Handling Radioactive Waste from the Proton Accelerator Facility at the Paul Scherrer Institut (PSI) - Always Surprising? – 13320

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ABSTRACT

The Paul Scherrer Institut (PSI) is the largest national research centre in Switzerland. Its multidisciplinary research is dedicated to a wide field in natural science and technology as well as particle physics. In this context, PSI is operating, amongst others, a large proton accelerator facility since more than 30 years. In two cyclotrons, protons are accelerated to high speeds and then guided along roughly 100 m of beam line to three different target stations to produce secondary particles like mesons and neutrons for experiments and a separately beam line for UCN. The protons induce spallation processes in the target materials, and also at other beam loss points along the way, with emission of protons, neutrons, hydrogen, tritium, helium, heavier fragments and fission processes. In particular the produced neutrons, due to their large penetration depth, will then interact also with the surrounding materials. These interactions of radiation with matter lead to activation and partly to contamination of machine components and the surrounding infrastructures. Maintenance, operation and decommissioning of installations generate inevitably substantial amounts of radioactive operational and dismantling waste like targets, magnets, collimators, shielding (concrete, steel) and of course secondary waste.

To achieve an optimal waste management strategy for interim storage or final disposal, radioactive waste has to be characterized, sorted and treated. This strategy is based on radiation protection demands, raw waste properties (size, material, etc.), and requirements to reduce the volume of waste, mainly for legal and economical reasons. In addition, the radiological limitations for transportation of the waste packages to a future disposal site have to be taken into account, as well as special regulatory demands.

The characterization is a task of the waste producer. The conditioning processes and quality checks for radioactive waste packages are part of an accredited waste management process of PSI, especially of the Section Dismantling and Waste Management. Strictly proven and accepted methods needed to be developed and enhanced for safe treatment, transport, conditioning and storage. But in the field of waste from research activities, individual and new solutions have to be found in an increasingly growing administrative environment. Furthermore, a wide variety of components, with a really large inventory of radioactive nuclides, has to be handled. And there are always surprising challenges concerning the unusual materials or the nuclide inventory.

In case of the operational and dismantling radioactive accelerator waste, the existing conditioning methods are in the process of a continuous enhancement – technically and administratively.

The existing authorized specifications of conditioning processes have to be extended to optimize and fully describe the treatment of the inevitably occurring radioactive waste from the accelerator facility. Additional challenges are the changes with time concerning the legal and regulatory requirements - or do we have to consider it as business as usual?

This paper gives an overview of the current practices in radioactive waste management and decommissioning of the existing operational accelerator waste.

Introduction

The Paul Scherrer Institut (PSI) is operating on-site, amongst others, a large proton accelerator facility since more than 30 years. In the PSI facility, hydrogen is ionized and the resulting protons are accelerated to 72 MeV for injection into a ring cyclotron where they are accelerated to 590 MeV. The beam intensity has increased from some few hundred µA in the beginning, to 2.2 mA today and in the future may be raised to 3 mA. Primary beam protons hit the targets and beam dumps where they cause nuclear reactions, including spallation, producing secondary protons and neutrons and lighter nuclei. The heart of the accelerator research at PSI is located in the Experimental Hall.

