Conditioning of Pu-containing radioactive waste generated in the Hotlab: implementation of an upgrade

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ABSTRACT

To achieve an optimal strategy for final disposal or interim storage, solid radioactive waste has to be sorted and treated. This strategy is based on the requirement to reduce the volume of waste designated for long term disposal, mainly for economical and safety reasons. In addition, the radiological limitations for shipping of the waste packages to a disposal site have to be taken into account.

In case of the operational Pu-contaminated and –containing waste from the PSI-Hotlab in Switzerland, the existing conditioning method needed improvement. The existing authorized specifications of the conditioning process had to be revised for an optimization of the interim-storage and the deep geological disposal. This was done by the PSI Section for Dismantling and Waste Management.
The improvement focused on technical aspects as well as improvement of the documentation for the certification by final depositor (Nagra) and the safety authority (ENSI). This work started in 2008. In 2012 ENSI delivered an authorization for realize the first conditioning according the new specification, formally as a certification test prior to final approval by the ENSI.

Technical construction of a waste package:

- The waste is packed in the Hotlab in 20-Liter-drums (4.4 gallons) and
- pressed by a 120 t press into a stainless steel pipe, which has been
- cemented in a 200-Liter-stainless-steel-drum (44 gallons) in the Section for Dismantling and Waste Management
- The waste has to be covered by 10 cm of concrete to comply with the standard requirements of the final depositor (Nagra)
- The 200-l-waste drum is made of corrosion-resistant steel

Radiological characterization:

- Maximum alpha-activity of 170 GBq as the sum of the activities of Pu-238, Pu-239, Pu-240, Pu-242 and Am-241
- Maximum beta-/gamma-activity of 3.9 TBq as Pu-241
- U-234 and U-238 limited to 47.3 MBq
- U-235 limited to 0.61 MBq
- All other radionuclides are limited to the A2-value according to ADR 2009 respecting the “sum-rule”

INTRODUCTION

The Paul Scherrer Institute (PSI) is the largest national research centre in Switzerland. Its multidisciplinary research is dedicated to natural science and technology, i.e. solid state physics and materials sciences, life sciences, elementary particle physics, nuclear and non-nuclear energy research, and energy-related ecology. PSI operates facilities dedicated to research in nuclear fields and particle physics and uses nuclear methods in materials and life sciences: for example proton therapy or research in the Hotlab. These inevitably produce radioactive wastes which have to be characterized, treated, stored and eventually disposed of.
The conditioning processes for radioactive waste packages are part of an accredited waste management process of the institute. This is a business of the “Sektion Rückbau und Entsorgung” (RBE) – Section Dismantling and Waste Management. Proved and accepted methods need to be developed for safe handling, conditioning and storage. In addition to this, PSI has the task of processing and storing the radioactive waste originating from Medicine, Industry and Research (German short form: MIF). The Swiss Federal Interim Storage facility for MIF waste is part of PSI. By law the Federal Interim Storage is a nuclear facility.

In the case of the operational Pu-contaminated and –containing waste from the Hotlab (under IAEA-control) the existing conditioning technique in a specialized package (compacted 20-Liter-drums cemented in a 200-Liter-drum) for the interim-storage and the deep geological disposal had to be improved with respect to the following criteria:

- According to the existing specifications, the waste sorts are divided into non-irradiated fissile material under IAEA control and wastes containing small amounts of burnt up fuel. They had to be unified to one method.
- A safe and easy conditioning process should result.
- To optimise the volume efficiency.

- To meet the needs of the Hotlab, the inventory of the Beta/Gamma-nuclides (mainly activation products) should be raised.
- To facilitate and shorten the proving process by the competent authority and to minimize administrative efforts, the changes shouldn't be fundamental.
- The risk of criticality is to be excluded.
- Additional requirements of the final disposer (Nagra) has to be fulfilled:
  - 200-l-stainless steel drums
  - smallest cover of 10 cm of concrete all around the waste
- Additional requirements results from the update of the shipment, interim Storage and final repository regulations

A technical report, called "specification", describes the improved conditioning process and the waste package. The conformity of the specification with the regulatory guidelines is investigated in advance by the Swiss Federal Nuclear Safety Inspectorate (ENSI). The ENSI is proofing the process, the possibility of shipment, interim storage (accident study) and the fulfillments of the demands of a deep geological repository.

