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Waste package characterisation

Paper presented at the Plenary Meeting
of the European Working Group
"Hot Laboratories and Remote Handling"
at AEA-T - Windscale, UK,
21-23 September 1998

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ABSTRACT

The waste packages emanating from the Hot Laboratory of the SCK•CEN contain a wide variety of radiotoxic materials. As a result there characterisation in terms of amount and nature of the hazardous components is extremely difficult. Ensuring safe disposal however requires an accurate characterisation of both the short- and long-term radiotoxicity.
This paper describes the methodology which has been developed and is adopted at present to characterise the high- and medium-level waste packages at the SCK•CEN Hot Laboratory.
It is based on :

- an estimation of the fuel inventory evacuated in a particular waste package ;
- a calculation of the relative fission product contribution on the fuel fabrication and irradiation footing ;
- a comparison of the calculated, as expected, dose rate and the real measured dose rate of the waste package.

To cope with the daily practice an appropriate fuel inventory estimation route, a user friendly computer programme for fission product and corresponding dose rate calculation, and a simple dose rate measurement method have been developed and introduced.

KEYWORDS

HOT LABORATORY

NUCLEAR FUEL

WASTE

PACKAGE

CHARACTERISATION

1 Introduction

The hot laboratory LHMA (Laboratory for High and Medium Activity) of the SCK•CEN is involved in nuclear fuel research encompassing a wide variety of fuel types. The R&D and fuel qualification programmes encompass a broad range of non-destructive as well as destructive tests producing hardly quantifiable fuel quantities being dispersed in the hot-cell installations. Moreover, parallel programmes are being performed in the same unique hot-cell installations resulting in cross–contaminations. This results in a complex mixture of a wide variety of radiotoxic materials which have to be evacuated as waste :

- different fuel types :
  - PWR-, BWR- and MTR-type fuels ;
  - UO_2, MOX and MTR fuels (e.g. highly enriched UAl_x) ;
  - at different U^{235} enrichments and Pu contents ;
  - at different burnups ;
  - with different cooling times ;
- in different forms of appearance (fuel rod cross-sections, pellets, pellet fragments, dust, ...)
- together with contaminated, spent hot-cell consumables (tissues, polishing clothes, ...).

Hence, the characterisation of the waste packages in terms of amount and nature of the hazardous components is extremely difficult.

On the other hand the Quality Assurance / Quality Control (QA/QC) programme of the Belgian National Institute for Radioactive Waste and Fissile Materials (NIRAS/ONDRAF) requires among others the qualification of the waste packages in terms of the radionuclide inventory. Indeed, the safe final conditioning and disposal requires an as accurate and complete as possible knowledge on the source terms for the performance assessment of the ultimate geological repository. Therefore a methodology based on γ-dose rate measurements complemented by code calculations has been developed and set up. It allows to assess both the short- and long-term radiotoxicity of the waste packages and is compatible with the daily operation practice of the post-irradiation examination hot-cell facilities.

2 Discussion

2.1 Waste packaging

In the hot-cells of the LHMA hot laboratory the waste is collected in tin-plate boxes of 244 mm diameter and 322 mm height (~ 15 l inner volume), which have a wall thickness of 0.3 mm. This is called the primary package.
The fuel material manutated in the hot-cell and its evacuation towards the tin-plate boxes is followed up and registered on a daily basis by the hot-cell operators according to the general fuel accountancy principles. The quantities of fuel material to be assigned to each object is estimated from available information like fuel rod cutting schemes, cutting width losses (e.g. ~ 0.7 mm per cut), weighted fuel fragments, weighted and/or visually observed fuel adhering on cladding samples, ... The hot-cell itself as well as the equipment contained in it are also assigned (minor) quantities of the fuel which has been handled inside, thus allowing to cope with their contamination and the characterisation of the waste produced during their clean up and/or dismantling.
It may be clear that foregoing fuel quantity estimations suffer from a lack of accuracy. However, the main goal is to have a qualitative assessment of the relative contribution of the different fuel types dealt with in the laboratory in the waste packages, i.e. to identify qualitatively and relative semi-quantitatively the different waste constituents.