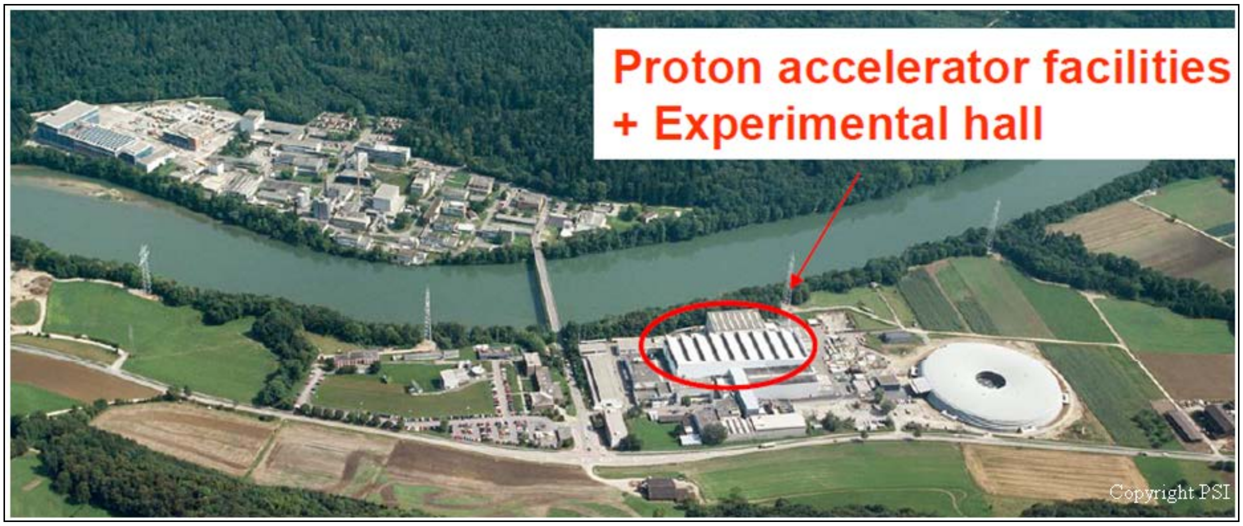


Fig. 1: Accelerator facility at Paul Scherrer Institut (Switzerland)

An unfortunate side effect of research with accelerators is, that the interaction of particle beams with matter might lead to the activation of accelerator components, beam line structures and their surroundings. This is comparable to nuclear research facilities. The induced radioactivity is either due to direct interactions (spallation) of the primary beam or to secondary particles (mainly neutrons) produced in a multitude of nuclear processes. For the radioactive waste we have to prepare technical solutions which are accepted by the supervising authority.

Ideally, we have for all appearing waste streams a technical report, called “ specification”. This document describes the waste packaging, conditioning process, the waste package, interim storage and the ability of transportation as well as the suitability for final disposal – a certificate from the Cooperative for the Disposal of Radioactive Waste (NAGRA). The conformity of the specification with the regulatory guidelines [1] is investigated in advance by the Swiss Federal Nuclear Safety Inspectorate (ENSI). At the end of the licensing process, the ENSI gives a type approval.

Radioactive waste management

Most of the radioactive material produced in accelerators consists of solid bulk components – often of metallic nature. Especially in case of the Swiss deep geological repository, large amounts of steel are a problem. The repository should be located in a clay formation. This formation is sensitive to gas production which arises by the corrosion of steel.

The main difference between waste produced in nuclear power plants and in accelerators are
- in general, no or little long-lived alpha activity (only for special target materials, e.g. Pb in SINQ target, LBE in Megapie and from U, Th- impurities)
- no fuel or criticality
- less contamination (except for special targets and activation of dust),
- less neutron-rich radioactive nuclides,
- but a large number of radionuclides and special nuclides from spallation (e.g., Fe-60).

Since the described approach in 2001 in [2] a lot of work has been done. The main waste streams are described in specifications [3], [4]. But the next task is, to keep the specifications up to date, especially the increasing demands of declaration. It is foreseeable, that described processes are to be revised or extended in the future. This will be a continuous process until the promised final repository is closed.

A further management problem is the transfer of all in the specifications written procedures to the workers, who are doing the practical work as it is performed.

It is important to stress that the minimisation of radioactive waste is best achieved with a proper study at the design stage of the facility.

Waste categorization by time of appearance

The waste produced at an accelerator facility is mainly solid. In accordance with Swiss legislation, the waste has to be solidified and conditioned to await final disposal. Mostly, the waste is embedded in cement.

The main waste streams generated by an accelerator could be categorized by the time of appearance. It is generated

- during operation and maintenance: replacements, decontamination and improvements of the facility and
- at the end of the lifetime, with the dismantling. This means that in addition to the operational waste, structural waste (mainly large amounts of concrete and shielding made of steel) has to be conditioned.

The amount of generated waste depends not only on the conditioning process itself and the packaging. It also depends on changes taking place in the research environment and the legal requirements (clearance level, needs of the operator of the final repository (NAGRA)etc.), too.

At PSI, the operational accelerator waste streams include in general:

- burnable waste (secondary waste, cleaning material, plastic, etc.) which is mixed with other waste to be treated at an incineration facility; this treatment is described in a technical report "Specification" by the operator of the facility (Zwischenlager Würenlingen AG, ZWILAG), a joint effort of the Swiss nuclear power plants, and has a type approval of the authority (ENSI). This kind of waste could be significantly lowered during the last years by the implementation of a free release measurement unit (measurement box additional to the manual measurements).

