

The German Product Quality Control of the Compacted Metallic Nuclear Waste – 15311

Yuliya Aksyutina *, Ivan Fast *, Holger Tietze-Jaensch *, François Guillaumin **, Philippe Pinson **

* Forschungszentrum Jülich GmbH

** AREVA NC

ABSTRACT

The treatment and safe disposal of spent nuclear fuel is a key challenge of nuclear energy utilization and has to be accomplished regardless the future nuclear of electricity production or phase-out policies. Until June 2005 approx. 6662 tons of spent fuel from German nuclear power plants was sent to the La Hague (France) and Sellafield (UK) plants for reprocessing. An equivalent amount of nuclear waste compounds has contractually to be returned to Germany, which is currently in progress. The returnable waste streams comprise mainly highly-active vitrified waste containing a major part of fission products and actinides, and intermediate-active super-compacted metallic waste containing reprocessed fuel element's structural materials and some technological waste from the reprocessing process itself.

Shearing and dissolution of spent fuel leaves scraps of fuel metal cladding, the end-pieces and spacers which are contaminated, activated and irradiating from their life in a reactor core. In fact, this metallic intermediate level radioactive waste makes up a bit more than half of the total volume of the reprocessed waste. At La Hague the metallic waste is super-compacted. Several, typically eight, compacted discs are stacked in a stainless steel container, named CSD-C as for "Colis Standard de Déchets Compactés". 4104 containers of such CSD-Cs in 152 transport/storages casks have to be returned to Germany. Each one of them has to be checked and quality certified for its conformity with German national product specifications and acceptance criteria by independent national product quality control inspectors (PKS). The residue package documentation has to be complete and the characterization methods have to be comprehensive.

Due to the heterogeneity of metallic waste the characterization and control of its properties is very complex. The nuclear characterization is performed by means of non-destructive measurements, where only a few key nuclides are measured directly. In fact, the proper calculation of the whole nuclear inventory relies on numerous supplementary simulations. The measurement and calculation methodology is scrutinized by PKS in detail and key points are benchmarked with our own simulations for the sake of an independent nuclear inventory declaration check.

With respect to the vast amount of documentation and parameters, which have to be checked and verified, PKS had to optimize their checking procedures. For this purpose dedicated proof tools have been developed that shall be applied for selected indicative cross-checks of the relevant parameters. This contribution is dedicated to the procedure of quality control by PKS and examples of actually performed computational verifications of declared nuclear inventory.

INTRODUCTION

The treatment and safe disposal of spent nuclear fuel is a key challenge of nuclear energy utilization and has to be accomplished regardless future nuclear electricity production or phase-out policies. In Germany reprocessing of spent nuclear fuel was compulsory until 1995, and then it became voluntary. After June 2005, transport to reprocessing has been forbidden by law, for spent fuel originating from power plants and since then all spent fuel has been stored at nuclear power plant sites. Spent fuel from German Nuclear power plants was sent to La Hague (France) and Sellafield (UK) for reprocessing until 2005. An equivalent amount of nuclear waste compounds has contractually and/or legally to be returned to Germany, which is currently in progress. These returnable waste streams are mainly of two types: highly-active vitrified waste

containing a major part of fission products and actinides; medium-active vitrified waste containing liquid solutions of tank and vessel rinsing and medium-active compacted metallic waste containing mainly structural material of reprocessed fuel elements. Waste of all three types has to be returned from France, while the UK will return highly-active vitrified waste, only. Each waste package has to be controlled and certified for its conformity with German national product specifications and acceptance criteria by independent national product quality control inspectors. The residue package documentation has to be complete and the characterization methods have to be comprehensive.

A large volume of compacted metallic waste has to be returned to Germany in forthcoming years. Due to their heterogeneity the characterization and control of such waste package properties is much more complex, than of vitrified waste. This talk is about the methods deployed by the German product quality control office (PKS) for the checking and verification of the compacted metallic waste package properties.

