Principles of Product Quality Control of German Radioactive Waste Forms from the Reprocessing of Spent Fuel: Vitrification, Compaction & Numerical Simulation – 12529

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ABSTRACT

The German product quality control is inter alia responsible for control of two radioactive waste forms of heat generating waste: a) homogeneous vitrified HLW and b) heterogeneous compacted hulls, end-pieces and technological metallic waste. In either case, significantly different metrology is employed at the site of the conditioning plant for the obligatory nuclide inventory declaration. To facilitate an independent evaluation and checking of the accompanying documentation numerical simulations are carried out. The physical and chemical properties of radioactive waste residues are used to assess the data consistency and uncertainty margins, as well as to predict the long-term behavior of the radioactive waste. This is relevant for repository acceptance and safety considerations.

Our new numerical approach follows a bottom-up simulation starting from the burn-up behavior of the fuel elements in the reactor core. The output of these burn-up calculations is then coupled with a program that simulates the material separation in the subsequent dissolution and extraction processes normalized to the mass balance. Follow-up simulations of the separated reprocessing lines of a) the vitrification of highly-active liquid and b) the compaction of residual intermediate-active metallic hulls remaining after fuel pellets dissolution, end-pieces and technological waste, allows calculating expectation values for the various repository relevant properties of either waste stream.

INTRODUCTION

The quality control of radioactive waste compounds has always been an integral part of the German safety and quality assurance concept for the disposal of radio-toxic waste in a deep geological repository. The latest German NPP was powered on-line back in 1989. Until 1995 reprocessing of nuclear fuel had been legally obligatory, then voluntary. Specific and dedicated waste conditioning methods and technologies had been engaged at reprocessing sites in France, Germany and the UK. As a result of the 2005 atomic act revision for the treatment and conditioning of arising HLW and ILW, the transport of spent fuel and reprocessing abroad has been banned. Now, all spent fuel has to be stored at the NPP sites until further decisions are made on how to proceed and dispose of the growing amount of current HLW and ILW.

The German Federal government is in charge of defining societal targets, safety standards and final disposal of German radio-toxic waste from nuclear facilities, namely the nuclear power stations, research and medical institutions and other facilities that legally deal with radioactive or fissile material. In addition, there is a vast program for the decommissioning of nuclear installations. All accumulated nuclear waste of the past has to be disposed of in a professional and safe manner. Therefore, a LLW and ILW repository for radio-toxic waste with negligible heat generation is under construction. This is the *Konrad* mine and it may be expected to become operational in the year 2019. At the same time, an operational repository for German HLW is still a long way ahead from now, and Germans are far from reaching a societal consent on that issue. Nonetheless, German waste currently reprocessed abroad has to be repatriated into dedicated national interim storage facilities just like the safe intermediate storage of other domestic HLW from the past and current operation of German NPPs.

Anyhow, the product quality control and quality assurance for nuclear waste treatment, conditioning and packaging has never been controversial and QA-methods have gradually been developed to a very high standard.

WASTE CONDITIONING AND PRODUCT QUALITY CONTROL PRINCIPLES

QC/QA methods accompany the waste treatment, packaging, conditioning and transport from the very beginning. Traceability and documentation are key issues. To understand German waste disposal safety philosophy, one has to start from repository considerations. Any repository in Germany has to meet the legal protection targets that are defined in a series of legal acts. Radiation safety for humans, livestock and crops has to be met and guaranteed for as long as radio-toxicity lasts, i.e. ~1 million years. Therefore, any obstruction or intrusion scenario for the disturbance of a radwaste disposal site must be analyzed and evaluated for both, the operational phase and the much longer dormant phase of the repository. Retrieval or rescue options may have to be considered, too. Therefore, it is absolutely essential to know and understand the radioactive waste form properties as precise as possible and to make predictions of the interaction with water of brine over a very long time span of up to ~1 million years. From these long-term repository requirements waste product specifications and long-term properties are derived and made obligatory. This requires the design of dedicated waste conditioning processes and a thorough subsequent verification of the radwaste properties by sophisticated and meaningful check criteria.

Despite the lack of a HLW repository, there is the target and obligation of deep geological burial. For long salt has been the first choice of host rock embedding, while nowadays clay formations are also discussed as an alternative. No decision has yet been made and safety case studies are underway. Nonetheless, waste compounds are being conditioned already and this production has to follow clearly defined guide-lines: First of all it needs approved product specifications that define the verifiable product properties and deliverables of the production process. Regular conditioning plant inspections and audits of the manufacturers and on-site quality inspectors are mandatory to ensure the actual conditioning scheme is maintained within the limits of the process quality approval. An individual compound checking and destructive product analysis can be replaced with production series approval, ensemble spot-checking during conditioning progress, quality assurance control and, last but not least, a 100% checking of the accompanying documentation on each individual waste container from the engaged reprocessing plants. Verifying its physical and radio-chemical properties and nuclear inventory declaration is a prerequisite for transportation.

The present requirement of quality assured and comprehensive knowledge of incongruent legacy waste with short falling documentation and data base needs some reconditioning and repackaging of relatively small volumes or waste series in accordance with dedicated work charts and execution plans. The advantage of a fully QA-based waste conditioning becomes quite obvious, after all. Today, all waste compound production must follow purposeful and dedicated industrial production schemes that are quality assured and then, all waste forms from any of these production lines are "identical" within, well defined and limited uncertainty margins. Their physical and chemical properties are, therefore, predictable and only certified waste compounds from valid, quality assured processes and with properties compliant with the product specifications are accepted for final disposal in Germany. Any lengthy testing of individual waste compounds along-side an individual control and execution plan, whether using destructive or non-destructive metrology, is gradually substituted with quality-assured surveillance and product control.