- waste from dismantling of components, machines and installations and from maintenance: solid bulk waste, mainly steel, different metals and consumable materials from installed components. The radioactive inventory depends on the location and can usually be calculated to an acceptable accuracy. This waste stream is described in specifications [3]; [4]. They involve also the main highly activated components like spallation targets, beam dumps and collimators. Only the principal occurring material types can be explicitly specified; for the presence of other, more exotic, materials, a total upper limit is given. This requires some attention to detail during the filling of the waste containers. The conditioning and waste management follows the normal procedures.

- dismantling of infrastructure: shielding and installed components; mainly concrete and steel. At the present time, this material should be conditioned in 4.5 m3 containers similar to the dismantling of research reactors and the established techniques described before.

Further more there is to consider
- "special waste**"** like special materials from targets and installations, electronic devices (e.g., aluminium, which gives rise to problems with the conditioning in a cement matrix), not common waste (dimensions, toxicity, material, nuclide inventory, activity etc.).

This waste has to be identified and described. The challenge is, to find out the best method for minimization and conditioning. This will be a never ending story until the research is at an end and the facility is decommissioned.

Special waste involves always “new” waste from research activities. This field of waste management is still a technical and administrative challenge: You have to be aware of upcoming problems in the future. This could be the use of problematical or complex materials (or mixtures) or changes in the requirements or updated safety models leading to inducing increased safety manners with complex consequences. Difficult waste should be recognized as early as possible to avoid a situation of unsolved problems at a later stage. For example: missing space and facilities for the storage and decomposition of large activated components (e.g., accelerator cavities) or for the treatment of large quantities of irradiated mercury (e.g. at SNS in Oak Ridge), etc. The lengthy approval procedures for such facilities must also be taken into account.

One also has to consider the formalities needed to establish new processes. From the point of view of the Swiss supervising authority, any waste which has no established conditioning method (described in a licensed specification), has to be formally declared as a “Grosskomponente” (“large component” [6]). This does not refer to the dimensions. It means a technically unsolved problem regarding the final conditioning of the waste, or, a waste stream which is not formally accepted by the authority (no licensed specification).

An example for good practice and the resulting administrative work: You have a high-dose-rate component (e.g. key nuclide is Co-60) which you don’t want to handle because of minimization demands regarding the accumulated dose of the operator. This high-dose-rate component can be shielded with low-activated material (e.g. key nuclide is again Co-60) in a concrete container. After some years, the low-activated shielding will be ready for free release and the high-dose-rate component can be handled. Now you can separate the waste, fill the container properly with waste and do the final conditioning. This process is not licensed and is formally treated as a “large component”. A separate description of the disposal process, including safety case considerations, etc. is required.

Waste categorization by location and activation; an accelerator facility from the perspective of waste management

An accelerator facility has several places of main activation with different characteristics. This means that waste streams can be categorised by their location or place of operation in the beam. For activation calculations, as established at PSI and described in [5], one needs in addition knowledge of the beam current, the time of operation, the material composition including impurities and a measured dose rate of the component.

The main parts of the PSI accelerator facilities are listed below and the different areas categorised according to their activation characteristics are illustrated in Fig. 2. The directly irradiated components are located mostly in the spallation neutron source SINQ and in the beam line with the meson production targets M and E and the beam dump. Secondary particle activation takes place close to these areas. Significant activation by thermal neutrons will be in the neutron beam lines of SINQ.

The PSI proton accelerator facilities:

72 MeV:	e.g. isotope production (Injector)
590 MeV / 2 mA:	muon and pion production spallation-neutron source (SINQ) ultra cold neutrons (UCN)
70- 250 MeV:	medical accelerator (COMET) – not yet categorized (see [7])

In addition, PSI operates an electron accelerator which produces negligible amounts of radioactive waste:

2.4 GeV:	Synchrotron-Light-Source SLS

The neutrons and mesons used in experiments at PSI are generated by collision of a beam of very fast protons with a special target made
– of lead in the case of the SINQ neutron sources and UCN and
- of carbon/graphite in the case of the meson sources.
The protons have been accelerated to a speed of approximately 80 % of the speed of light by PSI's proton accelerator facility. This facility came into operation in 1974 and was originally used for experiments in the area of elementary particle physics. Nowadays, it generates the most intense beam of protons (2.2 mA) in the world. It is planned to increase the beam current further, which will change the expected amount of activation.

