approval of the process,
- through the detection of any variations and discrepancies, to advise the process promoter and the research laboratories, so as to improve the process, the product and the specifications.

This paper describes an operation of this type conducted in 1982 by ANDRA, with the help of the CEA's laboratories and technical units, on low and medium-level waste packages produced by an industrial installation in an EDF nuclear power plant. The following are described: dismantling and sampling methods implemented, means employed, and the specific characteristics tested and the first results obtained.

However, since the state of progress of the analyses do not allow the presentation of an overall report on package quality, this will be covered by a subsequent paper.

DESCRIPTION OF THE PACKAGES

The radioactive waste packages inspected by ANDRA resulted from the packaging of high-level spent ion exchange resins from a PWR reactor in an organic polymer matrix.

After introduction into a metal drum, the mixed ion exchange resins were spin-dried and then immobilized in a polymer. The previous assembly placed at the center of a reinforced concrete container (see Fig. 1) was blocked in this envelope by a high density mortar offering the biological shield required for transport (package external contact dose rate $\leq$200 mrem/h). The biological shield could be reinforced locally by means of lead plates a few centimeters thick.

The immobilized ion exchange resins were of different origins and activities (including the primary water purification circuits and steam generator purge treatment), so that the package displayed variable activity levels. The specific activities contained were estimated at about 100 Ci/m^3 for the first two packages and at 150 to 200 Ci/m^3 for the second pair.

The main radionuclides thus immobilized were cobalt 58 and 60, cesium 134 and 137, and manganese 54. The contact dose rate of the bare matrices was estimated at about 150 rad/h.

DESCRIPTION OF THE OPERATION

Objectives

This inspection was intended to achieve the following.

(a) To obtain maximum data on the characteristics of this type of matrix, especially:

- filling rate of the primary container,
- matrix homogeneity,
- mechanical strength,
- cohesion between waste and matrix,
- radioisotope distribution,
- leaching resistance.

(b) To exploit the experimental aspect for a better definition of the means, the procedures,