MAIN PRODUCT QUALITY CONTROL FOR COMPACTED METALLIC ILW

Shearing and dissolution of spent fuel leaves scrap metal of fuel cladding, the end-pieces and spacers that are contaminated, irradiated and activated from their life in a reactor core. In fact, this metallic intermediate level radioactive waste makes more than a half volume of the reprocessing of spent fuel. The German share has to be returned from France, while in the UK it was agreed to be compensated with an equivalent extra portion of vitrified waste.

At La Hague the metallic waste is compacted. Several, typically eight, compacted discs are stacked in a stainless steel container. The product name is CSD-C, an acronym for “Colis Standard de Déchets Compactés”. The task of the Product Quality Control is to check the documentation and verify product conformity with the regulations of each of the CSD-Cs, to be returned to Germany. Four major pre-requisites have to be fulfilled before waste residues are approved for return:

1. Product specifications approval: a government approval is required for the anticipated or proposed specifications, i.e. a definition of the radioactive waste product, its physical and chemical properties as well as quality traceable identifications. These product specifications must comply with a number of repository relevant acceptance criteria (Table I).
2. Production process approval. Conditioning manufacturers have to describe their production process and provide the evidence that this process is appropriate and delivers waste compounds of the above mentioned specifications. This production process is subject of an official approval by the German authorities.
3. Process performance surveillance: once a process has been approved, all technical upgrades and modifications or deviations are subject for approval as well. All production standards must be maintained within the limits of production process clearance and approval. Therefore, the production process is constantly controlled by independent on-site inspectors. Their reports are received by PKS and studied. In addition, PKS performs six-monthly plant inspections and audits of the independent on-site inspectors.
4. Documentation check: when individual CSD-Cs are selected for the return their accompanying documentation, comprising the inventory declaration as well as physical properties, has to be checked by PKS. The German inspectors perform a 100% documentation check with independent data consistency calculations and properties derivations.

TABLE I. Repository Relevant Properties & Characteristic Values for Compacted Metal-Waste

No.	Property	Experimental Assessment (<i>italic</i>) / R&D definition (regular letters)
1	Total activity α and β	<i>Gamma-spectrometry, active and passive neutron interrogation, correlation calculation</i>
2	Activities of relevant nuclides	
3	Criticality safety	
4	Thermal properties	<i>Calculation (derived from neutron & gamma measurements)</i>
5	Dose rate (n, γ at surface & 1m distance)	
6	Surface contamination	<i>Wipe test</i>
7	Raw waste properties	Waste process management, identification of specific components
8	Compact-disk properties	Process design & management, cartridge layout & <i>height measurement</i>
9	Waste package properties	R&D characterization, process management, <i>visual inspection</i>
10	Hydrolytic stability of waste form	R&D characterization
11	(empty) container mechanical properties	Manufacturers design & QA, welding & corrosion QA
12	Weight	<i>Weighing</i>
13	Stackability & handling	Container design
14	Residue package identification	<i>Visual control</i>

The principles of Quality control of compacted waste residues are presented in Figure 1. In fact, production undergoes two distinct qualified processes (QP) that are detached from one another. The process QP1 comprises compaction process itself and the preparation of a CSD-C with certain predefined properties within the qualified domain. The inventory determination is performed in a subsequent but detached process QP2, the nuclear measurement station (Figure 1). Since the varying material composition of CSD-Cs affects the nuclear measurements, traceability is an important issue of QP1. The dosing of hulls, end-pieces and technological waste is performed under video-surveillance. The amount and type of end-pieces are noted.