Generated neutrons in the SINQ neutron spallation source are used to determine the arrangement or motion of atoms in materials. Because neutrons are particles that behave like very tiny magnets, they are also especially suited to the investigation of magnetic materials.

Generated mesons (pions and muons) are mainly used for investigations of magnetic fields within materials. They are elementary particles with properties very similar to those of electrons but being much heavier and unstable. When a muon decays inside a material, it carries information about the magnetic field in a sample.

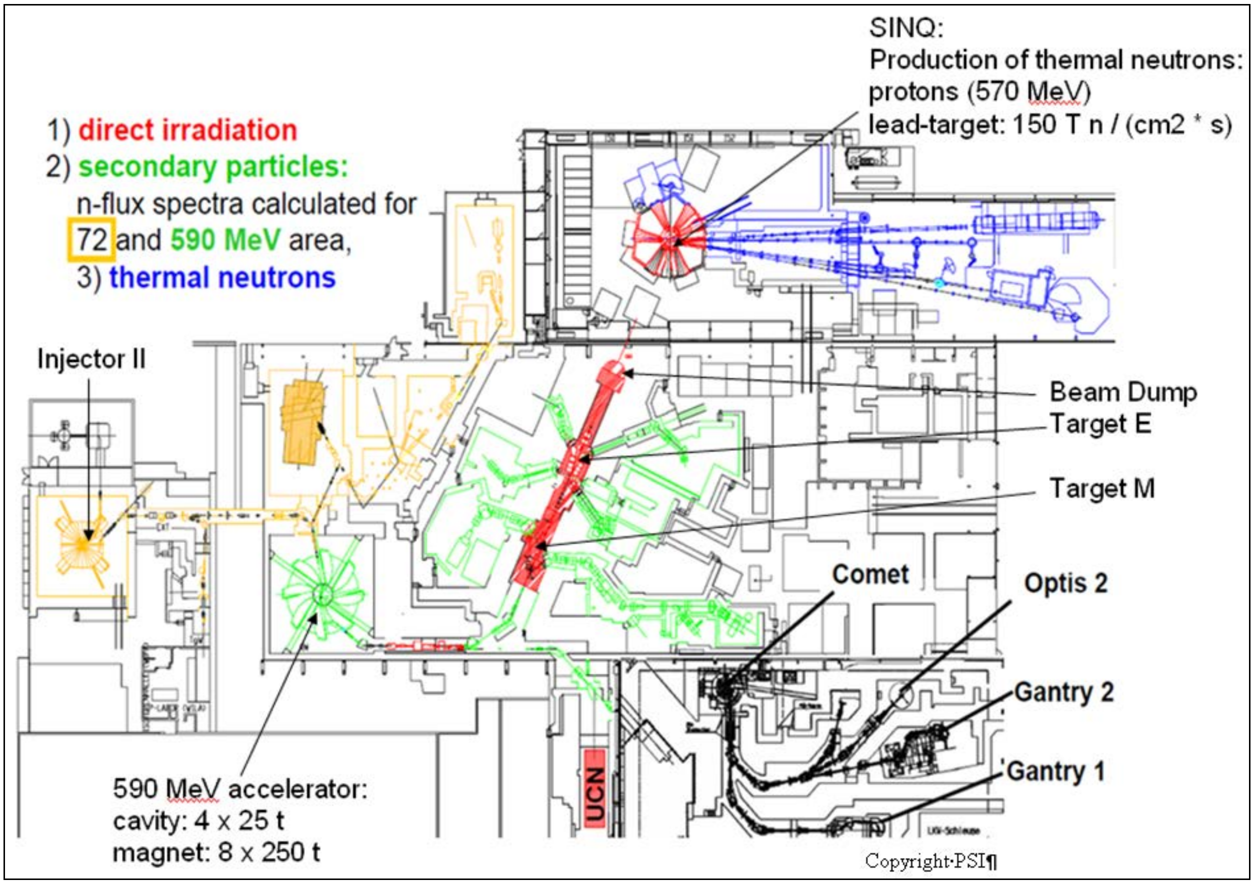


Fig. 2: The view of a waste manager: activated areas in the accelerator facility [5]; Copyright PSI

At the end of their operational lifetime, depending on the irradiation conditions, more or less all of the radioactive components from the marked location (see Fig. 2) must be treated as radioactive waste.