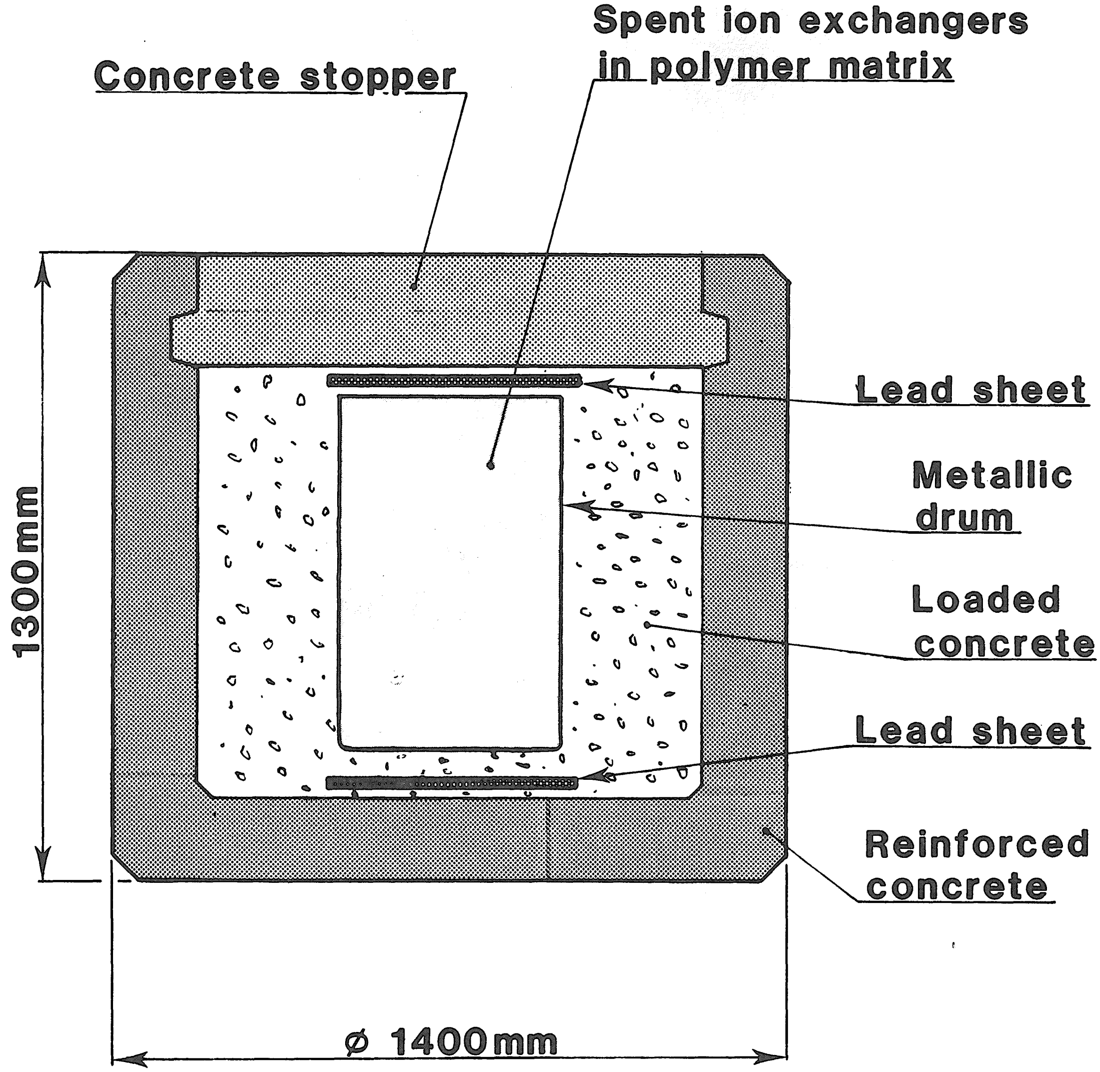

Fig. 1. Section of the Package.

and the radiological protection systems required to systematize this type of operation.

Sampling of Packages

Among the packages delivered to Centre de la Manche, ANDRA selected, for this inspection, packages containing medium-level irradiating wastes, rich in cobalt isotopes, with a significant Cs^{137} activity. The operation dealt with four packages.

Nondestructive Testing

A detailed visual examination showed that the apparent outer condition of the packages was satisfactory. No cement cracks or flakes were detected, particularly at the plug.

The external irradiation dose rates were measured systematically at various places. An external irradiation dose rate chart was prepared, followed by gamma spectrometry, in the testing facility of Centre de la Manche (Figs. 2 and 3).

These measurements were designed to allow the preparation of the dismantling and coring plan, and also to estimate the Co^{60} and Cs^{137} contents, and hence to predict the scale of the means to be implemented and the sensitivity of the radioactive measurements that could be made on samples: determination of homogeneity and radioisotope distribution, specific activities, and leaching resistance tests.

Dismantling and Coring

After transport to the Saclay Nuclear Research Centre, the internal metal envelope was partly exposed (grinding, cutting) in the installations of the Radiological Protection Department (SPR) followed by samplings of the matrices by axial and side coring (50 and 80 mm diameter diamond bits).

The cores were obtained without special protection, directly through the concrete shell playing the role of a shield, as the matrix could not be completely exposed due to the presence of the heavy concrete biological shield. The core positions were selected to allow the examination of different zones of the matrix: top, bottom, core and periphery (Fig. 4).

- Equipment

The operation was performed in an SPR operations bay allowing the access of trucks, fork-lifts, and equipped with an overhead travelling crane. The corer was installed on a metal frame about 2.50 m high for vertical coring, and the packages were positioned vertically under the drill bit by the crane.

- Coring

The variety of the successive materials to be penetrated to obtain a matrix core required many changes in the cutting conditions. A dry coring test proved futile, and it was, therefore, decided to use water as a lubricant and coolant. This was because, during dry cooling operations, we observed jamming of the cutter without the possibility of recovery, as well as softening of the matrix.

- Inspection of bit coolant

The volume of water used to cool the cutter during coring was estimated, and a sampling corresponding to each package was subjected to gamma spectrometry analysis before and after filtration through a 0.45 μ porosity cellulose diaphragm. Figure 5 shows the results obtained.

TESTS AND ANALYSES

These were conducted jointly by the Concrete and Coatings Laboratory and the Spent Fuel Research Laboratory (work on high-level materials), both located at the Saclay Nuclear Research Centre. The different test procedures were developed jointly with the Containment Evaluation and Inspection Bureau, located at the Cadarache Research Centre, and ANDRA. The tests were based either on existing standards, or on procedures set up specially for this type of inspection.

All the tests and analyses applied to the cores from the two first packages (Nos. 1 and 2) were conducted in hot cells equipped for determinations of the physiochemical, radiochemical, mechanical properties etc., of radioactive materials.

Description of the Facilities

The inspection facility, which featured several hot cells, is distinguished by the following:

- low 'background noise',
- easy decontamination,
- no interference with an external contamination source (conveyor, more contaminated adjacent cell),
- large volume, capable of accommodating a satisfactory number of experimental systems,
- easy manipulation,
- ready access favoring inter-laboratory and inter-cell transfers.