THE REGULATORY FRAMEWORK

The conformity of the waste package which has been developed to the requirements of a future deep geological storage has been verified in advance by the final disposer "National Cooperative for the Disposal of Radioactive Waste" (NAGRA). This check has been completed for this package end of 2011.

The ENSI is the controlling authority for nuclear facilities. In particular

- all activities concerning conditioning of radioactive waste (ENSI guideline B05 [1])
- responsibility for all material declared as radioactive waste (Art. 2 Nuclear Energy Ordinance (KEG) [2] connected with Art. 27 Radiation Protection Ordinance (StSG) [3])

Therefore the nuclear facilities of PSI including the radioactive waste management facilities are under responsibility of the ENSI. These are the so called "operation box" for waste treatment, the storage hall for all kind of radioactive waste and the Federal Interim Storage.

A conditioning process, described in a specification, and the produced waste packages had to fulfil the following ENSI guidelines:

- HSK-B-05 [1] for conditioning
- HSK-R-29 for interim storage [4]

The conditioning process must be approved by the ENSI before getting into routine operation.

QUALITY ASSURANCE FOR THE PRODUCED RADIOACTIVE WASTE PACKAGES

The conditioning processes for radioactive waste are part of the PSI accredited waste management procedure. This guarantees the accurate production of the waste packages. The procedures are audited by the regulatory authorities. The main elements for a waste package are:

- accredited Management-System for the conditioning process: ISO/IEC 17020 [5] and EN/ISO 9001 [6]
- Specification [7] for the waste package, proofed by the authority: this technical report describes waste, materials, nuclides, production, construction of the package and transportation.

THE WASTE, STARTING POINT HOTLAB

The origin of the Pu-containing waste is the PSI Hot-Laboratory. In the Hotlab the Division Hotlaboratory (AHL – operator of the Hotlab) and Laboratory for Nuclear Materials (LNM) are the principal research unit in Switzerland in the domains of materials behavior and ageing in nuclear installations. They do examination and analysis of fuel behavior, advanced fuels, reprocessing and damage analyses of core structural components. There is a focus on different aspects of nuclear reactor core internals. The most important is the fuel itself, consisting of ceramic fuel pellets and a surrounding cladding. The interest is on the physical, chemical and mechanical properties of these components and their respective influence on the performance. Further aspects concern the reprocessing of the fuel and sophisticated process optimizations.

COLLECTING THE PU-CONTAINING WASTE IN THE HOTLAB

Collecting of Pu-contaminated and –containing waste (fission- and breeding material containing products) in the Hotlab is part of the Hotlab waste management system.
Components of the solid organic and inorganic waste are in average:

- 10% PVC,
- 5% glass,

- 5% rubber,
- 45% steel,
- 5% ceramic,
- 20 to 45% cellulose and
- 0.124% of fuel.

Two different sources of Pu-containing waste exist in the PSI-Hotlab. Both types are collected into 20-Liter-drums (6 to 12 kg):

(1) The solid waste containing high level alpha activity out of the so-called Pu-trakt (laboratories dedicated to the handling of unirradiated fuel in the PSI Hot-Laboratory). This mixed waste results from the treatment of material containing fissile and breeding material in glove boxes and is under IAEA control
(2) Alpha- and fission product-contaminated remnants from the hot cells (irradiated material).

(1) For the material under IAEA-control, the main declaration is resulting from Hotlab´s strictly controlled documentation (0.1 g exact). This special information has to be transferred to the Section for Disposal, in order to insure the traceability by the IAEA (Safeguard). Furthermore, calculations (i.e. ORIGEN, input output calculation) and measurements (i.e gamma scanning) insure the proper declaration of the waste activity.