Physical background of the activation and involved materials

<u>Direct irradiation at major beam loss areas:</u> High energy protons (590 and 72 MeV) induce spallation reactions with emission of p, n, H-2, H-3, He-4 and heavier fragments and fission processes, leading to localized areas of very high activation (up to a few hundred Sv/h). Affected components are mostly:

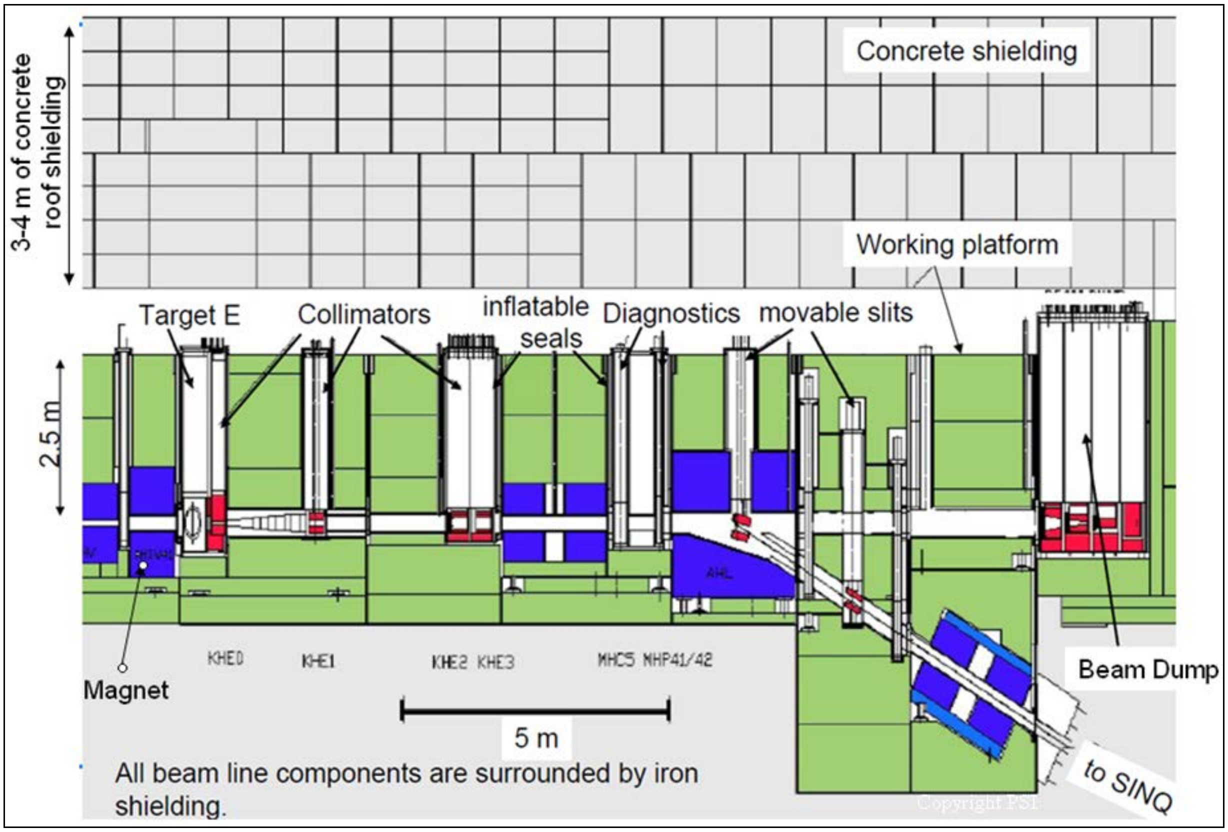
- targets (graphite, lead, zircalloy)
- collimators (copper)
- beam dumps (copper)

<u>Secondary particles fields:</u> Mostly neutrons remain after penetration of shielding material. These are responsible for neutron-activated materials in the surrounding areas of direct beam loss areas. This affects mainly components made of steel and concrete, for example the shielding.