The experimental systems employed and grouped together in two cells were essentially the following:

- periscope for visual inspection and filming (X6),
- 400 daN lifting unit,
- three telemanipulators, including one with 80 daN capacity,
- one 240 mm diameter transfer plug,
- one transfer carriage,
- metrology (dimensional and weight measurements),
- one gamma spectrometry installation (possibilities of working in translation motion or rotation on cores 75 mm in diameter and 800 mm long),
- one automatic bending machine (1200 and 6000N),
- one 300 kN compression machine,
- leaching stations.

The effective dimensions of these cells, which had cast iron biological shielding, were 2.10 x 2.80 x 4.85 m. These facilities were supplemented by gamma spectrometry and alpha/beta/gamma counting installations.

Measurements

This technical capability was used to perform the following examinations:

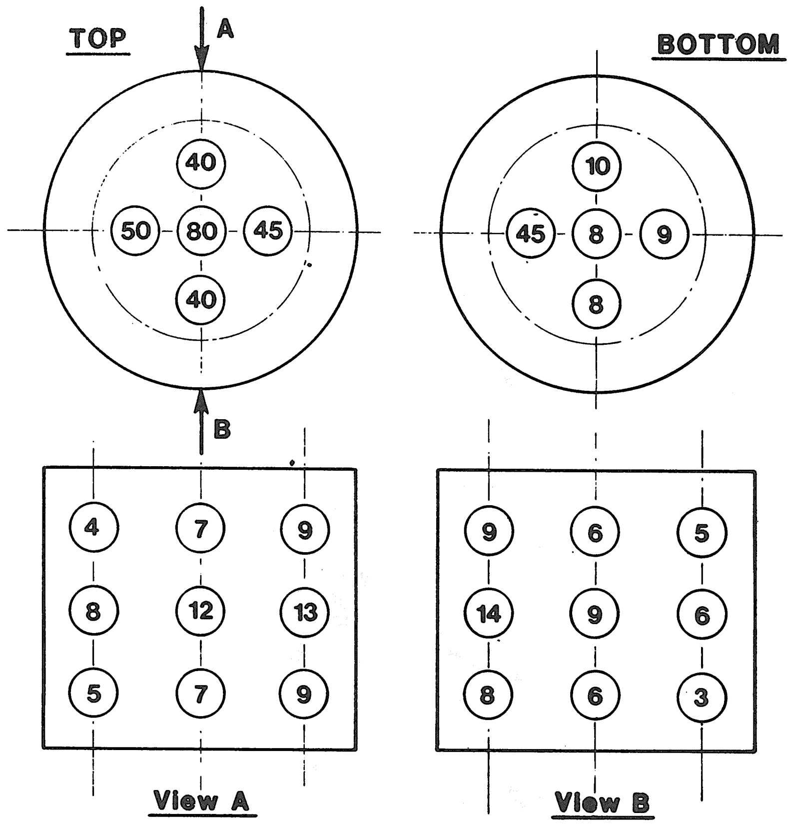

Fig. 2. Distribution of Surface Dose Rates (m rem/h) on the 1st Package.

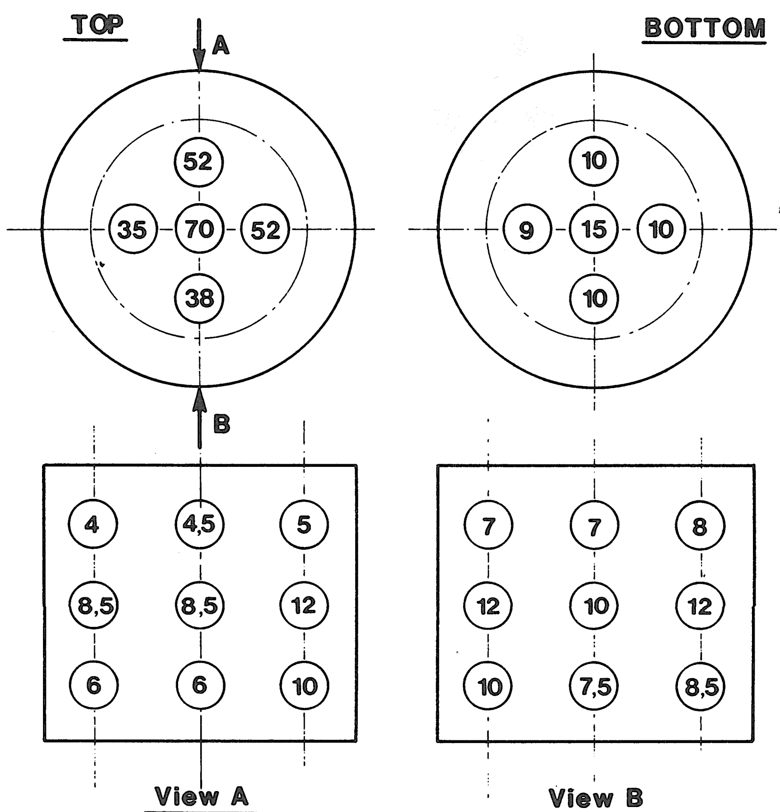

Fig. 3. Distribution of Surface Dose Rates (m rad/h) on the 2nd Package.