(2) The declaration of Uranium-, Plutonium- and Americium-contaminated waste (not IAEA controlled) is based on calculations and measurements. The Hotlab waste management procedures insure the compliance of the activity and dose rate limits. The documentation for each 20-Liter-drum includes the material, weight, dose rate, the nuclides, the activity and the reference date.

The 20-Liter waste drum are stored in shielded areas in the Hotlab and transferred batch wise to the Section RBE prior the final conditioning treatment.

PLANNING OF THE FINAL CONDITIONING IN THE SECTION DISMANTLING AND WASTE MANAGEMENT (RBE)

With the transfer of the drums to the RBE, the documentation takes place in the electronic database of Switzerland, the Information System for Radioactive Materials (ISRAM).

- The 20-Liter-drums with low dose rates are stored in the RBE-area, to be available for the next final conditioning campaign.
- Drums with high dose rates are transferred in shielded containers directly to the processing box at the RBE-area in time for a direct conditioning.

The conditioning is performed in a walk in cell equipped with a 120 t press for volume reduction.

The conditioning campaign has to be accurately planned, to optimize the number and proper sequence of pressed 20-Liter-drums for each produced 200-Liter-waste package. The resulting activity and dose rate of the produced 200-Liter waste package is pre-calculated. The calculation is done by a spreadsheet application (Excel), to insure the adherence to the limits of the specification. The choice for the handling of the 20-Liter-drums in order to keep the contain of the final package in the specification is made using the following criteria:

- dose rate
- activity
- material, weight and estimated volume

As a result of the planning, the filling of the first seven drums of each 200-Liter waste package is determined. The rest of the void of the waste package is filled with 20-Liter-drums, containing low dose rate and activity.

PREPARATION OF THE EMPTY 200-LITER PACKAGING

A 200-Liter-drum made of stainless steel is used. The inside of the drum is prepared with an end closed steel pipe (0.36 m diameter) surrounded by a special grout (10 cm = 0.33 foot). A 10 cm cement cover around the waste fulfills the demands of the standard models of a thermal attack of the final repository. The pipe is working as a press matrix for the 20-Liter waste drums. The bottom is filled with 10 cm grout. As a result, there is approximately 60 Liters (16 gallons) free volume for the pressed waste in the pipe.

FINAL CONDITIONING

In a walk in cell, the delivered waste drums are pressed to pellets in a pipe, stacked in 200-Liter-drums and embedded in a special grout. Approximately 6 to 12 drums are pressed in each 200-Liter-barrel, with a weight of about 80 kg +- 35 kg. After this, the remaining space between steel pipe and pressed waste is filled with a special flowable grout (“PSI-Vergussmörtel”).

The filled grout has to harden a predetermined time: After 14 days of hardening (when main hydration processes are finished) a compressive strength of 48 MPa is reached. This is well above the 10 MPa, demanded by the authorities (ENSI guideline B05 [1]). The resistance against leaching (< 5 micro m/d) of radioactive nuclides in dematerialized and gypsum water has been proven (ENSI guideline B05 [1]).
Finally, the remaining void is filled with grout as an inactive top in the drum. The waste must be covered by about 10 cm grout. After a final quality check, the conditioned waste package is ready to go to the interim storage. There it should be stored, until the Swiss deep geological disposal is constructed, in order to ensure the long-term protection of man and environment. This is under the condition that the Federal Interim Storage at PSI comply to the evolving safety regulation specially in view of the large alpha content of these waste packages. A new accident case study of the interim storage has been realized but is still not validated by the safety authority. The authority has additional claims on this study, which must be processed again and again. This takes time. If the interim storage of these waste package at PSI is not possible, another solution will have to be found. An option is the storage in the interim storage of the nuclear power plants (ZWILAG). This facility is located close to the PSI.

The whole practical and documental work, including the stop points for the quality checks (i.e. test of hardening, visual test for cracks in the grout or open water ...) as well as process-controlling check-lists are described and embedded in the management system of the RBE. This formalized tool offers the opportunity to be flexible and more up to date with the documentation of the production process in the case of changes. The formal work is concentrated on the changes; you don't have to repeat a specification of the whole process.