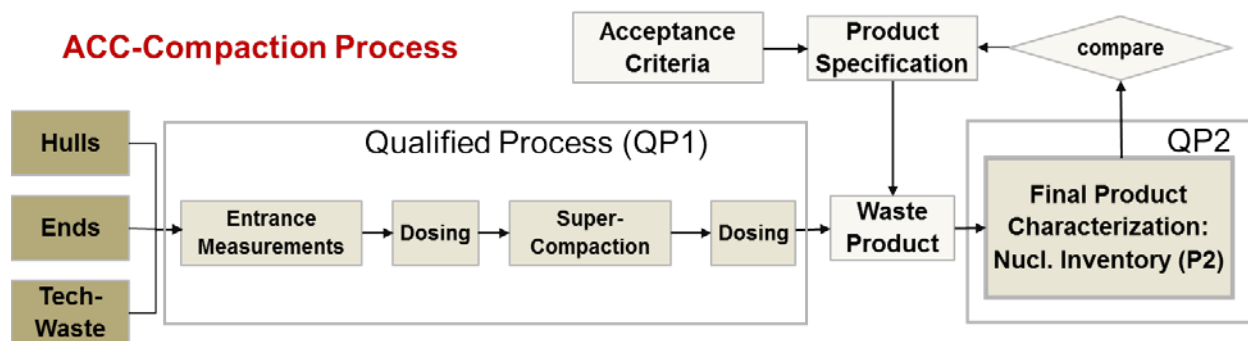


Fig. 1. Principle flow chart and quality objective of the metallic waste compaction processes

The characterization is performed by means of non-destructive nuclear measurements without taking credit from any biased information of the inbound waste streams, such as individual fuel type, burn-up or cooling time. Only a handful of key-nuclides can be measured and the complete nuclear inventory declaration relies substantially on simulations and is based on numerous correlations [1, 2]. The used experimental techniques and measurable nuclides are summarized in Table II.

TABLE II. Gamma and neutron measurable key nuclides for CSD-C; bold: group representatives

Measurement	Detected Radio Nuclide
Gamma Spectrometry	Co-60, Sb-125, Cs-134, Cs-137, Eu-154, Rh-106
Passive Neutron Counting	Pu-238 + Pu-239 + Pu-240 + Pu-241 + Am-241 + Cm-242, + Cm-244 + Cm-246
Active Neutron Interrogation (Prompt Neutron Counting)	U-235 + Pu-239 + Pu-241
Active Neutron Interrogation (Delayed Neutron Counting)	U-235 + U-238 + Pu-239 + Pu-241

Radio-nuclides, such as Cs-134, Cs-137, Eu-154, Co-60 and Sb-125 are measured for their gamma emission and key actinides are determined from active (U, Pu) or passive neutron counting (Cm-244). Cooling time and burn-up, derived from the activities ratios of Eu-154/Cs-134 and Eu-154/Cs-137, respectively, are the key parameters for the subsequent nuclear inventory calculation. The combination of prompt and delayed neutron counting, resulting from the active neutron interrogation, merged with the additional information derived from Eu-154 and Cs-137 activities, allows for the unambiguous determination of the U-235, U-238, Pu-239 and Pu-241 masses.

Due to vast complexity of nuclear inventory determination the PKS verification procedure has to be revised. The usual method of 100% independent calculation of all declared parameters has to be abandoned. Instead, the measurement and calculation methodology is studied by PKS in detail and several key points are benchmarked with our own simulations. In order to make documentation check a feasible task, despite the variety of parameters used for the inventory calculation, only limited amount of declared values are independently calculated by PKS. Examples of actually performed verifications will be presented during this talk.

THE DEVELOPMENT OF THE PROOFTOOL FOR DOCUMENTATION CHECKS

In the determination of nuclear inventory the so-called characteristic burn-up and cooling time play crucial role. The compacted waste residues may contain waste from several fuel elements with different irradiation history, e.g. different burn-up and cooling time. The idea is to determine some kind of mean values, which could be used to characterize the residue as whole, without using the information on its origin. The Figure 2 shows the results of our calculations for two broadly used correlations for burn-up determination, using activities Cs-134, Cs-137 and Eu-154. The calculations were performed for several values of initial enrichment, while it is usually unknown and defines the largest part of uncertainty in burn-up. We compared our calculations to experimental values from SFCOMPO online database [3]. This database contains selection of data with different characteristics and different irradiation histories, and is therefore perfectly suitable for benchmarking of our calculations. Since the measurement results are within our correlation bandwidth, we can conclude that our calculations cover all typical configurations and may be used also for checks on compacted residues.

