SPECIFICATIONS AND QUALITY RELATED ASPECTS IN RADIOACTIVE WASTE CONDITIONING IN BELGIUM.

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ABSTRACT

NIRAS/ONDRAF, the Belgian National Agency for Radioactive Waste and Fissile Materials, has established, in its waste management policy, quality assurance and quality control programmes. In the case of waste conditioning they must provide the necessary confidence that the product quality achieved during conditioning complies with the requirements defined by the acceptance criteria in force. The information on product quality they provide arises from : 1) The qualification of the conditioning process, an inspection and control programme implemented during the application of the process to a well-defined waste stream, the evaluation of the quality assurance programme applied by the conditioner. 2) The verification of the product characteristics specified by the conditioner using destructive and non-destructive measuring techniques on samples from the product development- and industrial demonstration phases, and from the actual application of the process on radioactive wastes.

To provide the necessary data for the safety assessments for final disposal concepts for conditioned wastes and to enable the quantification of their radioisotope inventories NIRAS/ONDRAF has, for wastes arising from nuclear power stations, initiated a programme to determine the concentration of longer-lived radioisotopes in the different waste streams. Its aim is to evaluate possible correlations between difficult to measure isotopes and some key-isotopes which can readily be determined using simple measuring techniques.

INTRODUCTION

As the responsible for the management of radioactive wastes in Belgium, NIRAS/ONDRAF has set up a comprehensive quality assurance and quality control programme for conditioned radioactive wastes.

These programs are based on waste acceptance criteria for each waste category and determine the necessary control measures to make sure that waste packages comply with the criteria.

The control measures are presented in the form of general conditions for acceptance to the waste conditioners.

The first part of the paper defines these general conditions and informs on their practical implementation. The second part describes the contribution of product characterization in the control of product quality, with emphasis on characterization programmes for high- and medium level wastes already being implemented at the moment.

Since one of the most important properties of a waste package concerns its radionuclide inventory, the way to determine it is a necessary part of a waste management program.

The third part of the paper deals in particular with the programmes followed in Belgium for the determination of disposal critical nuclides in PWR Waste streams.

WASTE ACCEPTANCE CONDITIONS AND THEIR IMPLEMENTATION

The general conditions defined by ONDRAF/NIRAS for the acceptance of conditioned waste are as follows :

- the conditioning process and installations must have been qualified by ONDRAF/NIRAS ;
- an inspection and control programme is to be defined and implemented for each conditioning process with various points for inspection by ONDRAF/NIRAS ;
- the quality assurance programme of the conditioner will be submitted to ONDRAF/NIRAS for approval and its application controlled by regular audits.

Each of these three conditions and their implementation are described in detail below.

Qualification Of The Treatment/Conditioning Processes And Installations By ONDRAF/NIRAS

For the qualification by ONDRAF, the producer must draw up a technical qualification file in order to demonstrate that the packages produced meet the acceptance criteria in force.

These acceptance criteria specify, for each waste category and type, the various properties to be met by the conditioned waste packages in order to satisfy the safety conditions and criteria associated to the different stages of their management : transportation, interim storage and disposal. These criteria refer to the waste form, the packaging used and the waste package itself.

The acceptance criteria for the various types of low- and intermediate- level waste of category A are currently being finalized.

Category A waste is waste containing mainly radionuclides with a half-life below 30 years and in which the

concentrations of radionuclides with a longer half-life are below the limits imposed by the shallow land disposal concept safety analysis. This disposal option with unrestricted release after 300 years is the reference concept considered for specifying the acceptance criteria.

Each qualification file must be prepared according to the principles imposed by ONDRAF/NIRAS.

Each file will include two parts :

- a first part related to the qualification of the process and to the characterization of the end product ;
- a second part related to the qualification of the installation.

In order to have his process qualified, the waste conditioner checks by means of preliminary tests, whether the process is adequate to deliver an end product compatible with the acceptance criteria. This process development stage takes into account the results of parametric tests concerning the influence of the product composition on its properties.The product is characterized on the basis of a comprehensive characterization programme to show compliance of the product with the waste acceptance criteria.

The part of the file related to the installation must give a complete description of the installation, including a descriptive note, process and engineering flow sheets, operating and control procedures.