On evacuation, the primary package is loaded in a polyethylene box, contained in a Pb-shielded wastecontainer and equipped with an α-tight coupling system fitting to the hot-cell access door. This polyethylene box of diameter 270 mm and with a wall thickness of 3 mm, further on called LC-box (from La Calhène system), constitutes the secondary packing. Tin-plate boxes with medium-level waste are preferentially individually loaded in LC-boxes of 395 mm height, and tin-plate boxes with high-level waste per two in LC-boxes of 735 mm height.
A first simple non destructive assay of the global secondary waste package is performed via a dose rate measurement as an appropriate and easily practicable tool. The waste-container, once decoupled from the hot-cell, is opened, and the dose rate is measured with a teledetector both in contact with the cover of the LC-box (measurement 1A) and at 1 m distance of the cover of the LC-box (measurement 1B) - fig. 1a.

Finally the secondary waste package is overloaded in a standard concrete container for evacuation towards the waste treatment plant BELGOPROCESS, the executive subsidiary of the Belgian National Institute for Radioactive Waste and Fissile Materials (NIRAS/ONDRAF). Six medium-level 395 mm LC-boxes are loaded in a 400 l drum contained in a so-called BPIII concrete container (fig. 2), while one high-level waste 735 mm LC-box is loaded in a steel case contained in a so-called type 1100 concrete container (fig. 3).
The overloading is performed by the intermediate of a Pb-shielded exchange-container, such that :

- a second measurement of the γ-dose rate of the secondary package can be performed - a γ–dose ratemeter is positioned in the waste-container/exchange-container coupling system at ~ 73 mm form the side wall of the LC-box - by rotating the LC-box while passing in front of the γ-dose ratemeter an estimation of the waste package homogeneity with regard to the radionuclides contained in it as well as the maximum γ-dose rate is obtained (fig. 1b) ;
- the LC-boxes are positioned upright with their cover on top, thus facilitating their retrieval by coupling on the La Calhène system at the BELGOPROCESS plant.

2.2 Waste package characterisation

Foregoing packaging practice yields essentially two types of information in relation to the waste package characterisation:

1. the fuel inventory, both with regard to the identity of the different fuel types and their relative contribution ;
2. the γ-dose rate measurement results on the global secondary package.

These data are sufficient for the full characterisation of the secondary packages in terms of the qualitative and quantitative inventory of actinides and fission products when complemented by code calculations.

1. Reactor code calculations (Origen - PWR, BWR; Fispin - MTR) allow to obtain a complete relative radionuclide inventory as arising from the fuel inventory and irradiation characteristics. Only in this way the radionuclide inventory can be made complete in a relatively simple manner. Indeed a series of critical nuclides, which have been recognised to be important relative to the long term risks of the waste package ultimate conditioning and disposal, are not easily measurable in the waste package because of their low content, their low specific activity or the particular characteristics of their radiation (e.g. ^{90}Sr, ^{99}Tc, ^{129}I, ^{135}Cs, the U- and Pu-isotopes and the actinides $^{241/243}Am$ and $^{242/244}Cm$).
2. Microshield code calculations allow to obtain the γ-dose rate expected to result from this radionuclide inventory being contained within the secondary package. As the radiation emitted by the radioistopes interact with the material inside the package itself (self-shielding), the calculation has to take into account these interactions and correct for them. This is done on base of three essential variable characteristics of the secondary package :
   - its height (i.e. 395 or 735 mm - related to the type of LC-box used) ;
   - the source configuration, i.e. a homogeneous or heterogeneous distribution of the radionuclides within the package, the latter one being simulated by a concentration of all nuclides in only 1/3^{e} part of the package - this source configuration can be estimated from the dose rate measurements as performed during the overloading into the exchange-container ;
   - its density (i.e. 0.3, 0.5, 1.0 or 1.5 g/cm³) - as estimated from the production route as well as from the weight of the LC-box ;

   in this way resulting in $2 \times 2 \times 4 = 16$ different configurations as being judged to be representative for the secondary waste packages.

   The comparison of the theoretical expected γ-dose rate obtained as such, with the actual measured γ-dose rate, allows to convert the relative radionuclide inventory into an absolute one.

Foregoing methodology is integrated in an user friendly home-made EXCEL based computer module (flow-sheet - fig. 4).

In order to eliminate the need for the user to perform code calculations, two types of basic tables are embedded in the programme :

- basic tables with the inventory of radionuclides in terms of activity per isotope per weight unit of fuel (Ci/Mg HM) ;
- basic tables depicting the corresponding γ-ray radiation yield per unit weight of fuel (γ/s.Mg HM) versus the γ-ray energy.

