Conditioning of Pu-containing Radioactive Waste Generated in the Hotlab: Where are We Now – 15400

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ABSTRACT

To achieve an optimal strategy for interim storage and final disposal, 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 as well as the improved regulatory requirements for material under IAEA control.

In case of the operational Pu-contaminated and –containing waste from the PSI-Hotlaboratory in Switzerland, the existing and over years practiced 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 issued an authorization to realize the first conditioning according the new specification, formally as a certification test (“Type Test”) prior to final approval by the ENSI. But, although a licensed method by the competent authority is available, modifications could be necessary and new tasks come up. In 2013 the PSI has produced 13 waste-packages as a Type Test. To obtain the legal authorization (“Type Approval”), some open issues ought to be answered. Waste is continuously generated and so the question arises: When will the next conditioning be done and applying which method?

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 Hot Laboratory (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 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 und used 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 (LMA) from the HOTLAB (material under IAEA-control) the existing conditioning technique in a specialized package (compacted 20-Liter-drums in a bottom closed steel cylinder are 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:

Technical construction of a waste package:

- The waste is packed and documented in the HOTLAB in 20-Liter-drums (5.3 gallons).
- After detailed planning and controlling, the packaged is moved to the Section for Dismantling and Waste Management and compacted by a 120 t press in a steel-press-matrix into a steel-cylinder.
- Afterwards the steel-cylinder, including the pressed waste, is cemented in a 200-Liter-corrosion-resistant-drum (53 gallons) made of stainless-steel.
- The waste has to be covered by 10 cm of concrete to comply with the standard requirements of the final depositor (Nagra).

A technical report, called “specification”, describes the improved conditioning process and the waste package and is a kind of “licensing documentation”. 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 requirements of a deep geological repository. However, although a licensed method by the competent authority is available, conditioning allowed either as a “Type Approval” or a “Type Test”, modifications could happen or new tasks arise.

THE REGULATORY FRAMEWORK

Each conditioning process needs approval from the regulatory authority prior its application. The work with the licensing documentation, the specification, started in 2008. The conformity of the waste package, which has been developed to the requirements of a future deep geological repository (i.e. corrosion resistant steel drums), 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.

As from 2009 the ENSI is the Swiss regulatory body (until 2008 HSK) 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 for conditioning [1]
- HSK-G-04 for interim storage [4]

The conditioning process must be approved by the ENSI before getting into routine operation (Type Approval). This proofing is in progress since 2011, as a “TypeTest”.

Further there are the IAEA regulations about unclear fuel to be considered. Only part of the material is nuclear material and therefore only a part of the waste is under Safeguards.

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

In Switzerland the HOTLAB of PSI is the only laboratory authorized by the safety authorities to handle large quantities of radioactive materials including commercial as well as experimental nuclear fuel, supervised by IAEA. This is the origin of the Pu-containing waste.

In the HOTLAB the Division Hotlaboratory (AHL – operator of the HOTLAB) and Laboratory for Nuclear Materials (LNM) are the principal research units 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.

These inevitably produce special radioactive waste which has to be treated, stored, documented and disposed of. The conditioning processes are part of an accredited waste management system of the institute. The waste management is performed by the Section “Dismantling and Waste Management”. Proved and accepted methods need to be developed for safe conditioning and storage. But, if you have a licensed method by the competent authority, modifications could happen, as described here.

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 conditioned in 2013 are approximately in average:

- 15% plastic/PVC/PE,
- 11% glass,
- 5% rubber,
- 43% steel,
- 5% ceramic,
- 10% cellulose and cellulose-like
- 10% others (aluminum, lead,copper etc.)
- 1% of fuel (including U-nat).

Two different sources of Pu-containing waste exist in the PSI-HOTLAB. Both types are collected into 20-Liter-drums (6 to 13 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 waste from the hot cells (irradiated material).

Material under IAEA-control

For the material under IAEA-control, the main declaration is resulting from HOTLAB´s strictly controlled documentation (to an accuracy of 0.1 g in reporting). 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.

Alpha- and fission product-contaminated waste

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-drums are stored in shielded areas in the HOTLAB (s-unit) and transferred batch wise to the Section RBE prior the final conditioning treatment. Some drums have high dose rates (5-10 mSv/h contact) which has to be manually handled. The handling and the sequence of conditioning is planned in advance to reduce the accumulated dose. In the past the handling and temporarily storage was no problem. In the last campaign the administrative classification of the waste and the location has been identified as an open issue. The regulatory requirements of the IAEA has to be clarified.