The general rules required for qualification files are adapted when treatment/conditioning installations have already produced waste packages for a number of years. In such case, the process development stage is not applicable to these installations and the product characterization directly concerns the end product produced in the active stage.

The qualification files related to the conditioning installations for category A waste, i.e. the installations of the nuclear power stations of Doel and Tihange and certain installations on the BELGOPROCESS site, have been prepared on the basis of the existing working conditions and taking into account the current acceptance criteria, namely the sea disposal criteria. In the same way, ONDRAF/NIRAS has started working out characterization programs according to the new acceptance criteria taking into account shallow land burial as the reference option for this waste category.

The categories B (medium level waste) and C (high level waste) waste characterization programmes currently in progress are discussed in detail in the second part of this paper.

The detailed validation of each file by ONDRAF/NIRAS and the satisfactory execution of the characterization programs are the bases for ONDRAF/NIRAS to deliver a definitive agreement for each installation.

This agreement remains valid insofar :

- all properties of the waste to be conditioned remain within the limits considered of the approval ;
- the critical parameters (as described in the file) of the process and the installation are not modified ;
- no frequent non-conformities versus the acceptance criteria arise during the process execution.

Inspection and Control Program

The second general condition imposed by ONDRAF/NIRAS for taking over the conditioned waste packages is related to the follow-up by ONDRAF/NIRAS of the waste production. This follow-up covers all stages from production to taking over of the packages by ONDRAF/NIRAS : supply of raw materials, possible pretreatment, actual conditioning and interim storage on the conditioner's site.

The various controls and inspections aim at :

- verifying that the conditioner observes the qualification file, especially that he remains within the limits of the qualification file ;
- verifying that the applicable specifications and procedures are followed correctly to ensure the final quality of the product ;
- ensuring the traceability of the waste during the whole process.

For each installation, the follow-up of the conditioned waste production will be done on the basis of a document describing chronologically each fabrication and control stage. This document is called LOFC (List of fabrication and controls).

Each fabrication or control step of the LOFC refers either to the operating or control procedure applied, or to the corresponding test or control sheet.

Each process will be the object of a specific LOFC defined by common consent between ONDRAF/NIRAS and the conditioner.

Each item of the LOFC will be countersigned during production by the conditioner's operator. The QA responsible of the conditioner and the inspector of ONDRAF/NIRAS will indicate their intervention points. In principle, the intervention points of ONDRAF/NIRAS are "whitness" points, except for the final reception on the site which is a "hold" point.

The final reception on the conditioner site of packages consists on the one hand in verifying the physical and radiological characteristics of each package as measured by the

conditioner and on the other hand of a comprehensive study of the package production file. The production control system implemented by means of the LOFC forms also allows to identify any non-conformity with regard to the applicable specifications.

The diagram in Fig. 1 shows the structure of the LOFC to be followed for the evaporator concentrates cementation process at the nuclear power station of Tihange.

The following five LOFC are used in the system :

LOFC 1 is related to the controls of the empty drums.

LOFC 2 is related to the quality control of the cement used as matrix. In order to ensure that the cement meets the specifications, a product sample is taken before each conditioning campaign in order to prepare a test sample and to test its compression strength.

LOFC 3 defines the quality control of the water used for the constituent cement of the coating matrix.

LOFC 4 defines the qualification of the cementation formula. It includes pretreatment of the solutions, sampling of the concentrate tank, determination of the cementation formula, preparation of test samples with different product