These tables have been generated by code calculations for different fuel (UO_2, MOX, MTR) and reactor (PWR, BWR, MTR) types and for typical enrichments, burnups and cooling times. Linear interpolation, automatically performed by the module, allows to obtain the isotope specific activities and corresponding γ-ray yield energy-spectrum for the actual fuel enrichment, burnup and cooling time.

Practical application of the module proceeds as follows (fig. 4).

1. Each type of fuel rod handled for PIE in the laboratory, is defined as a component, i.e. its characteristics of fuel and reactor type, enrichment, burnup and cooling time are introduced. With this information, the programme calculates automatically the radionuclide inventory and the corresponding γ-dose rate / fuel mass conversion factor for all 16 different waste package configurations. For this purpose the embedded basic tables, as described above, are used. As a result each component is stored with its characteristics as shown in the example in table 1.
2. For each particular waste package to be evacuated, a mixture of components is defined, i.e. the identification of the different constituent components as well as their relative portion (i.e. their weighing factor - as based on the fuel inventory information supplied by the hot-cell operators) is introduced. Similar to the components, the programme calculates with this information automatically the radionuclide inventory and the corresponding γ-dose rate / fuel mass conversion factor for all 16 different configurations. For this purpose the component tables, as available form the preceding point 1, are used. Again, as a result each mixture is stored with its characteristics as shown in the example in table 2.
3. Finally the waste package itself is defined, i.e. its configuration characteristics and best estimate γ-dose rate measurement result are introduced. On base of these data the programme automatically calculates the activity of fission products and actinides present in the waste package, using the mixture table and the appropriate γ-dose rate / fuel mass conversion factor contained in it. An example of the resulting waste package characterisation is shown in table 3.

As can been seen from table 3, the final radionuclide inventory includes also the calculated as expected amounts of U- and Pu-isotopes, i.e. the residual quantities of fuel material as expected to be present. These on base of the actual γ-dose calculated data for the U- and Pu-isotopes can be compared with the hot-cell operator declared values as a consistency check. It should be mentioned that appreciable deviations can occur (i.e. differences of the order up to 10 dependent on the mixture). This is however not surprisingly in view of to the inevitable rough estimations which have to be made. Indeed, the inherent inaccuracy of the hardly quantifiable dispersed fuel inventory and the simplifying approximations of package configuration needed in the γ-dose rate evaluations, are two inherent main sources of uncertainties among others, which don't allow for a more precise characterisation. Nevertheless, the resulting waste package characterisation is complete regarding the compilation of the radionuclides present and produces a reliable, best achievable quantification of them in terms of order of magnitude.

3 Conclusion

A simple methodology based on γ-dose rate measurements and reactor code calculations has been presented for the radiological characterisation of high and intermediate level wastes originating in the hot cells of the laboratory for post irradiation examination of reactor fuels at SCK•CEN. For the ease of implementation the method has been worked out in the form of a user friendly EXCEL module. This module contains the reactor code data for the different reactor fuels and the dose-rate-to-fuel-mass conversion factors for the different dose-rate measurement configurations. The required inputs to the module are the initial fuel characteristics and the irradiation conditions, and the relative contributions of the different fuels to the waste. The method allows to determine the radioactivity of the critical nuclides, as required by the national agency responsible for the nuclear waste, and requires only a simple dose-rate measurement omitting the need for any specialised waste assay installation.