Once the responsible authorities have defined the virtual or real repository acceptance criteria, four major steps are required for a quality assured and certifiable radioactive waste compound:

1. <u>Product specs approval:</u> The rad-waste conditioning manufacturer seeks approval for the disposability of his nuclear waste compounds with well-defined product characteristics and specifications, i.e. its physical and chemical properties as well as quality traceable identifications. Within the so-called product specification approval these individual, waste stream specific, product specs are assessed and evaluated to be compliant with a number of repository relevant acceptance criteria (Table. I, II) that are issued by the German Federal Ministry of Environment and Reactor Safety (BMU). These repository acceptance criteria are based on scientific evidence for a certain type of rad-waste and for a specific repository in Germany.

2. The second is <u>the process approval</u>: Conditioning manufacturers have to propose and describe their production process and provide evidence that this process is appropriate and delivers waste compounds of the above mentioned specs. The production process must be quality assured by the producer's QA department. This production process is subject to an official approval by German authorities in which all relevant production means, methods and equipment, flow charts and clearance controls as well as quality surveillance aspects and periodical checks are scrutinized, altered if so requested and eventually fixed.

3. <u>Audit and inspections</u>: Once a process is approved, all technical upgrades and modifications are subject to approval as well. All production standards must be maintained within the limits of production process clearance and approval. In addition to the manufacturer's quality inspections and declarations, independent on-site quality inspectors validate and certify the actual performance of the waste production processes, evaluate deviations and may issue quality inspection notes or corrective action requests that have to be dealt with and satisfactorily answered within a certain response time. The independent on-site inspectors work according to a well-defined and officially approved technical inspection and audit plan. Thus any arbitrariness is excluded.

Table I. German Repository Relevant Properties and Characteristic Values for Vitrified HLW [1]

No.	Property	Experimental Assessment (italic) / R&D definition (regular letters)
1	*Total activity α and β*	*Chemical & nuclear analyses: ICP-AES, ICP-MS , α, γ-spectrometry, (AAS), thermal induced mass spec (TIMS)*
2	*Activities of relevant nuclides*	
3	*Criticality safety*	
4	*Thermal properties*	*Measurements & calculation*
5	*Dose rate (n,γ at surface & 1m distance)*	
6	*Surface contamination*	*Wipe test*
7	*Raw glass matrix quality*	*Glass composition, tolerances*
8	*Raw waste properties*	*Chem. composition, oxide content*
9	*Mass balance, incorporation rate*	*Process handling, dosing*
10	*Homogeneity of glass blend & activity*	*Furnace temperature, process time, pouring rate*
11	*State of glass product*	*QA of 7-10*
12	*Hydrolytic stability of waste form*	*R&D characterization*
13	*(empty) container mechanical properties*	*Manufacturers design & QA, welding & corrosion QA*
14	*Weight*	*Weighing*
15	*Stackability & handling*	*Container design*
16	*Residue package identification*	*Visual control*

Table II. Repository Relevant Properties & Characteristic Values for Compacted Metal-Waste [1]

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

As for the German reprocessing program abroad, national inspectors working on behalf of the German authority, Federal Office for Radiation Protection (BfS), base their inspection and auditing on the existing QA structures and restrict themselves to aspects relevant to the re-patriation of radwaste to Germany. The regular summary reports of the independent on-site inspectors are received and studied. There are obligatory regular six-monthly plant and production inspections and audits to the manufacturer and the independent on-site inspectors. Technical modifications, periodical controls, deviations, liaison issues and any irregularities are discussed and explained in detail, and an official report to the German authorities is compiled with a statement of process compliance with the approval. This procedure is mandatory to maintain the production process approval for as long as relevant for German waste.

4. Doc-checking: Last but not least, a thorough 100% check of the radwaste documentation and inventory declaration follows, which is compiled in a dedicated container data file (CDF or CQF). This comprises generally key nuclide safety parameters (e.g. Cs, Sr content, long-lived α-emitters or fissile and minor actinides, e.g. Am, Cm etc.), in addition to basic waste form composition values as guaranteed parameters. The above mentioned 16/14 repository relevant properties (Table. I, II) plus a set of other declared values and safety relevant parameters or inventories are to be documented. The container data file is first checked by the independent on-site inspectors. That is Bureau Veritas in La Hague and Lloyds Register in Sellafield. The conformity certificates of compliance are a prerequisite for transportation. In addition, German inspectors perform another 100% documentation check with independent data consistency calculations and properties derivations. All guaranteed parameters are independently calculated again and compared with the declarations. Such parameters as residual fissile material content, gamma and neutron dose rates and heat load are calculated using a set of our own independent proof tools and programs. The resulting values are compared with measurements prior to loading into the transport and storages casks. Loading itself and final radiation measurements are witnessed by German inspectors.