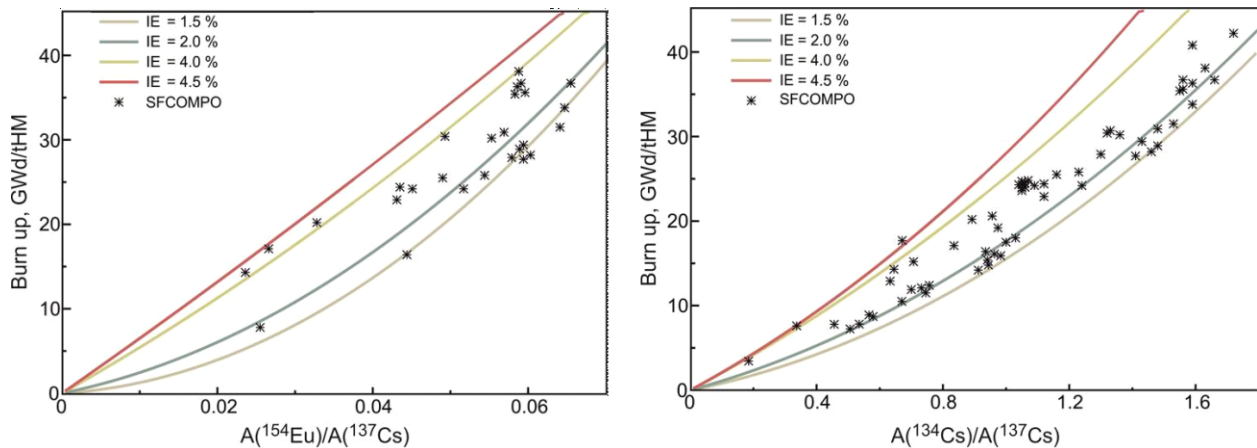


Fig. 2. The correlations for determinations of burn-up using the activities of Cs-134, Cs-137 and Eu-154.

Another interesting example is the calculation of the neutron and gamma dose rates. These parameters usually have to be documented for every waste residue and verified by independent inspector.

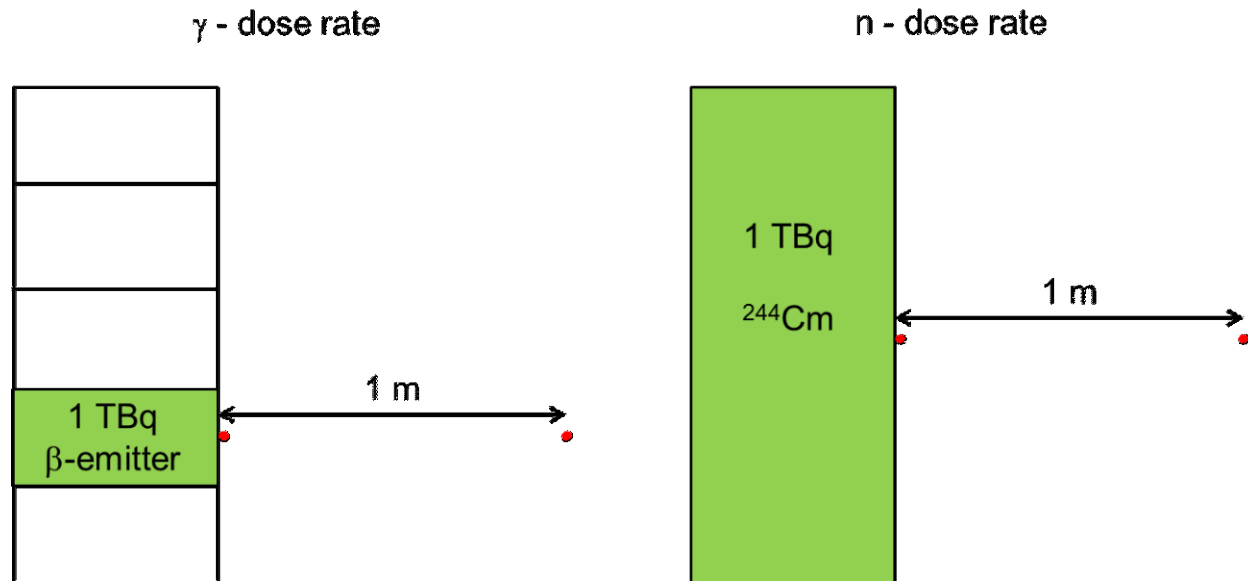


Fig. 3. The geometries used for the calculation of the dose rate factors.