4 Tables

Component Code	0296C1
Fuel Type	MOX
Reactor Type	BWR
Burnup (GWd / t_{HM})	43
Enrichment [%]	5.1
Cooling time [y]	6
Configuration	**Dose [µSv/h.Mg HM]**
1	4.6E+08
2	3.9E+08
3	2.7E+08
4	1.9E+08
5	2.3E+08
6	1.4E+08
7	3.1E+07
8	5.4E+06
9	6.4E+08
10	5.8E+08
11	4.5E+08
12	3.6E+08
13	4.4E+08
14	3.7E+08
15	2.0E+08
16	8.9E+07
Nuclide	**Ci/Mg HM**
H3	6.0E+02
C14	1.9E-01
S35	3.0E-07
Cl36	1.1E-02
Ca41	1.8E-04
Cr51	1.4E-21
Mn54	1.8E-01
Fe55	3.7E+01
Co58	1.4E-07
Co60	7.0E+02
Ni59	3.2E-01
Ni63	4.3E+01
Se79	2.2E+00
Kr85	4.5E+03
Sr90	3.9E+04
Zr93	1.6E+00
Mo93	3.9E-03
Nb94	1.8E-01
Tc99	4.3E-03
Ru106	1.4E+04
Pd107	3.3E-01
Ag110M	3.2E+01
Sb125	5.1E+03
Sn126	0.0E+00
I125	0.0E+00
I129	5.1E-02
I131	0.0E+00
Cs134	3.8E+04
Cs135	5.7E-01
Cs137	1.2E+05
Ce144	7.2E-03
Sm151	4.9E+02
Eu154	1.4E+04
Ra226	0.0E+00
U233	0.0E+00
U234	6.1E-01
U235	1.2E-04
U236	1.9E-03
U238	3.0E-01
Np237	8.6E-02
Pu238	4.0E+03
Pu239	4.0E+02
Pu240	1.2E+03
Pu241	1.7E+05
Pu242	7.5E+00
Am241	2.3E+03
Cm242	7.6E+03
Am243	1.9E+02
Cm244	8.6E+04
Total	5.1E+05

Table 1 : Example of component table as generated by the waste characterisation module.

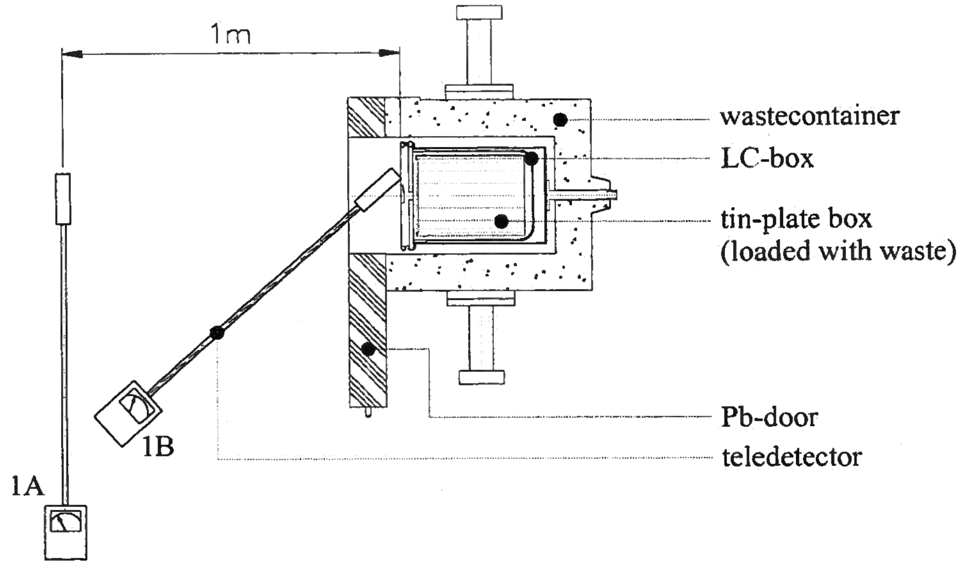
Mixture Code	0296M1
Configuration	**Dose [µSv/h.Mg HM]**
1	2.5E+08
2	2.1E+08
3	1.4E+08
4	1.1E+08
5	1.2E+08
6	7.7E+07
7	1.6E+07
8	2.8E+06
9	3.5E+08
10	3.2E+08
11	2.5E+08
12	1.9E+08
13	2.4E+08
14	2.0E+08
15	1.1E+08
16	4.8E+07
Nuclide	**Ci/Mg HM**
H3	3.8E+02
C14	1.3E-01
S35	9.1E-08
Cl36	8.8E-03
Ca41	1.3E-04
Cr51	4.1E-22
Mn54	5.9E-02
Fe55	1.8E+01
Co58	4.3E-08
Co60	4.3E+02
Ni59	2.5E-01
Ni63	4.2E+01
Se79	2.1E+00
Kr85	2.8E+03
Sr90	2.6E+04
Zr93	1.1E+00
Mo93	2.9E-03
Nb94	1.4E-01
Tc99	4.1E-03
Ru106	4.8E+03
Pd107	2.3E-01
Ag110M	9.8E+00
Sb125	2.5E+03
Sn126	0.0E+00
I125	0.0E+00
I129	3.6E-02
I131	0.0E+00
Cs134	1.5E+04
Cs135	3.7E-01
Cs137	7.7E+04
Ce144	2.3E-03
Sm151	3.9E+02
Eu154	8.3E+03
Ra226	0.0E+00
U233	0.0E+00
U234	8.3E-01
U235	1.4E-04
U236	1.5E-03
U238	3.0E-01
Np237	7.6E-02
Pu238	3.3E+03
Pu239	4.3E+02
Pu240	1.1E+03
Pu241	1.3E+05
Pu242	7.9E+00
Am241	2.4E+03
Cm242	7.5E+03
Am243	2.1E+02
Cm244	6.9E+04
Total	3.5E+05
# Components	3
Total # Units	1898
Component Code	**# Units**
0296C1	577
0296C2	740
0296C3	581