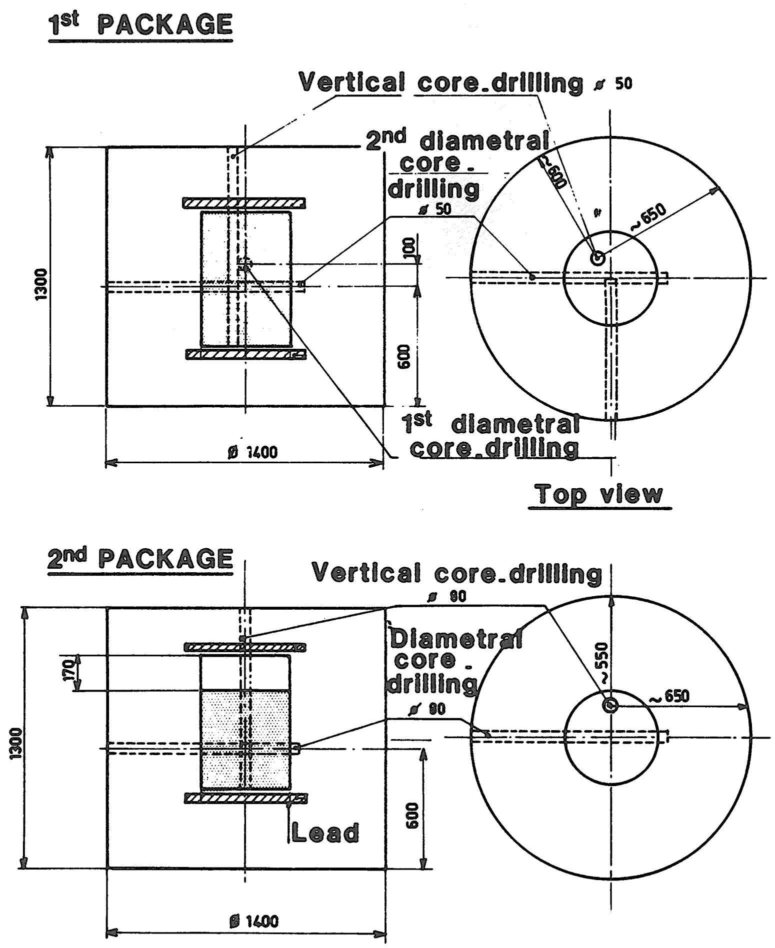

Fig. 4. EDF Packages Core Drilling Diagrams.

RADIONU- -CLIDES	PACKAGE n° 1				PACKAGE n° 2			
	A		B		A		B	
	1*	2	3	4	5	6	7	8
^{134}Cs	7.10^{-3}	6.10^{-3}	$5,7.10^{-3}$	$5,7.10^{-3}$	4.10^{-3}	4.10^{-3}	$4,3.10^{-3}$	$4,3.10^{-3}$
^{137}Cs	17.10^{-3}	19.10^{-3}	$12,4.10^{-3}$	$12,2.10^{-3}$	10.10^{-3}	9.10^{-3}	$8,2.10^{-3}$	8.10^{-3}
^{58}Co	40.10^{-3}	60.10^{-3}	$1,2.10^{-3}$	$1,1.10^{-3}$	20.10^{-3}	20.10^{-3}	$0,2.10^{-3}$	$0,2.10^{-3}$
^{60}Co	600.10^{-3}	880.10^{-3}	$26,6.10^{-3}$	$26,7.10^{-3}$	330.10^{-3}	330.10^{-3}	$5,6.10^{-3}$	$5,4.10^{-3}$
^{54}Mn	170.10^{-3}	230.10^{-3}	$4,6.10^{-3}$	$4,5.10^{-3}$	100.10^{-3}	100.10^{-3}	$1,5.10^{-3}$	$1,5.10^{-3}$

(A) Non filtered water

(B) Filtered water

(*) Sample reference

Fig. 5. Tool Cooling-Water Analysis (Ci/m^3).

Weight and dimensional measurements

The table below summarizes the observations made.

Table I.

block No.	core	diam (mm)	length (mm)	weight (g)	CDR* (rad/h)
1	diametral 1.D1	50	195	290	20
	End flaking.				
	diametral 1.D2	50	430	760	3
	No material detachment.				
	vertical 1.V1	50	530	850	2
	End flaking, peripheral detachment, stratified lateral appearance.				
2	diametral 2.D1	80	450	2270	7
	No material detachment.				
	vertical 2.V1	80	330	1700	4.5
	No material detachment.				