VITRIFIED HLW

First of all, the production of vitrified HLW has to meet the 16 repository relevant properties for every waste compounds (Table I). Inbound ingredients are in the liquid high-level waste from the spent fuel dissolution process and the glass frit. The chemical analyses of the incoming material flow define the initial state of the vitrification process. The vitrification itself takes place in a big melter, where the glass frit and the oxide forms (calcinate) of the original highly-active liquid (HAL) are blended and homogenized before the molten glass, now loaded with the HAL-calcinate, is poured into container vessels made of specific stain-less steel. Several pours per container are acceptable, depending on the melter capacity. Considerably rapid cooling will put the glass into a long-term meta-stable condition at temperatures well below crystallization temperature. Eventually, the filled glass containers are closed by welding a top lid and decontaminated. Table III and Figure 1a illustrate how the final product characterization is determined from inbound analyses and vitrification process control.

The incoming material is, as mentioned above, the highly active liquid (HAL) from the spent fuel dissolution process and the ingredients of the boron-silicate glass frit. The incorporation rate is the mass ratio of the radio-nuclide oxides to the total weight of glass product and it is a one of the key target process parameters. The maximum incorporation rate is limited by glass stability - unwanted phase segregation and out-leaking must be avoided under any circumstances. A typically achieved value is approx. 20% for a long-term stable glass product, which can be regarded as a homogeneous and isotropic product.

Criticality safety has to be obeyed at any time and the observation of key process parameters defines the final waste product properties. Final experimental checks of the gamma and neutron dose rates and comparison with the calculated expectation values allow consistency checks.

Table III. Inbound and outbound analyses for the vitrified (V) and *compacted (C)* (italic) or **both (bold)** product characterization

Inbound Analysis	Product Characterization
Objective: **Criticality safety for the vitrification plant** **Activity balance of inbound waste**	**Objective:** **Criticality safety for each individual waste package** **Determination of guaranteed & specific parameters** **Activity balance of outbound waste**
Inbound waste stream analysis: (V) chem. analysis of highly active liquid waste concentrate (HAWC) (V) chem. analysis of glass frit *(C) scrap metal collection drum, nuclear analysis*	**Outbound waste stream analysis:** (V) Vitrification process parameters & dose rate *(C) Compaction process params. & nuclear analysis* **Process traceability & product id**
Properties: (V) Homogeneous density (HAWC) *(C) Mainly axially heterogeneous density* (V) Homogeneous activity distribution *(C) Mainly axially heterogeneous activity distribution* **Traceable waste origin**	**Properties:** (V) Well defined glass properties *(C) density: ~4,4 g/cm³* *(C) hulls & end-pieces plus possibly added tech-waste* *(C) stainless steel (Fe) content*

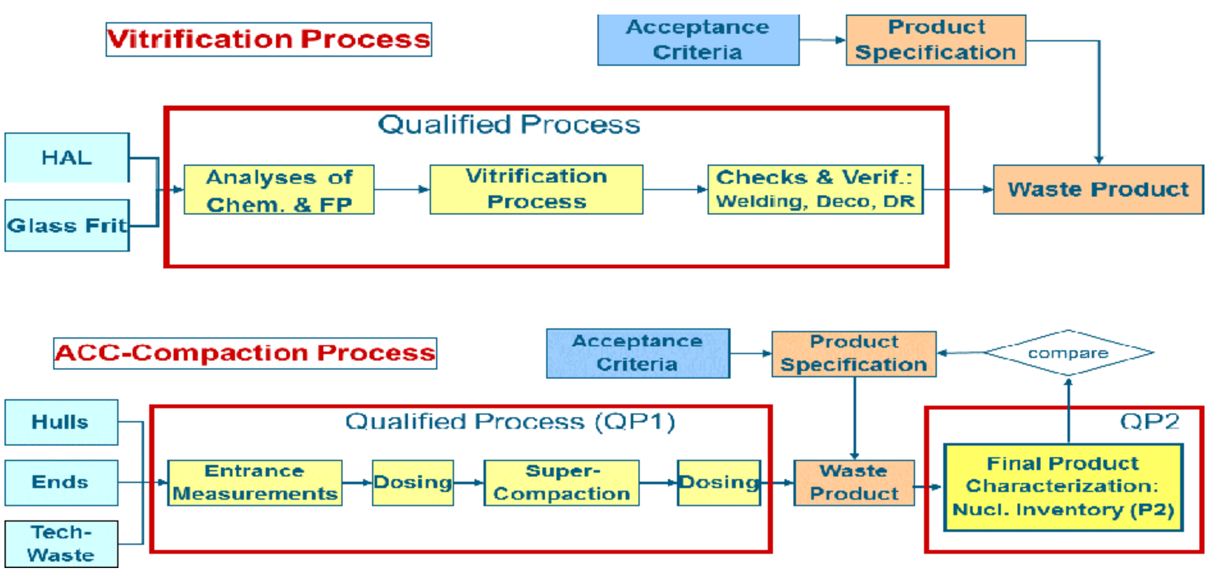

Fig. 1 a) Principle flow chart and quality objective of the waste vitrification process
b) Principle flow chart and quality objective of the metallic waste compaction processes