Table 2 : Example of mixture table generated by the waste characterisation module.

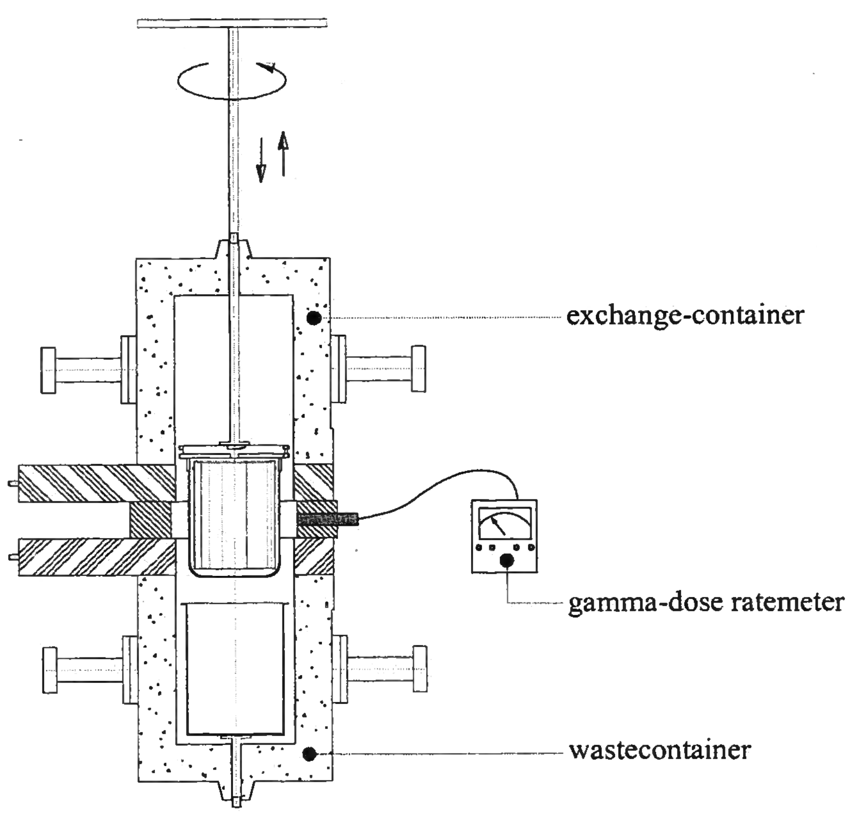
Waste package Configuration	
Height (cm) *Source Configuration :* *Density [g/cm³] :*	74 1/3[e] 1.5
Mixture code: **Actual γ-dose rate [µSv/h] :**	0297M02 1.0E+05
Nuclide	**Ci**
H3	1.3E+01
C14	4.9E-03
S35	1.3E-06
Cl36	2.9E-04
Ca41	4.8E-06
Cr51	7.8E-13
Mn54	2.5E-03
Fe55	5.1E-01
Co58	4.6E-06
Co60	1.3E+01
Ni59	8.3E-03
Ni63	1.2E+00
Se79	6.0E-02
Kr85	9.8E+01
Sr90	9.5E+02
Zr93	4.3E-02
Mo93	1.0E-04
Nb94	4.8E-03
Tc99	1.2E-04
Ru106	1.6E+02
Pd107	8.5E-03
Ag110M	5.3E-01
Sb125	7.2E+01
Sn126	1.2E-12
I125	6.7E-14
I129	1.3E-03
I131	0.0E+00
Cs134	5.0E+02
Cs135	1.5E-02
Cs137	2.8E+03
Ce144	1.3E-04
Sm151	1.3E+01
Eu154	2.9E+02
Ra226	0.0E+00
U233	9.2E-10
U234	1.9E-02
U235	1.8E-05
U236	8.5E-05
U238	8.3E-03
Np237	2.4E-03
Pu238	1.1E+02
Pu239	1.1E+01
Pu240	3.4E+01
Pu241	4.0E+03
Pu242	2.0E-01
Am241	7.9E+01
Cm242	2.1E+02
Am243	5.0E+00
Cm244	2.0E+03
Total	1.1E+04

Table 3 : Example of waste package characterisation as generated by the module.