CHANGES TO THE EXISTING PROCEDURE

Summarizing of process steps

According to the old specification, the path for Pu-contaminated (fission product containing) radioactive material and Pu-containing fissile and breeding material from the Hotlab was separated already during the waste conditioning in the Hotlab. This procedure was described and established by PSI and approved by the ENSI in two separate specifications. The difference between these specifications was the ratio of radionuclides in the waste and the IAEA-documentation. There was only a small difference, however, between the material components / composition. The substantial formal difference is the nuclide inventory, under IAEA-control (Pu-containing material) or not (Pu-contaminated). The conditioning practice and building of the waste package is for both paths the same.
Changes take place in the research fields of the Hotlab as well as in the radioactive waste composition, especially the involved nuclides. As a result, in the Hotlab a strict separation of the two waste streams is no longer practical. It is also not helpful for the conditioning at the RBE. In reflection of the past, it has shown that there isn't a significant difference in the radiological characterization between the two Pu containing-waste paths any more, except the IAEA-declaration. An easy way for the logistic and planning of the conditioning of the 20-Liter waste drums is to take them together and have a wider choice for the planning of the filling of the final package (200-L Drums). Drums with a high dose rate and / or activity could be easier mixed with others in order to obtain a more uniform waste package. The storing of the 20-Liter-drums is also reduced, with the option of a wider choice of drums to be mixed.

To keep the conditions of the specifications, the resulting dose rate and loaded activity of a 200-Liter-drum has to be considered:

- To avoid high activity concentrations in 200-Liter-drums, for each drum a small number of 20-Liter-drums with high dose rate and activity content is mixed with selected drums of low dose rate and small activity. This results in a moderate total activity per drum.
- To avoid high dose rates at the drum surface, those 20-Liter-drums with high dose (max. 50 mSv/h) rate are positioned in the middle of the steel pipe.

Today there is only one specification for the conditioning of Pu-containing radioactive waste.
The two theoretical separately existing methods for the conditioning, divided into

- containing fission and breeding products (IAEA-controlled) and
- Uran-, Plutonium- and Americium-contaminated waste

are unified to one method. The inventory of nuclides named in the specification is opened. The strict declaration of the IAEA-inventory for fissile material is still guaranteed.

To be ready for future changes in research, there is a limitation for all other nuclides to the A2-value equal to the transportation rules for radionuclides:

- Two separately existing methods for the conditioning, divided into fission (Pu-containing) and non-fission (Pu-contaminated) products with an existing gap for the approved nuclide-content, are unified to one method

- To meet the requirements of the Hotlab, the inventory and the level of radioactivity content is carefully adjusted to the following values:
  - Maximum alpha-activity of 170 GBq as the sum of the activities of Pu-238, Pu-239, Pu-240, Pu-242 and Am-241
  - Maximum beta-/gamma-activity of 3.9 TBq as Pu-241
  - U-234 and U-238 limited to 47.3 MBq
  - U-235 limited to 0.61 MBq
  - All other radionuclides are limited to the A2-value according to ADR 2011 [8] respecting the "sum-rule"

With these radiological limitations of this special Pu-type, there is a production rate of approximately 12 waste packages per year.

Organization
A potential safety hazard was excluded with the implementation of an operational regulatory: Nitric acid solutions are sometimes wiped away with cellulose cloth. For that reason, cellulose nitrate could occur in the waste, with the known risk of fire hazard. To be absolutely sure to prevent a thermal excursion, all acid fluids are immediately neutralized with sodium hydrogen carbonate ($NaHCO_3$) and dried in the Hotlaboratory prior to conditioning.

Technical
Technical changes in the walk in cell made the work more efficient. A 100 t press was boosted to 120 t.

- To maximise the volume efficiency, the performance of the press was boosted and as a result, the press-matrix was changed from a concrete pipe to a steel pipe to withstand the required forces

A technical change is the use of corrosion-resistant steel for the 200-l-drums.