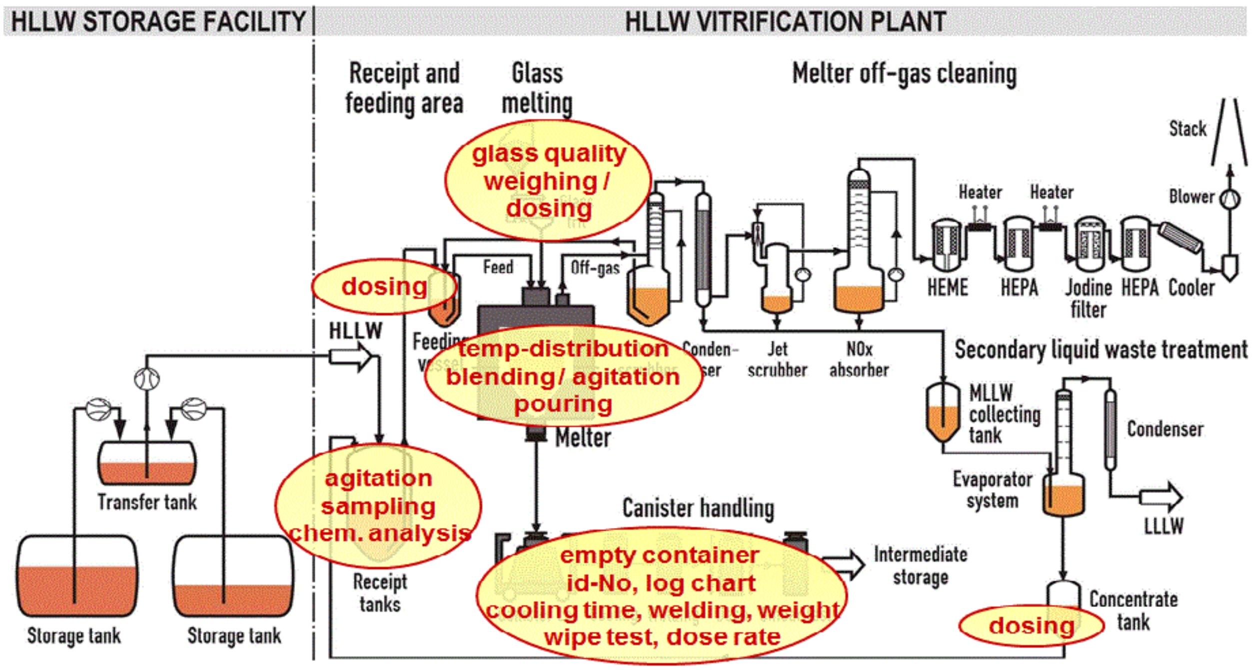

Fig. 2 Vitrification process flow chart and product quality control points (underlying chart from WAK GmbH, Karlsruhe, Germany [10])

The vitrification process itself is controlled and surveyed with persistent readings and regular periodical calibration controls of mass balancing and blending devices (scales and dose meters), the melter temperature distribution and the pouring rate as a measure for the viscosity of the agitated and homogenized molten glass / HAL calcinate blend (Figure 2). The ingredient glass frit and empty containers are also inspected, checked and quality assured. The guaranteed parameters (GP) of a vitrified waste product are declared by the manufacturer. They are verified and validated in a conformity certificate issued by the on-site and national independent inspectors. The GPs for each waste container comprise:

- Glass frit composition (mass ratios of SiO_2, B_2O_3, Al_2O_3, other ingredient oxides in wt-%)
- Actinide masses of total uranium, plutonium and curium content in grams
- Cs-137 and Sr-90 nuclide specific activities in Bq
- Total alpha and beta/gamma activities in Bq
- Alpha and beta/gamma surface contamination in Bq/cm²
- Gamma and neutron dose rate at the surface and at 1m distance in Gy/h
- Heat load in Watt
- Calcinate fraction in wt-% Container mass and ID number, incl. production date and origin.

Proper lid welding is checked, as it is important for undisturbed handling, packing and transportation. The dose rates and heat load are calculated from a few of key nuclides, typically: Sr-90/Y90, Ru106/Rh-106, Sb-125, Cs-134, Cs-137/Ba-137m, Ce-144/Pr-144, Eu-154 for the gamma dose rate and mainly Am-241 and Cm-244 for the neutron emission rate, respectively.

A derived complementary long list of additional nuclides, basic other glass frit and process data, HAL chemical analyses and feed dosing as well as melter temperature and pour data are produced and documented for further cross-checks, if needed.

COMPACTED METALLIC MLW

Shearing and dissolution of spent fuel leaves a lot of scrap metal of fuel cladding, the end-pieces and spacers that are contaminated, irradiated and activated from their life in the reactor core. In fact, this metallic medium level radwaste makes the largest fraction of waste volume from the reprocessing of spent fuel. And the German part of it has to be returned from France. In the UK it was agreed to compensate this with a larger portion of vitrified waste, whereas in Karlsruhe, their highly-active liquid concentrate (HAWC) results from the legacy of early German reprocessing plant prototypes, and the metallic by-products have been dealt with differently. The French product name is CSD-C, an acronym for “Colis Standard des Déchets Compactés”. Chopped scrap metal is collected into compaction cans, in which it is dried and then compressed into disks. Typically 8 of such disks are sorted and then stacked in a stainless steel container, similar to that of the vitrified product.

