Research for the safe management of nuclear waste

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Decommissioning of nuclear facilities

- In Germany …
  - about 300,000 m³ LILW up to 2080
  - repository Konrad for low- and intermediate level waste will start operation in the next decade

- … and abroad
  - EU: about 40 NPP will reach their envisaged lifetime at about 2025 and …
  - about 300 NPP world-wide by 2030

- Moreover:
  - decommissioning/dismantling of research reactors, research installations, reprocessing facilities, …
Geological Disposal seems to be the best option

OECD NEA RWMC collective statement, 2008
• There is overwhelming scientific consensus worldwide that geological disposal is technically feasible

30+ years of R&D needed for the first license applications in Europe
• Sweden 76: March 2011 (35 yrs) operation 2029 (53 yrs)
• Finland 78: Dec. 2012 (34 yrs) operation 2022 (44 yrs)
• France 70's: 2015-2017 (35-40 yrs) operation 2025 (ca.50 yrs)

C.Davis, EC
Licence granted for Finnish used fuel repository

12 November 2015

The Finnish government has today granted a construction licence to waste management specialist Posiva for a used nuclear fuel encapsulation plant and final disposal facility at Olkiluoto.

The Olkiluoto repository will consist of a network of disposal tunnels up to 450 metres below ground (Image: Posiva)

The government included several conditions in the construction licence. When applying for an operating licence, Posiva is required to submit analyses concerning the environmental impacts of the facility, the retrievability of the used fuel, the transport risks involved and any changes that may have been introduced in the project. A maximum of 6500 tonnes of uranium may be disposed of in the facility.

The used fuel will be packed inside copper canisters at the encapsulation plant, from where they will be transferred into the underground tunnels of the disposal facility, located at a depth of 400-450 metres, and further into the deposition holes lined with bentonite buffer.

The site for Posiva's repository at Eurajoki near Olkiluoto was selected in 2000. The Finnish parliament approved the decision-in-principle on the repository project the following year.
Swedish regulator approves repository application

29 June 2016

The licence application by Sweden’s radioactive waste management company for an integrated system for the final disposal of used nuclear fuel and radioactive waste has been endorsed by the country’s Radiation Safety Authority (SSM). A final decision to licence the facilities will be made next year.

Svensk Kärnbranslehantering AB (SKB) submitted applications to build the country’s first repository for used nuclear fuel, together with a plant to encapsulate the fuel prior to disposal, to the SSM in March 2011. The integrated facility - the encapsulation plant and the Clab interim storage facility - is referred to in SKB’s application as Clink. SKB has since made both clarifications and additions to the applications. The company has also submitted an application to extend the storage capacity of the Clab facility from the current 8000 tonnes of fuel to 11,000 tonnes.

The applications are being reviewed by the SSM and the Land and Environment Court in Stockholm. The SSM is considering questions of nuclear safety and radiobion at the facilities as laid down in the country’s Nuclear Activities Act. The review undertaken by the Land and Environment Court is based on the Environment Code.

An artist's impression of the planned repository concept, with its 66-kilometre network of underground tunnels (Image: SKB)

WNA Links
- Nuclear Power in Sweden
- Radioactive Waste Management

Related Links
- Svensk Kärnbranslehantering AB (SKB)
- Swedish Radioactive Safety
Radioactive wastes

- Radioactive wastes:
  - generated in all stages of the nuclear fuel cycle, but also in various industrial activities (incl. NORM), research and medicine

- Wastes in the nuclear fuel cycle:
  - uranium mining & processing: mine tailings, waste rocks, …
  - enrichment & fuel fabrication: depleted uranium (if considered as waste), …
  - nuclear energy generation: spent nuclear fuels containing fission products (e.g. $^{137}\text{Cs}$, $^{135}\text{Cs}$, $^{131}\text{I}$, $^{99}\text{Tc}$, $^{90}\text{Sr}$, $^{79}\text{Se}$, ...) and actinides (e.g. $^{239}\text{Pu}$, $^{239}\text{Np}$, $^{242}\text{Am}$, ...) due to neutron capture; activated structural materials; operational wastes (e.g. ion-exchange resins, wiping tissues, ...)
  - reprocessing: vitrified wastes, secondary wastes
  - decommissioning wastes: activated materials/metals, rubble, ...
Categories of nuclear wastes

- **Spent fuel (HLW)**: 5% of the total waste, 99% of the radioactivity. 
  - Final Disposal: 5% of the total waste, 99% of the radioactivity, \( \Delta T >> 3 \text{ K} \)

- **High-level waste (HLW)**: waste with negligible heat production.

- **Low-level waste (LLW)**: waste with negligible heat production.