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-waste-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. If the amount of waste-drums is high, these are placed in the right sequence for compacting on the RBE-area.

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 contents of the final package within 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.

Furthermore the IAEA has to be informed in advance because of the special status of parts of the waste (fuel). There are nowadays open issues to be clarified, concerning the status of the waste package and the involved fuel.

CONSTRUCTION 200-LITER PACKAGING

A 200-Liter-drum made of stainless steel is used. The bottom and the inner side of the drum are prepared with a 10 cm (0.33 ft) grout cover.

A 10 cm cement cover around the waste is fulfilling the requirements of the standard models of a thermal attack of the final repository.

An end closed steel cylinder (0.36 m diameter) filled with the compacted waste is placed in the 200-Liter-drum. The steel-cylinder is surrounded by a special grout (10 cm = 0.33 feet). The cylinder is working as a press matrix and a handling-tool for the 20-Liter waste drums.

As a result, there is approximately 60 Liters (15 gallons) free volume for the pressed waste in the cylinder.

FINAL CONDITIONING

In a walk in cell, the delivered 20-Liter-waste-drums are pressed to pellets in a steel-cylinder, stacked in a new special constructed steel matrix (see Figure 1). For dismantling purpose the press matrix is divisible. Approximately 6 to 13 drums are pressed in each steel cylinder, with a weight of about 80 kg ± 35 kg.

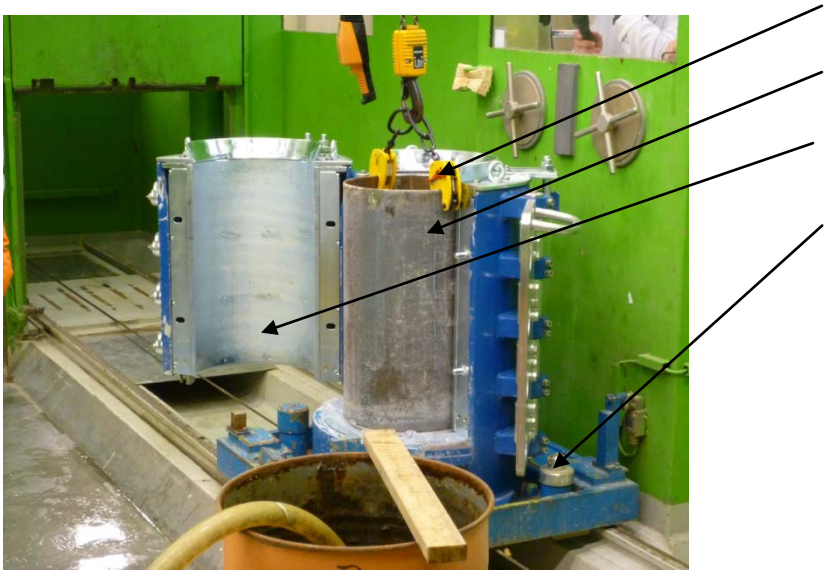

Fig. 1. Construction of the new fabricated press matrix

Fig. 2. Filled 200-l-drum

After this, the remaining space between steel cylinder and pellets is filled with a special flowable grout (“PSI-Vergussmörtel”). After that, the cylinder is carefully placed in the prepared 200-Liter-drum and the remaining empty space is again filled with a special flowable grout (see Fig. 2).

The filled grout has to harden in 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, required 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 again 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 repository is in operation, 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 with the evolving safety regulation especially 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 requirements on this study, which must be answered again and again. This takes time. Actually the storage in the Interim Storage of this special waste is accepted by the supervisor, because an external study has excluded in the Interim Storage the risk of criticality.

The whole practical and documentation 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.

In 2013, a total of 13 waste packages were produced formally as a Type Test. 130 waste drums (20-Liter) were successfully compacted in the cylinder and embedded in mortar in a 200-Liter-waste package.

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-Liter-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. Of course, the produced waste packages are reduced because of the stronger press.

One lesson learned in 2013 is to do with the in-drum compaction process. It was found that if the full force of the press was used to produce maximum compression of waste in the upgraded press, unless the bottom of the drum is precisely planar, the compaction process can produce cracks on the grout filled bottom of the drum. If the bottom of the 200-Liter-drum isn’t perfectly manufactured, cracks can`t be ruled out on the grout filled bottom. To use the full force, a separately steel-cylinder is used in a special constructed inner press and afterwards placed in the 200-Liter-drum. At least it is filled with grout.