Although the CSD-C product properties and the production processes are distinctly different from those for vitrified residues, product quality control and QA concepts are to a large degree comparable. It should be mentioned here, that the AREVA NC Quality Control department accounts for the manufacturer’s responsibility for the process and product quality independently from the operations department. Different metrology is used for the inventory determination, though. The characterization is performed by means of non-destructive nuclear measurements without taking advantage of any biased information of the inbound waste streams, such as individual NPP fuel type, burn up or cooling time as various types of disks can be piled in the same CSD-C. Since the varying material composition of CSD-C influences the nuclear measurements, traceability is an important issue. The dosing of hulls, end-pieces and technological waste is performed under video-surveillance. The number and type of end-pieces are noted. The actual compaction process takes place where the filled compaction can is compressed into a so-called compacted disk by mechanical force of up to 200 MPa pressure. The control of incoming scrap-metal masses allows predicting and verifying the thickness and density of produced disk for optimizing the container filling. The resulting compacted disks are temporarily stored before putting them into an empty container, which is then welded and decontaminated and stored just like the vitrified products. The production chart is shown in Figure 1b.

The final waste product properties have to match the approved specification requirements. In fact, production undergoes two distinct qualified processes (QP) that can be detached in time from one another: the first QP1 is about compaction itself and preparing a CSD-C for inventory determination and property characterization in a subsequent but detachable QP2, the nuclear measurement station (Figure 1b). Here, the nuclide inventory is determined by means of sophisticated and combined methods of gamma and neutron spectroscopy [2, 3]. Only a handful of key-nuclides are measured and the complete nuclear inventory declaration is based on numerous correlations. Such nuclides as Cs-134, Cs-137, Eu-154, Co-60 and Sb-125 are measured for their gamma emission and key actinides are determined from steady-state or passive neutron counting (Cm-244). Time resolved neutron spectroscopy yields the mass content of U-235, Pu-239 and Pu-241 while delayed neutron counting yields U-235, U-238, Pu-239 & Pu-241. The mass ratios of U-235/U-238 and Pu-241/Pu-239 are obtained from accompanying correlation calculations; just like cooling time and burn-up is derived from the gamma intensity ratios of

Eu-154/Cs-134 and Eu-154/Cs-137, respectively. The experimentally observable nuclides are shown in Table IV. They are grouped and the bold one is taken as a group representative in the container data file CQF.

Table IV Gamma and neutron measurable key nuclides for CSD-C; bold: group representatives

Metrology	**Measurable Nuclides**
Gamma Spectrometry	Co-60, Sb-125, Cs-134, Cs-137, Eu-154
Passive Neutron Counting	Pu-238 + Pu-239 + Pu-240 + Pu-242 + 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

All other repository relevant properties (Table II) and a required inventory declaration, guaranteed parameters and radiological properties, namely dose rate and thermal properties are derived from this key nuclear measurement. Only the total mass, head welding and contamination swabbing is established independently. Quite obviously, numerous CSD-C can be quite heterogeneous, each of them in a slightly different way. External radiological assessment is much hampered by anisotropic shielding effect inside the CSD-C product and the correct interpretation is consequently rather challenging and not quite easy.

WHY SIMULATING WASTE PROPERTIES? THE NUMERICAL APPROACH AND PROGRAM SELECTION

AREVA NC submits their principle methods and ways of nuclear inventory determination to the scrutiny and approval of their independent on-site inspectors and national surveyors of their foreign customers. However, the underlying neutron and gamma spectra are not attached to the container data files for customers as a comprehensive interpretation would also require the knowledge of the measurement cell geometry and calibration data. As this is, generally, not disclosed to the average customer for the sake of industrial know-how protection, external independent product quality control poses a real challenge, and this is much facilitated by the supplementary numerical simulation of the residue properties.

The German safety authorities require the documentation of each waste residue to be checked and evaluated individually before shipment. Some important compacted metallic waste residue properties are derived from data, such as the aforementioned nuclear measurement cell and calibrations data, that are not generally disclosed except for some individual checking in the course of process quality approval by the external surveyor Bureau Veritas who acts under a non-disclosure agreement. For instance, the correlation functions need to be re-evaluated and checked for their validity under different extreme conditions, like low or high burn-up or long cooling-times. The simulation of such a waste stream also provides expectable waste properties on a theoretical basis and far beyond the limits of the actual production process incurred. We have decided to use world-wide accepted and benchmarked codes and combined them in a new way for the numerical calculation of the waste residue characteristics. The necessary benchmarking of our new combination tools is easier and only needs to be done against already bench-marked and published reference data and proved experimental results, e.g. [4].

Another validity check is derived from activity calculations for individual radio nuclides, e.g. Cs-134, Cs-137, Eu-154 or Sr-90 and others. Here too, we have obtained satisfactory results, and the only input parameters to be used are the reactor core geometry, fuel element composition and design geometry, initial enrichment, burn-up and cooling time. There is no room to go into details here but the results are published elsewhere [5, 6]. An important parameter is the criticality of a specific assembly. Our calculations are in good agreement with AREVA NC declarations for real waste residues (cf. [5]). Controlling the validity range of nuclide correlations used to assess radio nuclide content that is not obtainable easily from other experimental results allows independent inventory declaration checking. For instance, the Pu-mass ratio Pu-241/Pu-239 is derived via correlation with the ratio of Eu-154/Cs-137 β–activities (Eq. 1). However, the uncertainties of the correlation factors a, b in Eq. 1 are defined by the initial enrichment. The parameters A_β are the β–activities of Cs-137 and Eu-154, respectively. An evaluation of the complete numerical calculation is achieved by comparing all the numerical results with experimentally proved properties of real waste compounds, such as vitrified HLW-containers or compacted metal residues and will be published later [6].

$$\frac{mass(Pu-241)}{mass(Pu-239)} = function\ (burn-up) = a\left(\frac{A_\beta(Eu-154)}{A_\beta(Cs-137)}\right)^b \qquad \text{(Eq. 1)}$$