- **Intermediate-level waste (ILW)**: 95% of the total waste, 1% of the radioactivity. 
  - Final Disposal: 95% of the total waste, 1% of the radioactivity, \( \Delta T < 3 \text{ K} \)

- **Heat producing waste**: HLW-glass and cladding material.
Nuclear waste in Germany

- non-heat-generating wastes: 304,000 m³ (L-/ILW) (until 2080)
- heat-generating wastes: 28,100 m³ (HLW)
- used nuclear fuel (until 2022): 17,220 t<sub>HM</sub>
  - reprocessed: 6,670 t<sub>HM</sub>
  - LWR-fuel for direct disposal: 10,550 t<sub>HM</sub>
- HLW-glass (CSD-V): 670 m³
- MAW-glass (CSD-B): 25 m³
- compacted waste (CSD-C): 740 m³
- other wastes (e.g. HTRSF, RRSF): 5,710 m³

[source: BfS 2015]
Safety principles of geological disposal

- General protection objectives:
  - protection of people and the environment against the dangers arising from ionizing radiation

- Approach:
  - isolation of waste from the biosphere and against inadvertent human intrusion
  - concentration and containment
  - (retardation)

-Containment of waste by “defense in depth” multi-barrier system

- Passive safety over geological time scales

- First priority: long-term safety
# Host rocks for geological repositories

<table>
<thead>
<tr>
<th>Crystalline rocks</th>
<th>Clays/Clay rocks</th>
<th>Salt</th>
</tr>
</thead>
<tbody>
<tr>
<td>+ mechanical stability</td>
<td>+ low hydraulic conductivity (diffusional transport)</td>
<td>+ low hydraulic conductivity</td>
</tr>
<tr>
<td>+ thermal conductivity</td>
<td>+ high radionuclide-retention capacity</td>
<td>+ plasticity (convergence)</td>
</tr>
<tr>
<td>• moderate radionuclide-retention capacity</td>
<td>+ self-sealing capacity</td>
<td>+ thermal conductivity / temperature resistance</td>
</tr>
<tr>
<td>− fractures</td>
<td>− low thermal resistance</td>
<td>− water soluble</td>
</tr>
<tr>
<td>− advective transport</td>
<td>− low mechanical stability</td>
<td>− low radionuclide-retention capacity</td>
</tr>
</tbody>
</table>

- **Sweden, Finland, Czech Republic, Germany**
- **France, Switzerland, Belgium, Germany**
- **USA (TRU), Poland, The Netherlands, Germany**
Safety requirements & criteria in Germany

- Safety principles
  - concentration and containment of radionuclides in the isolating rock zone (IRZ)
  - release of radioactive substances from the final repository only negligibly increases risks associated with natural radiation
  - no (planned) intervention or maintenance in the post-closure phase

- Comprehensive site specific safety analysis and safety assessment covering a period of one million years required

- Retrieval of waste must be possible during operational phase

- Manageability of containers for up to 500 years must be guaranteed for recovery
Potential regions for a HLW-repository in Germany

[based on BGR 2007]
Decommissioning & waste management

- Technologies for decommissioning and dismantling of nuclear facilities and the safe management of associated waste streams developed during recent decades
- Current R&D focussing especially on optimisation regarding, e.g. radiation exposure of personal, economic aspects, clearance, etc.
- However, some special (“problematic”) waste streams arising during decommissioning have not been in the focus of R&D activities in the past, i.e.
  - safe, efficient, and cost effective processing methods not readily available
  - technologies for the treatment of the waste types not yet established for routine waste management, and/or
  - waste-type failed to meet criteria for acceptance for available processing technologies or waste acceptance criteria for disposal
Problematic waste streams

- irradiated graphite (i-graphite)
- spent ion-exchange resins (SIER)
- radioactive toxic metals, e.g.
  - mercury
  - cadmium
  - beryllium
- contaminated NAPL and decontamination fluids
- asbestos & PCB containing wastes
- mixed waste containing chemotoxic and/or hazardous constituents
- (legacy wastes)

- often comparatively small waste streams
Irradiated reactor graphite (i-graphite)

Primarily graphite is used as a moderator

... but also as a structural component

- control rod channels
- coolant channels
- reflector bricks
- fuel compacts

i-graphite arisings:
- Germany: ~ 1000 Mg
- Worldwide: > 250,000 Mg

Variety of different types of reactor graphite with individual structure, composition and history
Irradiated reactor graphite: issues

- Complex structure of reactor graphite depending on the fabrication technology
- Individual impurity inventories result in unique inventory and distribution of APs
- Unique radiation-induced structure and surface properties / reactivities
- Unique RN release behavior in every irradiated graphite type affected by storage conditions