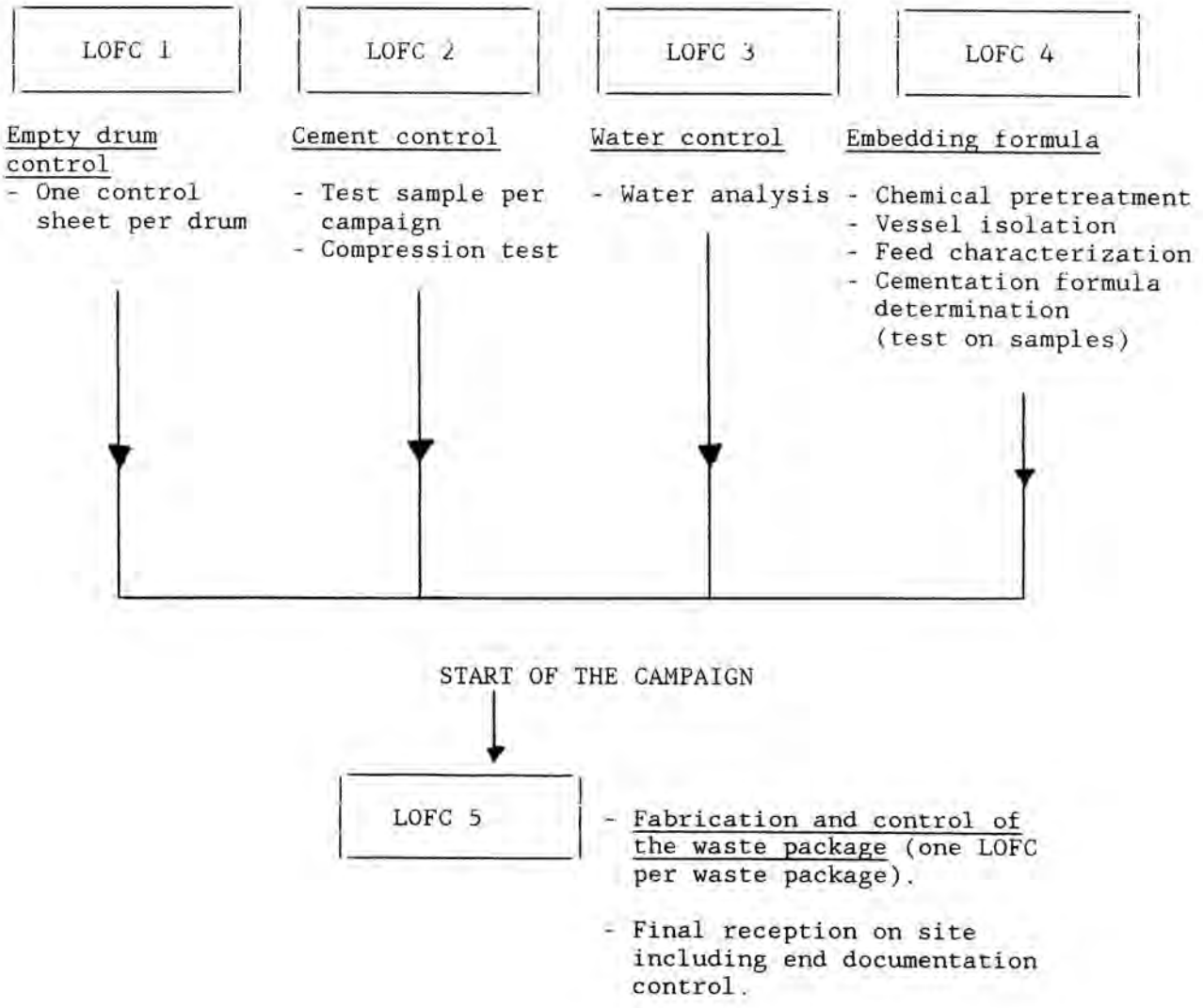

Fig. 1. LOFC Structure for Cementation Process.

compositions and testing the resulting compression strengths.

When the LOFC forms 1 to 4 have been validated, the cementation campaign can start. This campaign is the subject of LOFC 5 which is specific to each waste package.

Quality Assurance Program

The third general condition required by ONDRAF relates to the conditioners quality assurance program.

Each conditioner must work out and follow a quality assurance program covering all activities and installations associated to supply, treatment/conditioning and interim storage before expedition. The programme will be submitted to ONDRAF/NIRAS for approval and will be in accordance with well recognized code, such as the IAEA 50 CQA code.

ONDRAF/NIRAS will verify the follow-up and the application of the quality assurance programme of the conditioner by means of audits which will be organized periodically. These audits will be performed independently of the production inspections and controls defined in the paragraph above.

HIGH- AND MEDIUM-LEVEL WASTE CHARACTERIZATION

General

The first step in the verification of the quality of waste packages achieved during conditioning processes is based on the implementation of the quality control and quality assurance programme defined in the previous chapter.