Basically, the idea of our numerical methods is to mimic the waste reprocessing flow chart in its principle steps of processing (Figure 3): First, there is the nuclear fuel life-time in the reactor core: initial enrichment, fuel type, composition, fuel geometry and reactor core geometry, fuel position history and burn-up. Then, the discharge and cooling time in decay pools and interim storages can be combined before the spent fuel element is submitted to shearing into pieces and subsequent fuel dissolution. Now, reprocessing splits-up into the two lines of waste residue production: a) vitrification of dissolved fuel and b) mechanical compaction of shred metal claddings and end-pieces. Supplementary additions of technological waste from the reprocessing process itself can be introduced in the compaction line. A full separation of metallic waste from liquids, rinsing fluids, sludge and slurries, organics and other volatile components prevents uncontrollable interactions and reactions inside the MLW compacted waste that still contains a lot of fairly long-lived radio nuclides. Numerical simulation of the two reprocessing lines, vitrification and compaction allows assessing the physical, nuclear and radio-chemical properties and to predict the long-term behavior.

Our calculation methods can be divided into three individual parts (Figure 3) for the full numerical assessment of any specific type of waste compound under consideration:

A) *Burn-up calculations:* The final burn-up is the most dominant factor for considering spent-fuel properties. Additional reactor parameters are the initial enrichment, soluble neutron poison, linear power density, etc. The basis for the safety analysis and evaluation of the waste characteristics is the expected radioactive inventory at the time of unloading the fuel-element from the reactor. Intermediate storage is modeled as an additional decay period before reprocessing operations. The SCALE software package [7] is our main code for burn-up and decay calculations. SCALE is a modular software framework consisting of different functional modules which are executed in a predefined order from a control routine. The TRITON control routine [8] is mainly used for multi-group 2D and 3D depletion calculations. The nuclide concentrations are calculated with the point depletion code ORIGEN-S, which is derived from the original Oak-Ridge Isotope Generator (ORIGEN) but has been complemented with up-to-date decay parameters and neutron cross-sections.

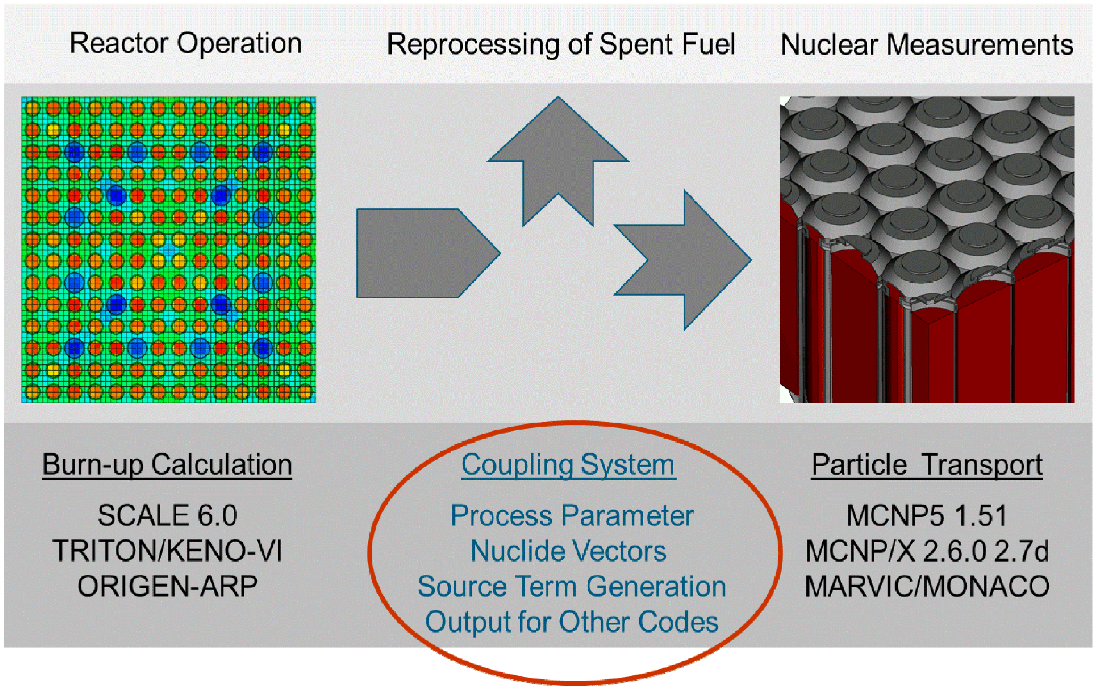

Fig. 3 Tri-fold numerical calculation scheme of A) burn-up, B) Monte Carlo particle transport using world-wide accepted standard codes and C) coupling code for our assessment of specific waste compound streams

B) *Particle transport calculations:* In order to estimate the effects of the particle transport in terms of dose-rates, neutron fluxes or conditions for waste characterization systems the MCNP/X software has been chosen. Depending on the waste properties like the source terms, matrix composition and density, the MCNP/X code is used to estimate the potential gamma and neutron dose rates. The MCNP/X model parameters have to be in compliance with the parameters for the considered waste stream. This is done in the third part of the software compilation, a coupling code that combines the Scale output with the MCNP-X input files.