Important examples for neutron capture at thermal energies are Co-59 (n,g) Co-60 (important reaction in steel) or Ag-107 (n,g) Ag-108m (important impurity in copper).
The resulting dose rates after decay of the short lived nuclides lie approximately between micro and milli Sv/h.

Affected components (see also following Fig. 3 of a cross section of the proton beam line):

- vacuum chambers (corrosion free steel) and components
- beam diagnostic monitors
- magnets, collimators (steel, copper)
- degrader, targets (graphite)
- target (lead)
- accelerator cavities (aluminium, copper)
- shielding (steel, concrete)
- electrical installation (plastic)



All beam line components are surrounded by iron shielding.

Fig. 3: Cross section of a part of the beam line (target E to beam dump); [5]

Typical materials that can be found at particle accelerator facilities are:

- iron and zinc: especially for magnets and cable trays
- copper: used for collimators, beam dumps, magnet coils and for electric cables,
- normal and stainless steel: used for beam tubes, vacuum chambers, supports, pipes for water-cooling systems and machine components, shielding etc.
- aluminium: for power cables and pipes, vacuum chambers, cavities, target hulls, components
- plastics and resins: used as insulator of electric cables (PVC)
- graphite: degrader, targets
- concrete: used for walls and as biological shielding from radiation
- lead: used as shielding material, target material

Waste originating from the components listed above is, therefore, an unavoidable consequence of the operation of accelerators. One characteristic of accelerator waste is the presence of a large number of radionuclides. For a proper declaration one has to have an established calculation procedure [5].

Separation of waste - volume economy

For minimizing purpose the waste is divided into categories:

- waste , which will not decay within 30 years below the currently accepted level of exemption of 0.1 mSv/h [9]. This waste is conditioned.

- weakly activated waste, which is expected to decay within 30 years to below the currently accepted level of exemption. This waste is stored on site. Obviously, this material has to be measured before any future exemption.

- short-lived radioactive substances with half-lives measured less than 60 days need be stored for only relatively short periods to decay to insignificant values.

- waste already below the current exemption limit [9]. This material is checked and released. A significant waste reduction of secondary waste by free release measurement has been established in the last years.

Characterization and waste declaration

In the case of accelerator waste, the nuclear characterization is very important. This is mostly done by calculation [5] and verification measurements. For comparison: in nuclear power plants, the fingerprint method is currently used , where a predefined radionuclide inventory is scaled to the measurable radionuclides to obtain the final radionuclide inventory and activity.

To ensure an appropriate calculation, the documentation of some facts (if available) is very important:

- operation cycles
- location of the parts in the facility
- construction plan
- energy of the beam at loss point
- amount of beam lost at loss point
- distance to beam loss points
- function/task of the Material (degrader, magnet, shielding, collimator etc.)
- Material composition and impurities
- dose rate at defined points

These records are included in an application for disposal of radioactive components for PSI-internal quality assurance.

The standardized documentation of the waste includes

- materials
- nuclides (qualitative, quantitative, mass specific)
- construction/dimensions
- special materials or toxic materials (such as lead, aluminum, graphite, etc.)
- hazardous components: corrosive, flammable materials (metallic Uranium targets, etc.)
- etc.

Conditioning

The ordinary waste could be embedded in concrete in steel drums. The current techniques used for waste disposal involve the conditioning of waste in concrete containers [3] with radioactive waste immobilized in cement. For this purpose, a system of concrete containers (4.5 m^3) has been developed; in addition to the existing conditioning in drums. The conditioning is done by embedding in concrete. The container fulfils the industry package demands [10] and is accepted for final disposal. Large components have to be disassembled in a hot cell.

During the disassembling, activated aluminium is removed for separate conditioning because of the hydrogen production caused by the reaction of aluminum with strongly basic mortar. A melting procedure was developed to reduce the problem to a level accepted by the operator of a final repository (NAGRA). This process was established at PSI originally for the dismantling of a research reactor, described in [8]. Today, the dismantling of this reactor is coming to an end and the further operation of the melting facility, especially the licensing, is questioned by the authority. The established procedure for the conditioning of aluminum has to be formally clarified again.

Special treatment of targets, highly activated beam dumps, collimators etc. is described in [4] and currently in the licensing process for type approval by the competent authority ENSI.