See Figs. 6 and 7
* CDR = contact dose rate

- Visual examinations

These were generally achieved by macrographic inspection.

- Check of uniform radionuclide distribution

These tests were performed by a longitudinal measurement of the cores using a gamma spectrometer with a finely-collimated Gi/Li detector. These tests served to measure the activity of the different gamma radionuclides present in the cores by increments of 1 mm. Collimation was provided by a 100 x 1 mm rectangular window with 1 mm step-by-step movement. The spectra were recorded on magnetic tape, offering the possibility of determining the activity of any zone by summation. The initial activity A_0 of the leach samples was determined in this way.

Figures 8 and 9 show the plots obtained on cores 1.D2 and 2.V1 for cobalt 60. The specific activities for the different radionuclides are shown in Fig. 10.

- Mechanical properties

The value of the compressive breaking stress was mainly estimated by means of the 300 kN machine. The test specimens were 50 or 80 mm diameter cylinders with slenderness ratio 2.

- Leach tests

These tests were performed on specimens sampled by dry cutting of the initial cores. Table II gives the dimensional, weight and radiochemical characteristics of the leached sections.

Table II.

samples	block 1	block 2
diameter (mm)	53.6	83.4
length (mm)	80.0	66.0
weight (g)	130.0	305.0
volume (cm^3)	180.4	360.4
area (cm^2)	180.0	282.0
initial activity 137 Cs	$5.0 \cdot 10^{-1}$	1.3
A_0 in mCi:		
. 58 Co	1.1	2.8
. 54 Mn	4.0	9.5
. 60 Co	14.0	34.0

The following procedure was selected: static leaching in a 1 litre beaker, using drinking water (see Fig. 11) kept at 20°C and replaced on the third, tenth, twenty-fourth and fifty-second days (sequences of 3, 7, 14 and 28 days). A sample was taken for gamma spectrometry analysis at the end of each sequence. The results obtained, combined with the longitudinal determinations mentioned earlier, yielded the following results:

- ai — activity released per sequence and per radionuclide
- Σai — cumulative activity released per sequence and per nuclide
- $\frac{ai}{A_0}$ and $\frac{\Sigma ai}{A_0}$ — fraction released and cumulative fraction released (CFR) per radionuclide.

The numerical values obtained will be compared subsequently with those expected from leach tests on full-scale blocks (effect of volume/area factor, validity of scaling up from results on samples). The absence of alpha emitters was also confirmed by the spectrometric method. Conclusions will be drawn later from the overall numerical results obtained to date and partly processed.

MEDIUM- AND LONG-TERM TESTS ON FULL-SCALE MATRICES

Based on the lessons drawn from the foregoing experiment, we decided to supplement the inspection by medium- and long-term tests.

In order to assess the validity of scaling up of the values obtained on small samples taken by coring, we decided to set up the means to enable us to conduct tests on full-scale packages. To do this, and faced with the difficulties previously encountered in removing the biological shield, we had a package prepared using industrial procedures, but in a removable packing.

One of these matrices was mainly used for overall water effects, by a total immersion test for a number of weeks. Another will be used for full-scale and long-term leach tests.

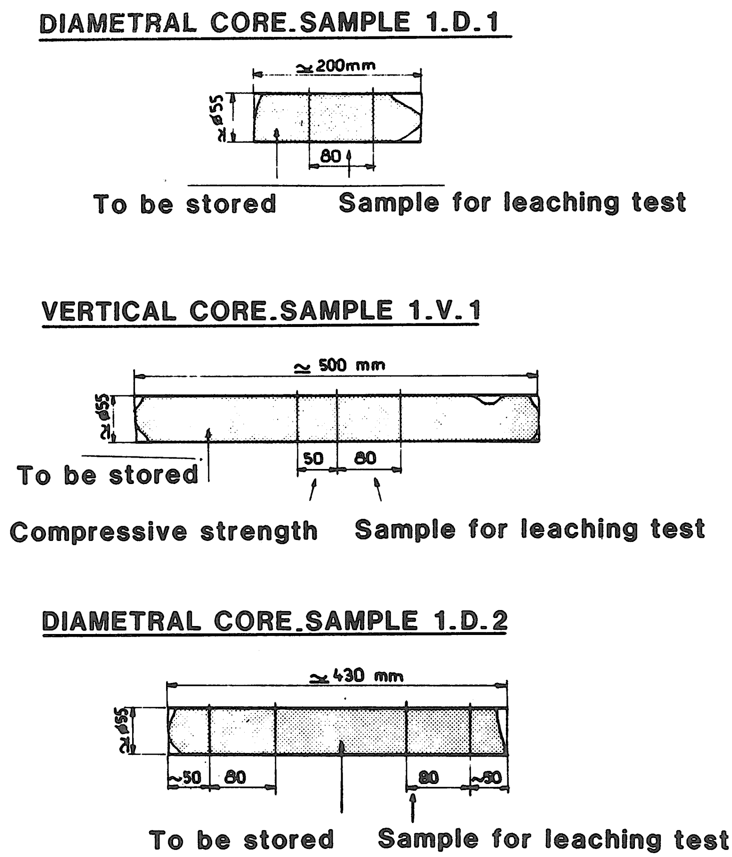

Fig. 6. EDF Package No. 1 Sawing Up Diagram.