Additionally, a programme has been elaborated and is being implemented which aims at the verification of the product characteristics as specified by the producer. Investigated are material properties such as composition, homogeneity, microstructure, physico-chemical properties, chemical stability, etc....

The experiments are performed on inactive or slightly active simulants and on fully active materials in the laboratories of the Nuclear Research Center at Mol (CEN/SCK).

The degree of detail and depth with which the different waste forms are studied depends to some extend on their type and origin. Investigated are:

- Waste forms resulting from the conditioning of wastes generated during the reprocessing abroad of spent fuel from Belgian power plants (Cogema - La Hague).
- Waste forms generated in the former Eurochemic reprocessing plant which has terminated its active phase and is managed by Belgoprocess, since recently a branch of NIRAS/ONDRAF. On its site all wastes resulting from the industrial and medical use of radioisotopes in Belgium are conditioned, in addition to the wastes from earlier reprocessing activities.

Waste Form Characterization

Investigated vitreous waste forms include the borosilicate glasses produced at both the R7/T7-plant at La Hague and the PAMELA-plant at Mol. For the latter 3 glass compositions (SM 513 LEWC, LEWC = Low Enriched Waste Concentrate, SM 527 HEWC and SM 539 HEWC, (HEWC = High Enriched Waste Concentrate) are studied, in addition to the vitromet (glass beads in a lead matrix). For the Cogema-glass only one composition has been specified.

For the PAMELA-glasses the characterization work is made on samples (MT) from the three project-phases: product development (MT1), industrial demonstration on a 1/1 scale with inactive simulants (MT2) and the actual conditioning of the wastes (MT3).

The characterization program covers:

- the chemical composition (MT1, 2 and 3);
- the concentration and distribution of radionuclides (MT3);
- the homogeneity of individual specimen, the homogeneity within a single canister and between canisters (MT2 and MT3);
- the thermal characteristics and the behavior towards crystallization (MT1);
- the determination of the leaching rate under standard conditions (MCC1, MCC5), (MT1);
- phase separation phenomena.

The very first results obtained show:

- discrepancies between expected and found concentrations for some elements for the technical (MT2) glass SM 527, still to be confirmed by further analyses;
- an apparent homogeneous distribution for Si, Al and Ca, and to a lesser extend for Na, in individual specimen taken at different heights in 2 full size containers with the inactive glass;
- a homogeneous distribution for Si, Al and Ca within one of the containers and higher variations in the second container, with an inverse observation for Na;
- Ru-Rh-segretations in the active LEWC-glass SM 513 with also spots enriched in Fe-Cr-Ni;
- a homogeneous distribution for Si and Al in the active glasses SM513 and SM527, while Na and Ca show less homogeneity.

For the Cogema-glass only results for the MT1 sample

are available and they generally confirm the data given by the producer for chemical composition, thermal behavior and leaching. Although they have been requested no MT2-samples are yet available. Sampling of the active material is not foreseen at the R7/T7 plant and as a substitute a 400 g-glass sample prepared at CEA-Marcoule following the Cogema specifications is used for characterization purposes. The analyses include the determination and distribution of the radionuclides present, the chemical homogeneity and the leaching behavior of the radionuclides under standard conditions. The necessary subsamples have recently been cut.

For the bitumenproducts, resulting from the conditioning of EUROCHEMIC MLW-effluents, the program covers:

- the chemical composition of the embedded salt fraction (MT2, MT3);
- the determination of changes in density, penetration, softening-, flash- and selfignition point in function of time (MT3);
- the determination of leaching rates under standard conditions (MT2);
- the leaching behavior of the radionuclides present (MT3);
- the swelling behavior when contacted with water and clay/water mixtures (MT2, MT3);
- the influence of pressure (repository pressure, 4 MPa) on swelling (MT3).