- The gas production in the final deposit should be reduced by using stainless steel waste Drums instead of zinc coated drums.

An improvement was made by covering the waste with 10 cm mortar without steel reinforcement.

- The standard model for the safety case studies from the Nagra are satisfied

Integration to the accredited management system
The existing and accredited management system is used to accelerate changes and to minimize administrative efforts with work instructions.

- To facilitate and shorten the proving process by the competent authority and to minimize administrative efforts, the detailed description of the process (work instruction) is embedded into the existing accredited management system

Criticality
In comparison to the old specification the limitations for fissile nuclides were not increased. This credit is taken to exclude in an established way the risk of criticality. The mass of the fissionable material in this waste package is far away from its theoretical critical mass.

- The risk of criticality is excluded in an established way (no possible criticality event resulting from the activity limitation)

Open request from the authority
The activity and the fissile inventory of the waste package is over the limits of a transport Type A package. To fulfill the regulations and the requirements of the ENSI, the PSI has to find a package B(M)F to realize a standard shipment of these waste packages (to the final repository).
Further more the alpha-activity of the produced waste packages overcome the limit used for the actual accident case study of the Federal Interim Storage and a new study has to be realized and approved by the ENSI. Until

this revision is accepted by the ENSI, the new waste package cannot be transferred to the interim storage. The new calculation for the accident case study is in process with varying conditions.
Finally, the laboratory used for the conditioning "Waste Laboratory" has an old approval. The lab was renovated ten years ago for the safe conditioning of Pu-containing waste This action was proved by the competent authority. The Laboratory will have to be relicensed for the future conditioning of alpha containing waste (after the first test conditioning phase).

CONCLUSION

In the case of the operational Pu-contaminated and Pu–containing waste from the PSI-Hotlab the existing conditioning method in a specialized package for the interim-storage and the deep geological disposal has been improved. The changes are soft, to keep the experience and to accelerate the process of approval by the final disposer and the authority.

- Two separately existing methods for the conditioning, divided into
  - non-irradiated fissile material under IAEA-control and
  - waste containing small amounts of burnt up fuel contamination
  with an existing gap for the approved nuclide-content, has been unified to one method
- HNO_3 conc. is neutralised in advance by addition of $NaHCO_3$ to prevent a thermal excursion during the whole process
- To maximise the volume efficiency, waste drums are compacted by a 120 t press. The performance of the press was boosted and as a result, the press-matrix was changed from a concrete pipe to a steel pipe to withstand the required forces
- To meet the requirements of the Hotlab, the inventory and the level of radioactivity content is carefully adjusted to the following values:
  - Maximum alpha-activity of 170 GBq as the sum of the activities of Pu-238, Pu-239, Pu-240, Pu-242 and Am-241
  - Maximum beta-/gamma-activity of 3.9 TBq as Pu-241
  - U-234 and U-238 limited to 47.3 MBq
  - U-235 limited to 0.61 MBq
  - All other radionuclides are limited to the A2-value according to ADR 2009 respecting the "sum-rule"
- To meet the requirements of the final depositor the 200-l-drums are made of stainless steel.
- To facilitate and shorten the certification process by the competent authority and to minimize administrative efforts, the detailed description of the process (work instruction) is embedded into the existing accredited management system of the disposal section.
- The risk of criticality is excluded (limitation of included activity).

The radioactive waste is embedded in a safe and stable manner in a concrete matrix in a 200-Liter-drum. The waste is covered by a minimum layer of 10 cm concrete. The grout completely fulfills the requirement of the guideline B05 (conditioning of radioactive waste) of the ENSI, the approving authority.
The method, especially the waste package, is accepted by the final disposer (NAGRA). The approval by the authority (ENSI)is on going. To reduce the hazard of unconditioned alpha toxic waste and to get experienced with the conditioning process ENSI approved the conditioning of a limited amount of waste in order to test and prove the new procedure in 2012/2013.

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