C) *The third part of the calculation system* is a new and self-developed C++ class framework, which links the burn-up calculations with the particle transport simulation taking into account the declared waste residue properties. The binary nuclide concentration file of ORIGEN-S is used to store the most relevant information about the waste package, e.g. the waste matrix composition and the neutron and photon source terms. The object-oriented design of the framework is based on a C++ model of a nuclide with all the relevant information on nuclide identifier, whether measured or declared. All other program classes depend on this implementation and extend the functionality of the software. A nuclide-vector, for instance, is modeled as a collection of independent nuclides. Several utility classes act on the objects for statistical evaluation or conversion of the units. Since it is not known which waste streams are to be considered or what analyses are needed for future evaluations, this frame-work is also made available for rapid prototyping with the ROOT software developed at CERN [9]. All classes are stored in a ROOT-compatible shared library enabling the user to rapidly develop new analysis cases. One of the main features is the creation of MCNP/X models for a conceivable waste-stream. A general simulation input template is used. Many parts of the model are replaced by calculations using a SCALE nuclide binary file. Examples are the waste matrix composition or the gamma and neutron source-terms, which are then used to evaluate the dose-rates and other results of the particle transport.

RESULTS, APPLICATIONS AND PROOF TOOLS

To demonstrate the quality and power of our numerical tools, we have chosen to compare the declared and calculated nuclear properties of vitrified residues from the VEK vitrification plant of WAK Karlsruhe GmbH [10]. The declared glass composition, masses and nuclide concentration from a 30% subset of the electronic data files have been used to calculate binary data input files for the MCNP-X particle transport calculations. As an example, the calculated thermal output power and neutron contact dose rate are presented here and compared with VEK-waste declarations obtained from measurements (Figure 4 a, b). The agreement is strikingly good. Both the thermal power and the neutron contact dose-rate are guaranteed parameters and must not exceed 0.74 kW and 2.0 mGy/h, respectively. The bulk of the VEK containers include approx. 16 wt-% of calcinate loaded into the glass frit matrix. Obviously, less active containers had been produced in the start-up and close-down phases. Container no. 84 is less active because of a melter drain for maintenance. The program follows these depleted containers very well, just from the aforementioned input values without any further parameter variation.

The dose-rate calculations result from internal binary MCNP/X input files. The individual compound specific radio-nuclide content is the only adjustment parameter. It seems justified to restrict the dose-rate calculations to the two main uncertainty reasons, i.e. neutron emission from spontaneous fission events and (α,n) reactions in HLW loaded glass. The error bars in Figure 4a are the combination of the statistical uncertainties of the Monte-Carlo simulation and the uncertainties associated with the neglect of the unspecified nuclides contributing to the total α-activity, which causes the approximate 2-3% systematic discrepancy between measured and calculated data. The source-terms and associated energy dependent spectra are calculated with ORIGEN S [11]. Nonetheless, both the thermal power and dose rates are well below the permissible limits of 0.74 kW and 2.0 mGy/h for each waste container.

The measured vs. calculated dose rates are in very good linear correlation. In fact, linear regression between the two data-sets yields a slope coefficient of 0.9995 with an R^2-factor of 0.99 of the regression function. The computational framework of coupling the SCALE/ORIGEN-S functionality with MCNP/X Monte-Carlo calculation is capable of predicting the derived parameters from the VEK high-level glass waste-stream. The next steps will be to extend the analysis to the whole campaign of the VEK waste-products and finalize the neutron and γ-dose-rate analyses. The results so far seem very promising for consistency checking of different homogeneous and heterogeneous high- and intermediate-level waste-streams for German radwaste repositories. These numerical simulations yield numerous applications. First of all, the independent assessment and evaluation of radwaste inventory declarations contribute widely to the obligatory product quality control of waste residues to be disposed of in a repository site. The German authority for nuclear waste (BfS) requires from the national inspector to check radwaste properties of the current production during on-site inspection visits. Therefore, an independent proof tool is needed to accomplish this task. This so far pending task can now be completed by a spin-off of the comprehensive computational method. Moreover, it can be extended to all kinds of waste, homogeneous or heterogeneous at whatever activity levels, HLW, MLW or LLW. Nuclide correlations and the embedding material matrix are general key input parameters for the numerical assessment of the physical and radio-chemical properties of the radioactive waste under consideration.

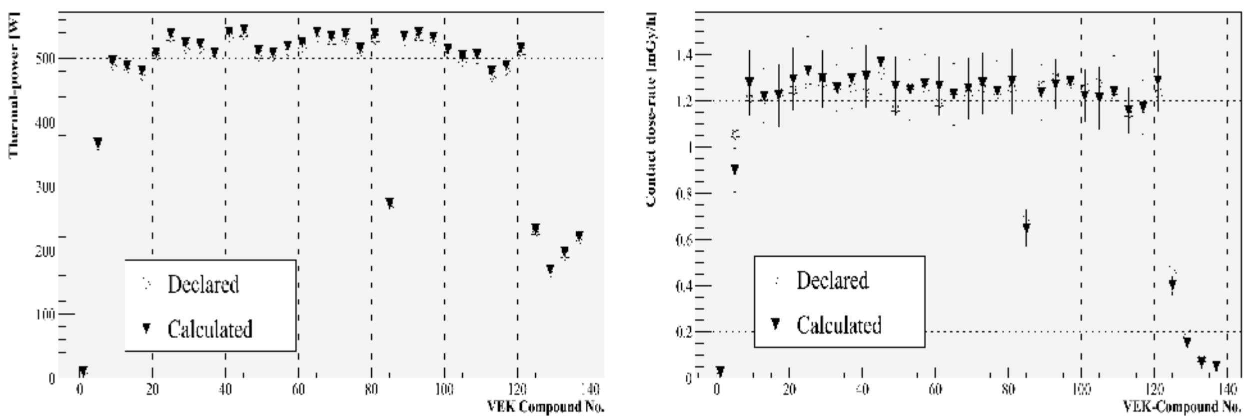

Fig. 4 Comparison of measured [12] and calculated thermal power (a, left) and neutron contact dose-rate (b, right) of VEK vitrified waste, plotted vs. the consecutive container id no.