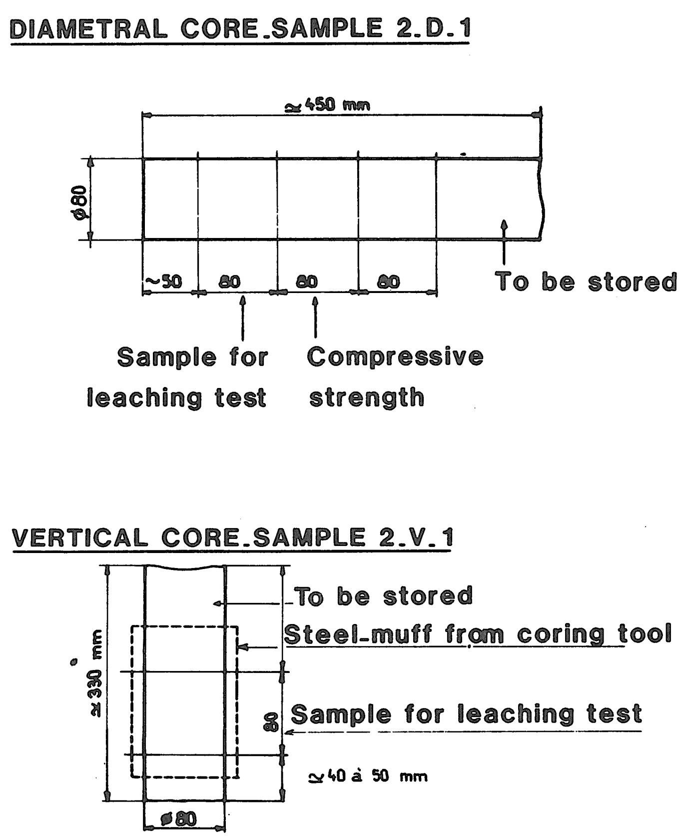

Fig. 7. EDF Package No. 2 Sawing Up Diagram.