The first results for ageing reveal some changes in physical properties after four years (Table I), [1] :

TABLE I
Physical Properties Of Eurobitum
As A Function Of Ageing
(each result is the average value for four samples)

	TIME OF ANALYSIS		
	AS PRODUCED	AFTER 1 YEAR	AFTER 4 YEARS
Self-ignition point (°C)	312	313	293
Flash point (°C)	262	261	272
Softening point (°C)	101	101	104
Penetration (10^{-1} mm)	20.0	19.3	18.8
Density ($g.cm^{-3}$)	1.32	1.32	1.36

- The softening point increases (from 101 to 104°C), the penetration decreases (from 20.0 to 18.8 x 10^{-1} mm), and the density increases (from 1.32 to 1.36 $g.cm^{-3}$). This suggests a hardening of the bituminizates together with a shrinking, probably as a result of different phenomena such as: (1) a change in the structure of the bitumen matrix due to radiolytic H_2 formation; (2) continued shrinking during longterm storage; (3) escape of volatile bitumen components.
- The flash point increases from 262 to 272°C, and the self-ignition point decreases from 312 to 293°C. Both phenomena might be related with the creation of radiolytic gases, altering the combustion process. In case of the self-ignition point, also changes in the chemistry of the solid material might contribute to differences in self-ignition behavior.

The determinations will be repeated after 6.5 and 10 years.

As a result of the small sample sizes (0,7 ml) used in the first active experiments, high degrees of swelling were observed (up to 900%) [1]. The equivalent leached thicknesses for $NaHO_3$ and $CaCO_4$ are comparable to those for (larger) simulated specimen. As much as 15% of the beta-emitter inventory (mainly ^{90}Sr, ^{137}Cs and ^{60}Co) was leached after one year corrosion. Of the three nuclides ^{137}Cs was best retained. The fractional release of the alpha inventory was limited to 0.1 % after one year. To overcome the disadvantages of the use of small samples, the only available at the time, the programme foresees to repeat these experiments on larger active samples.

Of the Cogema bitumen products only inactive simulates are available and tests are still in progress at the time. Also here work on active samples is included in the programme.

For cemented Cogema waste forms the characterization programme covers technological wastes and hulls and end caps.

For the first the long term behavior of the used cement container will be examined by corrosion tests in water. In addition the diffusitivity of a number of radionuclides is determined.

For the cemented hulls and end caps, special attention will be given to the interaction between the hulls and the cement matrix by:

- microscopic (optical, SEM) examination of the interface hull/cement;
- investigation by SEM and autoradiography of the

eventual absorption of radionuclides and corrosion products in the matrix material.

THE DETERMINATION OF DISPOSAL CRITICAL NUCLIDES IN PWR WASTE STREAMS

Safety studies related to the shallow land disposal of low- and intermediate level wastes indicate that the long term risk is determined by the presence of longer-lived nuclides such as ^{14}C, ^{59}Ni, ^{63}Ni, ^{90}Sr, ^{94}Nb, ^{99}Tc, ^{129}I, ^{135}Cs, ^{137}Cs and the transuranium elements.

In order to improve its knowledge of radioactive inventories in PWR waste streams, the only reactortype in use in Belgium, NIRAS/ONDRAF started a sampling and analysis programme which is sponsored by the Commission of the European Communities. Three waste types are investigated: evaporator concentrates, ion-exchange resins from the purification chain of the primary loop and filters from the main auxiliary systems.

The program aims at:

- the development of sampling procedures, sample preparation and analysis techniques necessary to establish the radionuclide inventories of these waste streams;
- an investigation of the representativity of these samples, with an evaluation of the possible parameters liable to affect their composition;
- the establishing of a practical method for routine use to determine the concentrations of the given radionuclides e.g. by correlation with the concentration of easily detectible key-nuclides.

In a first phase samples were collected at the two reactor sites, Doel and Tihange. They are now being analyzed at the Nuclear Research Centre at Mol, using sampling - and analytical techniques specifically developed for the three waste streams. The programme also includes a destructive verification of radionuclide inventories in the cemented evaporator concentrates and ion-exchange resins.

In parallel NIRAS/ONDRAF has initiated a study to make a theoretical evaluation of the concentrations of the given radionuclides in the three waste streams based on:

- the calculation of their activities in the primary loop, assuming a percentage of leaking combustibles corresponding to the observed average in the Belgian PWR's;
- the distribution of the radionuclides in the different waste streams considering the physico-chemical forms in which they are present;
- the exploitation procedures for the treatment and conditioning of effluents and wastes which are applied.

Both programmes are scheduled to be finished mid-1990. The results available today, however, are not yet conclusive.

REFERENCES.

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