5 Figures



1a : on evacuation



1b : on overloading

Fig. 1 : γ-dose rate measurements on the secondary waste package

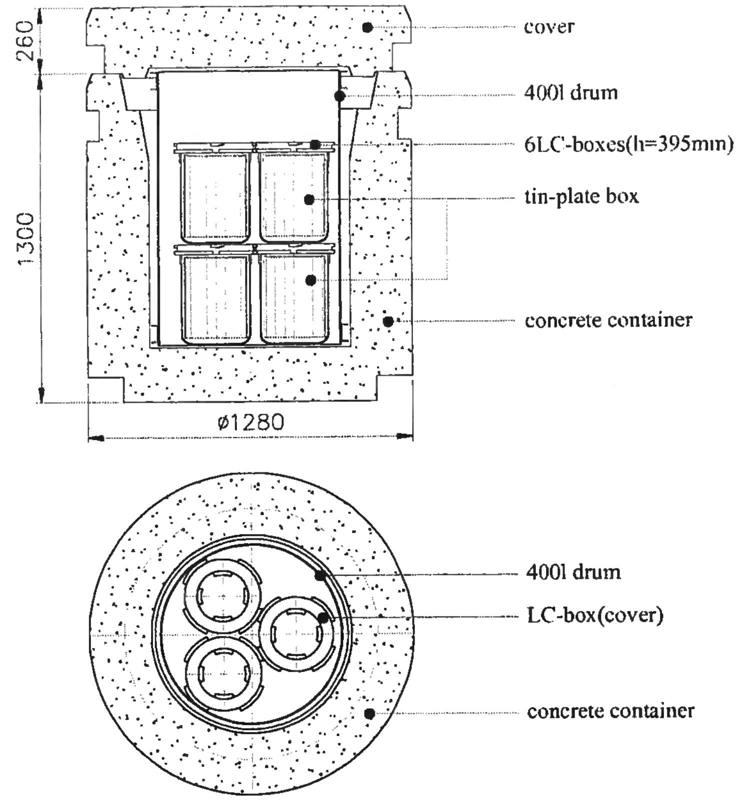


Fig. 2 : medium-level waste packaging in a standard BPIII concrete container

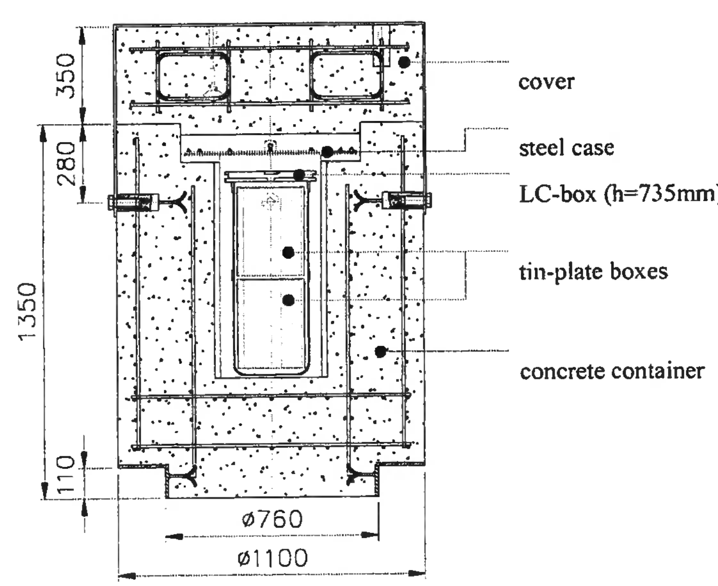


Fig. 3 : high-level waste packaging in a standard Type 1100 concrete container.