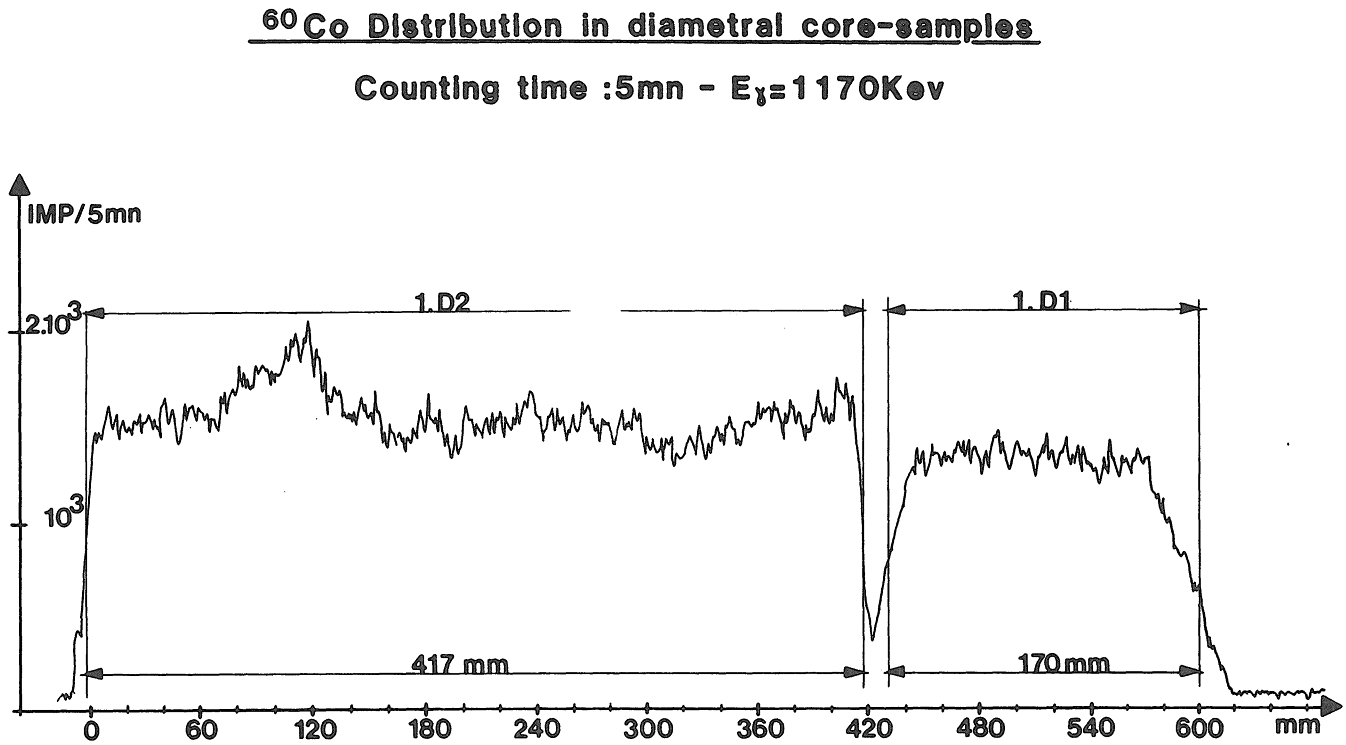

Fig. 8. EDF-Package No. 1.

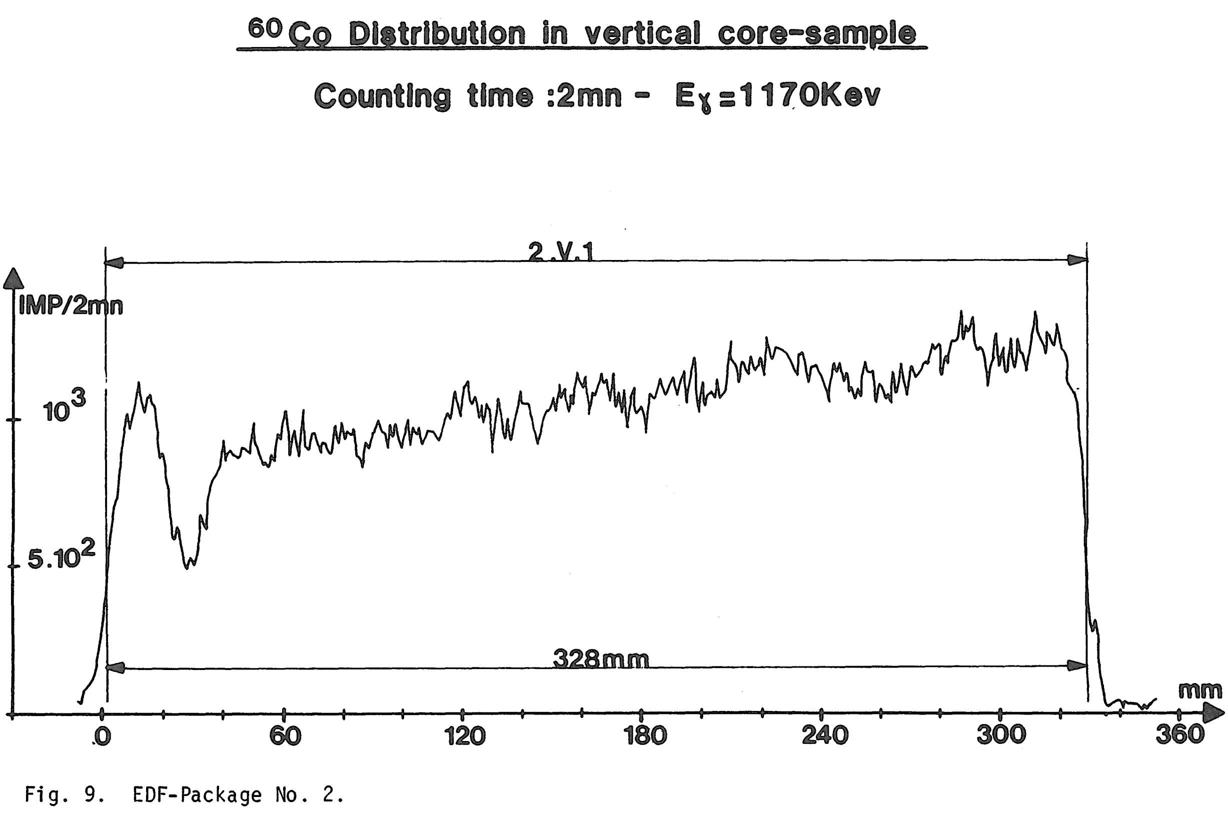

Fig. 9. EDF-Package No. 2.