However, much further reaching is the assessment of repository inventory propagation in time. Clear understanding of how the radioactive waste properties develop under many environmental conditions is a requisite for the solid foundation of any repository safety case. Conceivable scenarios can be calculated and features, events and processes of theoretical repository propagation can be assessed. We are following a bottom-up approach in which the waste residue properties are calculated and then any interaction with surrounding material, e.g. bedrock can be introduced.

SUMMARY AND OUTLOOK

The principles of the German product quality control of radioactive waste residues from the spent fuel reprocessing have been introduced and explained. Namely, heat generating homogeneous vitrified HLW and heterogeneous compacted metallic MLW have been discussed. The advantages of a complementary numerical property simulation have been made clear and examples of benefits are presented.

We have compiled a new program suite to calculate the physical and radio-chemical properties of common nuclear waste residues. The immediate benefit is the independent assessment of radio-active inventory declarations and much facilitated product quality control of waste residues that need to be returned to Germany and submitted to a German HLW-repository requirements. Wherever possible, internationally accepted standard programs are used and embedded. The innovative coupling of burn-up calculations (SCALE) with neutron and gamma transport codes (MCPN-X) allows an application in the world of virtual waste properties. If-then-else scenarios of hypothetical waste material compositions and distributions provide valuable information of long-term nuclide property propagation under repository conditions over a very long time span. Benchmarking the program with real residue data demonstrates the power and remarkable accuracy of this numerical approach, boosting the reliability of the confidence aforementioned numerous applications, namely the proof tool set for on-the-spot production quality checking and data evaluation and independent verification. Moreover, using the numerical bottom-up approach helps to avoid the accumulation of fake activities that may gradually build up in a repository from the so-called conservative or penalizing nuclide inventory declarations.

The radioactive waste properties and the hydrolytic and chemical stability can be predicted. The interaction with invasive chemicals can be assessed and propagation scenarios can be developed from reliable and sound data and HLW properties. Hence, the appropriate design of a future HLW repository can be based upon predictable and quality assured waste characteristics.

REFERENCES

1. Kienzler, B., Loida, A. (Eds.), Endlagerrelevante Eigenschaften von hochradioaktiven Abfallprodukten, Laboratory Report, Forschungszentrum Karlsruhe, FZKA 6651, 2001, pg. 70ff, ISSN 0947-8620, http://bibliothek.fzk.de/zb/berichte/FZKA6651.pdf

2. Toubon H., Gain T., Perot B., Lyoussi A., Raoux A.-C., Melhman G., De Brimont X., Tulle P. A., 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

3. Toubon, H., Vuillier, Gain, T. Huver, M., 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.

4. Raoux, A.-C., Lyoussi, A., Passard, C., Denism, C., Loridon, J., Misraki, J., Chany, P., Transuranic waste assay by neutron interrogation and online prompt and delayed neutron measurement, Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms Volume 207, Issue 2, June 2003, Pages 186-194

5. Schneider, S., Tietze-Jaensch, H., Bosbach, D., Odoj, R., "Numerical Tools for the Evaluation of Super-Compacted Rad-waste residues", Proc. Supercomputing in Nuclear Application and Monte Carlo Conference, (SNA-MC 2010), Oct 2010, Tokyo, Japan

6. Schneider, S., Ph.D. thesis, to be published, Aachen Technical University, Germany, 2012

7. Bowman, S. M., "SCALE 6: Comprehensive Nuclear Safety Analysis Code System," Nucl. Technol. 174(2), 126-148, May 2011.

8. De Hart, M.D., Gauld, I. C. and Williams, M. L., "High-Fidelity Lattice Physics Capabilities of the SCALE Code System Using TRITON", Joint Int. Topical Meeting on Mathematics, Computation & Supercomputing in Nuclear Applications (M&C + SNA2007).

9. Brun, R. and Rademakers, F., "ROOT - An Object Oriented Data Analysis Framework", Proc. AIHENP'96, Lausanne, 1996, Nucl. Inst. & Meth. in Phys. Res. A 389 (1997) 81-86.

10. Grünewald W., Plaschke M., Roth G., Tobie W., Weiss K-H., Weisenburger S., Stollenwerk A., Gauthier R., Eissler A., „Process Control and Glass Product Quality Assessment Applied in the VEK Plant" Proceedings of GLOBAL 2011, Makuhari, Japan Dec. 11-16, 2011 Paper No. 392621

11. Gauld, I. C. and Murphy, B. D., Updates to the ORIGEN-S Data Libraries Using ENDF/B-VI, FENDL-2.0, and EAF-99 Data, ORNL/TM-2003/118, ORNL, Oak Ridge, TN, May 2004.

12. VEK container quality files, Karlsruhe 2009-2011, WAK GmbH, unpublished

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