An example: special waste, concerning the size and the material

An example of minimization and handling of special waste are the aluminum cavities of the 590 MeV accelerator (see Fig. 4). The standard conditioning method at PSI is the embedding in cement. This is not possible, for the reason of the gas producing reaction of aluminum in cement. First we have to minimize the waste. This is done by cutting the cavity and dividing it into the inner, highly activated part (remaining waste which has to be melted and conditioned) and the rest which is stored until it is decayed.

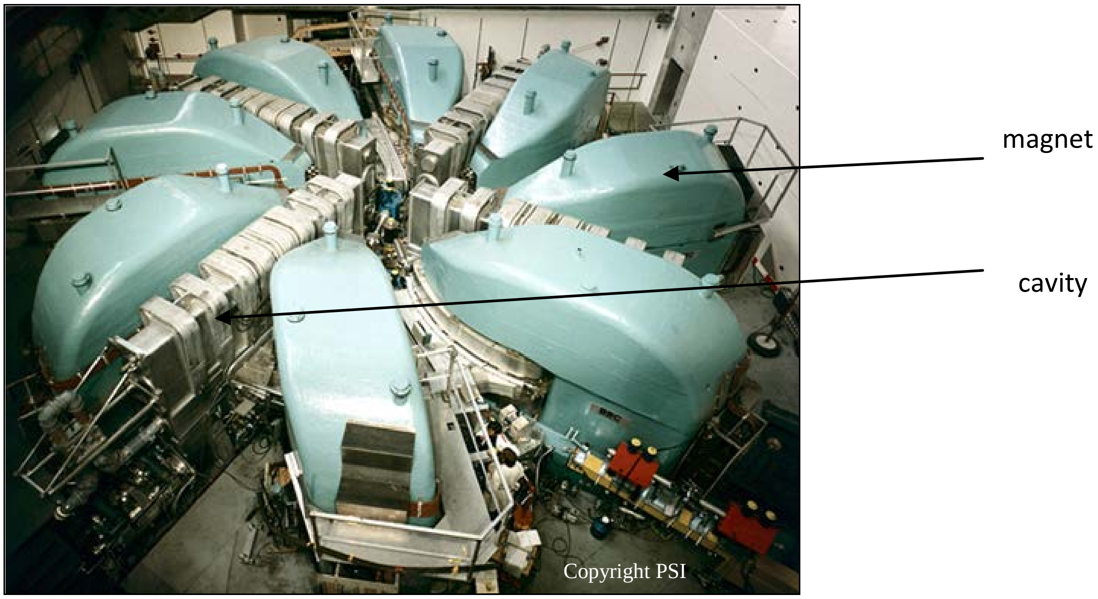


Fig. 4: 590 MeV accelerator with cavities and magnets

Conclusion

At PSI, radioactive waste from different origins has to be managed. This involves free release measurements and sorting for decaying and a intelligent packaging concept to minimize radioactive waste. In general, there are established waste streams for the main materials, components and machines, declared as radioactive waste. In this context, a process has been established to manage accelerator waste routinely on a practical and regulatory level. For the main waste streams the acceptance of the authorities (Swiss Nuclear Safety Inspectorate) and the final disposal agency (NAGRA) has been given. This is business as usual. Surprises arise from the discovery of unexpected waste items or because of the fact that an accelerator is a still developing research facility: so we still will have technical challenges with new problems, arising by the use of special materials or large and huge components. Another expected change is the waste volume: minimization will create in future a higher concentrated activity per mass or volume and the produced waste packages. This means that administrative changes have to be implemented in already predetermined upper activity limits and average activities of licensed waste packages.

But as long as there is no final repository in operation, waste management means acting in a dynamic system: with time, technical and administrative changes will unavoidably occur.. This could involve

- changes in the regulatory demands,
- upgrades in the facility (e.g. materials, higher beam current),
- changes in clearance levels, demands of the final repository, regulation etc. as well as
- changes in the demands of minimizing of radioactive waste or
- the opportunity (hopefully?) to do the final conditioning as late as possible to remain flexible for the coming requirements (upgraded embedding matrices, documentation demands, new waste minimization concepts etc.) in the future.

Every time when changes take place, we have to qualify the described processes again. As well as we have to prove the impact on the already produced waste packages. From the point of view of radiation protection, one has to be very careful with the next step, the repackaging of conditioned waste and the justification of such a work.

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