Package Nb	Core-Sample	^{137}Cs		^{134}Cs		^{58}Co		^{54}Mn		^{60}Co	
		(1)	(2)	(1)	(2)	(1)	(2)	(1)	(2)	(1)	(2)
1	DIAMETRAL D1	0,88	$3{,}0.10^{-3}$	0,54	$1{,}9.10^{-3}$	1,9	$6{,}5.10^{-3}$	7,1	25.10^{-3}	25	86.10^{-3}
	DIAMETRAL D2	2,7	$3{,}6.10^{-3}$	1,5	2.10^{-3}	5,8	$7{,}7.10^{-3}$	21,5	28.10^{-3}	75	99.10^{-3}
	VERTICAL V1	3,3	$3{,}9.10^{-3}$	1,5	$1{,}8.10^{-3}$	7,0	$8{,}2.10^{-3}$	22,5	$26{,}5.10^{-3}$	74	87.10^{-3}
2	DIAMETRAL D1	6,8	$3{,}0.10^{-3}$	3,3	$1{,}5.10^{-3}$	16,8	$7{,}4.10^{-3}$	53,8	$23{,}7.10^{-3}$	188	83.10^{-3}
	VERTICAL V1	3,2	$1{,}9.10^{-3}$	2,1	$1{,}2.10^{-3}$	10,1	6.10^{-3}	33,2	$19{,}5.10^{-3}$	112	66.10^{-3}

Fig. 10. Activities of Core-Samples--(1) Whole activities (mCi), (2) Specific activities (mCi/g).

Potassium as K^+:	2,5 mg.l$^-$
Sodium as Na^+:	12 mg.l$^-$
Calcium as Ca^{++}:	85 mg.l$^-$
Chlorine as Cl^-:	28 mg.l$^-$
Carbonates as HCO_3^-:	225 mg.l$^-$
Sulfates as SO_4^{--}:	35 mg.l$^-$
Nitrates as NO_3^-:	18 mg.l$^-$
Phosphates as PO_4^{---}:	traces
Dry residues:	309 mg.l$^-$
pH :	7,9 mg.l$^-$

Fig. 11. Chemical Composition of Leaching-Water.

Facilities

The following means were used to conduct this new series of tests:

- Dismantling

A hot cell equipped in particular with a group of telemanipulators (gripping capacities between 150 and 1000 dN), and a lifting unit capable of moving 1500 kg loads.

- Immersion

A 200 litre stainless steel tank, without water circulation, placed in the hot cell.

- Leaching

A full-scale leach test station using real packages, created in 1979 at the Saclay Nuclear Research Centre, was not designed to handle packages with contact dose rates over 10 rad/h. It was decided to set up new facilities (highly-protected leaching loop due to the high dose rate), designed to leach matrices between 0.1 and 0.5 m^3 in volume and with contact dose rates up to 200 rad/h.

Immersion Tests

The tests planned on this package essentially concerned its water behavior: swelling, wetting/drying cycle, radionuclide release etc. At the time of its production, the matrix subjected to immersion tests exhibited the following radiochemical characteristics: activity 200 Ci/m including:

- 24% ^{58}Co (T = 71 d)
- 16% ^{60}Co (T = 5.26 a)
- 23% ^{134}Cs (T = 2.05 a)
- 24% ^{137}Cs (T = 30.23 a)
- 13% ^{54}Mn (T = 303 d).

Removal of the biological shield to gain access to the matrix was carried out in about five hours in the new facility.

Before water immersion, the contact dose rate measurements yielded values between 45 and 65 rad/h (values obtained using a collimated babyline chamber), providing an idea of the homogeneity of the block. Immersion, prolonged for a few weeks, made it possible to observe the material's behavior in time, to examine the structural and dimensional stability aspect of the matrix, and to measure the activity released by the block by gamma spectrometry on successive water samples.

Leach Test

This test will be started shortly. The following test parameters have been selected:

- leachate: drinking water (see Figure) kept at 23 ±3 °C, replaced periodically (sequences of 15, 30, 30, 90, 90 and 90 days),
- samplings at the end of each sequence for:
  - radiochemical analyses designed to determine the values of the activities released, including 134 and 137 Cs, 52 and 60 Co, and 54 Mn,
  - physiochemical analyses to identify anion/cation exchanges between matrix and leachate, and to secure data to evaluate the durability of the matrix.

CONCLUSION

This operation was conducted over several months due to the high activity of the packages, which forced us to use means and installations designed for radiation work. However, through the use of simple systems, not involving heavy shielding and costly handling systems, we succeeded in gathering a large body of high-grade data.

All the objectives which we set were attained:

- confirmation of the feasibility of a dismantling and coring operation on a high-level package,
- gathering of analytical data (mechanical, chemical and radiochemical) to check the conformity of the package to specifications,
- development of specific methods and inspection procedures.

The results will now be processed to make a judgement on the quality of the package, to check its conformity to specifications, and to make an overall appraisal of the package and its production process.