Conditioning of Pu-containing radioactive waste (LLW) generated in the Hotlab of the PSI - 10199

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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 reasons. In addition, the radiological limitations for transportation of the waste packages to a disposal site have to be taken into account.

In case of the operational Pu-contaminated and –containing low-level waste (LLW) from the PSI-Hotlab (under IAEA-control) in Switzerland, the existing conditioning method needed improvement. The existing authorized specifications of the conditioning process are to revise for optimization of the interim-storage and the deep geological disposal. This was done by the PSI Section for Dismantling and Waste Management.
The work is focused on technical aspects as well as aspects of documentation and the approving authority.

Technical construction of a waste package:
- The waste is compacted in the Hotlab in 20-Liter-drums and
- pressed by a 120 t press into a steel pipe, which has been cemented in a 200-Liter steel standard barrel in the Section for Dismantling and Waste Management

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”

The results are shown here.

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 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 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.

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

- According to the 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 requirements 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.

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 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).

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.

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

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

The process must have the approval 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 and the regulatory controls guarantee the accurate production of the waste packages:

- 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, 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 understanding 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.

In general, the solid waste coming from the so-called Pu-trakt (of PSI Hot-Laboratory) is separated into waste containing alpha activity in a high degree. Those are collected in 20-Liter (6 to 12 kg) drums and in addition to this, the radioactive waste is process orientated divided into:

- fissile and breeding material containing mixed waste (IAEA-controlled) from the glove boxes and
- Alpha- and fission product-contaminated remnants from the hot cells.

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

The declaration of Uranium-, Plutonium- and Americium-contaminated waste (not IAEA controlled) is based on calculations and measurements. An internal Hotlab regulatory takes care, to drop below 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.

In the following, they are stored and shielded in the Hotlab and transferred to the Section RBE for the final condition.

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 shielding containers directly to the processing box at the RBE-area, simultaneously with the conditioning process.

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

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. In advance, the resulting activity and dose rate of the produced 200-Liter waste package is calculated.
The selection takes place by following criteria

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

PREPARATION OF THE EMPTY 200-LITER PACKAGING

A standard 200-Liter barrel made of steel is used. Inside, the barrel is prepared with a steel pipe (0.36 m diameter) surrounded by a special grout (10 cm). 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 are 65 Liters 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. Proximately 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 stay a predetermined time to harden: 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]).
At least, the remaining void is filled with grout as an inactive top in the barrel. After a final quality check, a finally conditioned waste package is ready to go to the interim storage. There it is stored, until the deep geological disposal is constructed, in order to ensure the long-term protection of man and environment.

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

The path for Pu-contaminated (fission product containing) radioactive material and Pu-containing fissile and breeding material from the Hotlab was separated: Arising in the Hotlab and ending with the conditioning at the RBE. This procedure was described and established by PSI and approved by the ENSI in two 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. 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, a strict separation of the two waste streams is in the Hotlab practical, but not useful for the conditioning at the RBE. The past has shown that there isn't a significant difference in the radiological characterization between the two Pu-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 planning. The storing of the drums is also reduced.

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

- To avoid high activity concentrations in 200-Liter-barrels, for each barrel a small number of 20-Liter-drums with high activity content is mixed with selected drums of low activity. This results in a moderate total activity per barrel.
- To avoid high dose rates at the barrel 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 <u>other radionuclides</u> are limited to the A2-value according to ADR 2009 [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 hazards 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.

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

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 critical mass of the fissionable material in this waste package is far away from its nuclear properties of criticality.

- The risk of criticality is excluded in an established way (non possibility of criticality with this activity limitation)

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 is improved. The changes are softly, 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, had to be 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 pressing. 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 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 of the disposal section.
- The risk of criticality is excluded (see the limitation of included activity).

The radioactive waste is embedded in a safe and stable manner in a concrete matrix in a 200-Liter-barrel. The grout completely fulfills the requirement of the guideline B05 (conditioning of radioactive waste) of the ENSI, the approving authority.

The method is in the process of approval by the final disposer (NAGRA) and the authority (ENSI).

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