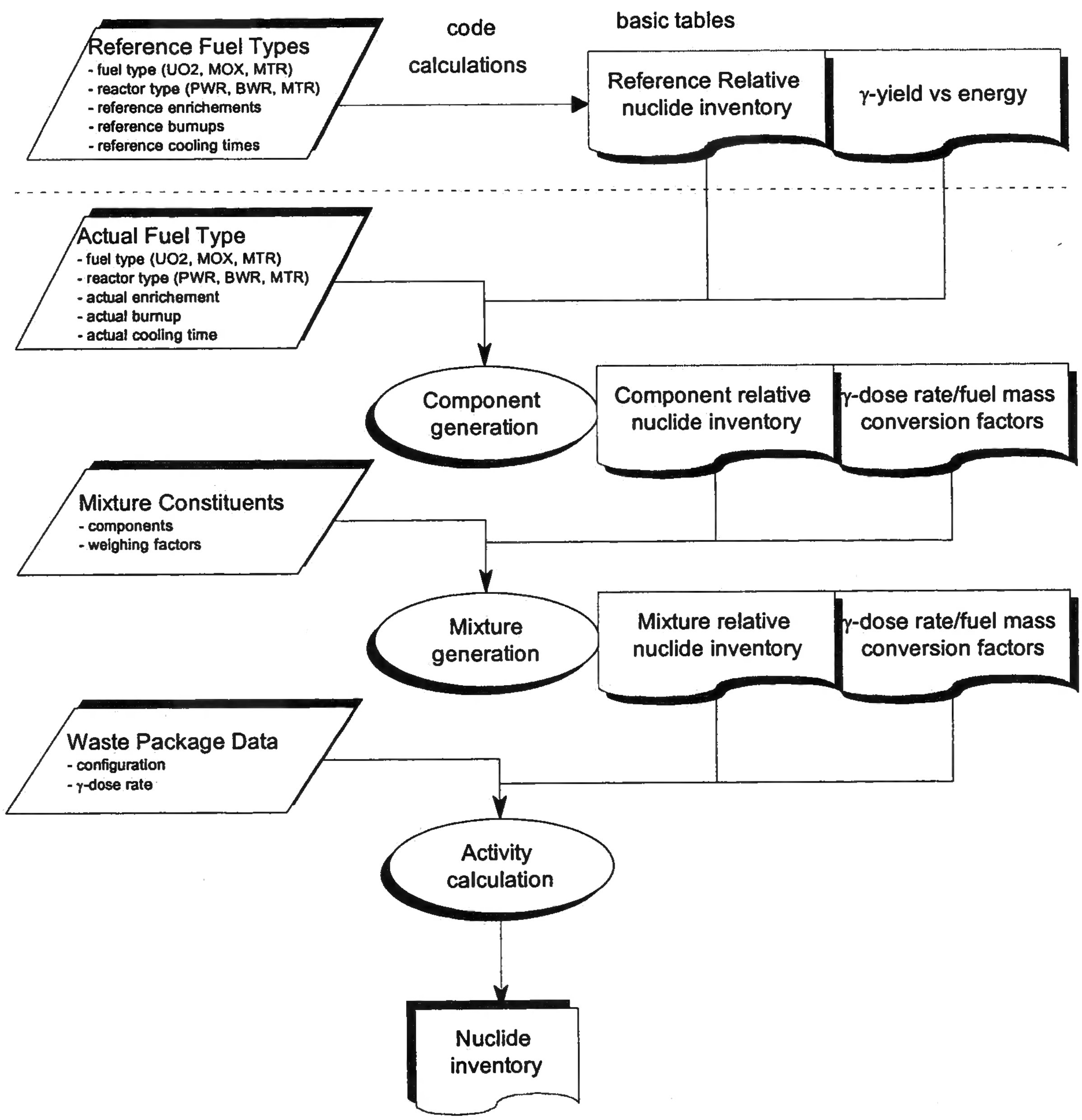


Fig. 4 : Flow-sheet of the waste characterisation module

6 Abbreviations

BELGOPROCESS:	Belgian Waste Treatment Plant (Executive subsidiary of NIRAS/ONDRAF)
BWR:	Boiling Water Reactor
HM:	Heavy Metal
LHMA:	Laboratory for High and Medium level Activity
Mg:	Mega gram
MOX:	Mixed Oxide Fuel
MTR:	Materials Test Reactor
NIRAS/ONDRAF:	Belgian National Institute for Radioactive Waste and Fissile Materials
PIE:	Post-Irradiation Examination
PWR:	Pressurized Water Reactor
RMO:	Reactor Materials Research Department of the SCK•CEN
SCK•CEN:	Belgian National Research Center