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. This was successfully practiced in 2013.

- 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 cylinder. This was also successfully practiced in 2013.

Now 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 Uranium-, Plutonium- and Americium-contaminated waste are unified to one method. The produced and in advance calculated waste packages were done successfully, but it took its time.

The strict declaration of the IAEA-inventory for fissile material is still guaranteed. An open issue is still the exact IAEA-definition. Clarifying is still in progress.

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 (ADR) [8] 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
- No longer in-drum-compaction of the waste is performed. After compaction waste in a steel cylinder these are cemented in a 200-Liter-drum. The waste is covered by 10 cm of concrete, to shield the raw waste containing a significant amount of organics in case of fire.
- 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 2013 [8] respecting the “sum-rule”

With these radiological limitations of this special Pu-type, there is a production rate of approximately 6 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 and dried in the HOTLAB 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 cylinder to a steel cylinder to withstand the required forces.
- No longer in-drum-pressing of the waste is performed.

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

- The gas production in the final repository 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

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 requests 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). This issue is still open.

Furthermore the alpha-activity of the produced waste packages overcome the limit used for the still valid safety case study of the Federal Interim Storage. The new calculation for the accident case study is done as well as the criticality study under accident conditions of the Interim Storage. The new study has to be approved by the authority ENSI. This is still in progress.

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). The realization of the new safety case study is planned. The licensing documentation will be finished at the end of 2015.

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.

- 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 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 cylinder to a steel cylinder to withstand the required forces
- To meet the requirements of the final repository the 200-l-drums are made of stainless steel.
- No longer in-drum-pressing of the waste is performed.
- The successfully conditioned 13 waste packages are within the limits (activity and material).

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. Administratively the grout fulfils the

requirement of the guideline B05 (conditioning of radioactive waste) of the Swiss Federal Nuclear Safety Inspectorate (ENSI).

The method, especially the waste package, is accepted by the final disposer (NAGRA). The approval by the authority (ENSI) is ongoing. 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 (Type test) and prove the new procedure in 2012/2013. This has been successfully done. In 2013, a total of 13 waste packages are produced. But there are still old and new open issues to be handled:

- Shipment in the future (find a package B(M)F)
- Interim Storage (until accident case study is accepted); criticality under accident condition is excluded
- Further conditioning campaigns (qualifying the Waste Laboratory/new accident case study)
- Requirements and status of waste packages (IAEA-control)

It was planned to repeat the conditioning campaign every second year, to restrict the amount of waste to be handled. This will imply a formal Type Approval by the authority or an extended approval for a Type Test.

Inevitably waste is produced and so the question is again: when will be the next conditioning done and applying which method?

REFERENCES

1. ENSI guideline HSK-B05: Anforderungen an die Konditionierung radioaktiver Abfälle (requirements to the conditioning of radioaktive waste), 1. February 2007 http://www.ensi.ch/de/document/richtlinie-hsk-b05-deutsch/
2. Nuclear Energy Ordinance (KEG), 21. March 2003 http://www.admin.ch/ch/d/sr/c732_1.html
3. Radiation Protection Ordinance (StSG), 22. March 1991 http://www.admin.ch/ch/d/sr/c814_50.html
4. ENSI guideline G-04: ENSI guideline G-04: „Auslegung und Betrieb von Lagern für radioaktive Abfälle und abgebrannte Brennelemente", 1. March 2012 (requirements to the storage of radioactive waste) http://www.ensi.ch/de/document/richtlinie-ensi-g04-deutsch/
5. ISO/IEC 17020:2012 Accredited quality management system - requirements
6. EN/ISO 9001:2008 Quality management systems - requirements
7. J. Müth, P. Schleuniger, U. Ehrlicher, E. Hartmann, H.-F. Beer: Spezifikation des Abfallgebindetyps 4 zur Konditionierung von plutonium-haltigen/-kontaminierten Abfällen", TM-90-09-05 Rev. 3, Paul Scherrer Institut (2013)
8. ADR 2013: Accord européen relatif au transport international des marchandises Dangereuses par Route http://www.astra.admin.ch/themen/schwerverkehr/00246/00406/05800/index.html?lang=de it is conform to the UN Recommendations on the Transport of Dangerous Goods, Revision 17 (2013) http://www.unece.org/trans/danger/publi/unrec/rev17/17files_e.html

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