For heterogeneous compacted waste residues, the usual approach of dose rate measurement at one point is not representative, and more detailed measurements at different points are very time-consuming. Therefore, a special theoretical algorithm was developed by AREVA NC, which allows determining the conservative dose rate values, based on measurement results. PKS studied this method in details and, additionally, performed simulations in order to verify independently the dose rate coefficients. The geometries are schematically shown in Figure 3. The green areas represent the volume, where activities of relevant nuclides are distributed and red points correspond to the relevant point for dose rate calculations. Since for gamma measurements the residue is split into five zones [1, 2], the same principle is used for dose rate calculations.

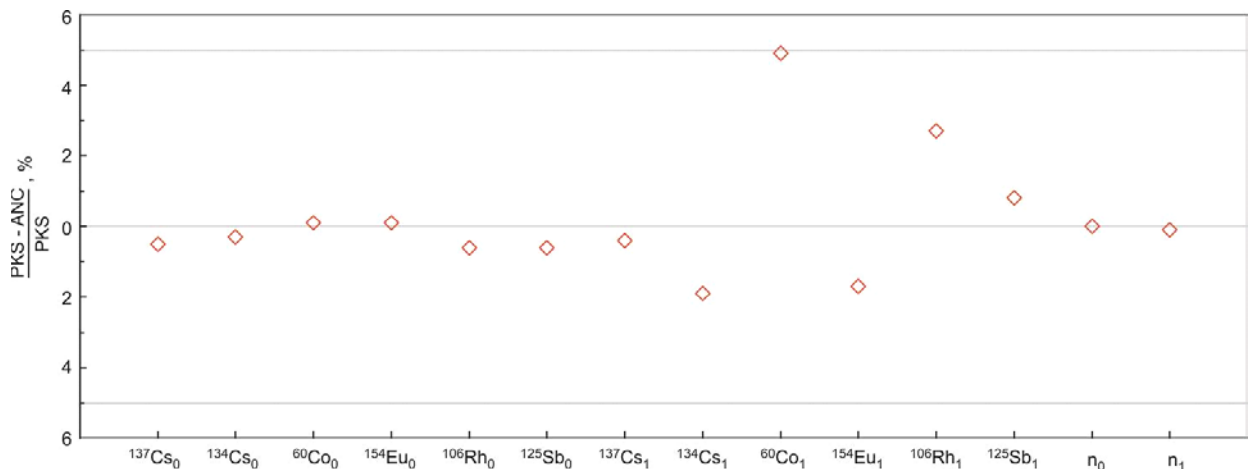


Fig. 4. The comparison between the gamma and neutron dose rate factors, performed for key nuclides at the surface and at 1 meter distance.

The results of comparison are shown in Figure 4. Here the calculations of PKS for contribution of the measurable gamma and neutron emitters to the dose rate at the surface and at 1 m distance were compared to corresponding values from AREVA NC. The observed deviations are due to some differences in geometry and material, used for simulations.

SUMMARY

The principles of the German product quality control of radioactive waste residues from the spent fuel reprocessing are introduced and explained. The challenges of controlling heat generating, highly heterogeneous compacted metallic ILW are discussed and as well as proposed solutions are demonstrated. Several examples of the verification calculations are presented.

REFERENCES

- [1] H. Toubon, T. Gain, B. Perot, A. Lyoussi, A.-C. Raoux, G. Melhman, X. de Brimont, P.A. Tulle, Method for ascertaining the characteristics of the radiological content of canister of compacted hull and nozzle waste resulting from reprocessing at the La Hague of irradiated fuel assemblies from light water reactors, Proc. Safewaste 2000, Montpellier
- [2] H. Toubon, Vuillier, T. Gain, M. Huver, Calibration and hot testing of the advanced nuclear measurement systems used for waste characterization in COGEMA's new ACC compaction facility, 2003, Proceedings of Waste Management 2003, Tuscon, AZ.
- [3] <http://www.oecd-nea.org/sfcompo/Ver.2/Eng/>