No standardized technical solutions for the safe management of irradiated graphite available worldwide

long-lived activation products (AP):

impurities\((n,X)\)APs (\(^{14}\)C, \(^{3}\)H, \(^{36}\)Cl etc.)
R&D issues for i-graphite

- Phenomenological investigations of i-graphite so far do not allow for an understanding of i-graphite behaviour during treatment & disposal
- Fundamental investigations of the properties of i-graphite required to develop safe waste management solutions

R&D themes:
- chemical speciation, binding sites and partitioning of radionuclides
- mechanisms of radionuclide release (e.g. $^{14}$C, $^3$H, $^{36}$Cl)
- theoretical understanding & modeling of i-graphite surface reactivity
- multi-scale modelling of graphite waste behaviour

R&D aims:
- Characterization and understanding of i-graphite properties
- Development of approaches for graphite waste minimization
- Understanding of i-graphite behaviour during storage and disposal
Radionuclide distribution in i-graphite

Distribution of APs “follows” their precursors – impurities

Inhomogeneous distribution of APs in irradiated graphite causes difficulties in an „up-scaling” of RN inventory from radioanalysis

TOF-SIMS depth profile of $^{35}$Cl in reactor graphite

Autoradiographic images of hot-spots in reactor graphite
Treatment: Crushing and fractionation (e.g. sieving) of graphite followed by characterization of structure and $^{14}$C distribution

Hypothesis: $^{14}$C is accumulated on the pore surface of the binder material

$^{14}$N(n,p)$^{14}$C (95%)

$^{13}$C(n,$\gamma$)$^{14}$C (≈5%)

$^{17}$O(n,$\alpha$)$^{14}$C (<0.01%)

Binder fraction

- 1-0.5 mm
- 500-250 μm
- 250-100 μm
- 100-50 μm
- <50 μm

Filler fraction

Crystalline filler particles (ca. 70%)

Porous pitch binder (ca. 30%)
Thermal treatment of i-graphite

Selectivity of treatment:

\[
\frac{wt(14C)}{wt(C_{tot})} > 1
\]

C\text{tot} mass loss, [%]

HT treatment in inert atmosphere improves selectivity of $^{14}$C separation
Future perspectives: use of model materials

- Lack of understanding of AP distribution and speciation
- No unique model on release behaviour of APs
- Unique structures & reactivities of every i-graphite type

Approach: use of model materials (HOPG, coke, binder) for separation of individual effects

Experiments with simplified model systems/materials to interpret and predict the behaviour of i-graphite
Management of mercury from nuclear facilities

- Various (historic) applications for mercury (Hg) in nuclear facilities
  - coolant for early experimental fast reactors
  - shielding in prototype reactors (e.g. PFR, DFR; UK)
  - target material in accelerators
  - window seals in hot cell facilities
  - catalyst in isotope separation and uranium metal dissolution

- Issues:
  - radiological characterisation
  - chemotoxic hazards / waste acceptance criteria
  - decontamination / re-use
  - immobilisation
Management of radioactive Hg @ FZJ

450 kg of contaminated Hg, collected during decommissioning of FZJ hot-cells

Unknown inventory (fission products, actinides, etc.)

Surface dose rate \( \leq 1 \text{mSv/h} \)

No authorization for mercury storage at FZJ interim storage

Disposal limit at KONRAD: 43 kg Hg

Up-coming project - PROcess of radioactive MErcury Treatment and Handling for Elimination Under Safety-Standards

PROMETHEUS – a joint BMBF-project on characterization, decontamination and clearance of radioactive mercury
Objectives & tasks of PROMETHEUS

PROMETHEUS (2016-2018):

- Radiological characterization ($\alpha, \beta, \gamma$) / nuclide vectors / scaling factors
  RN-speciation in contaminated mercury collected at FZJ

- Characterization of chemical composition of contaminated Hg (e.g. speciation, inactive additives, etc.)

- Optimization of existing decontamination techniques: vacuum distillation, Hg conversion, membrane extraction, electrochemical, etc.

- Immobilisation of radioactive residues after processing

- Development and optimisation of $\beta/\gamma$-counting technique for clearance of Hg
Management of irradiated beryllium

- Be (or BeO) employed as neutron reflector and moderator in research reactors
- Issues
  - activation during operation
  - contamination possible depending on cladding
  - highly chemotoxic
  - incompatible with cementitious waste forms

\[ ^9\text{Be}(n,\gamma)^{10}\text{Be} \rightarrow ^9\text{Be} \quad \beta^- \quad 1.38\text{e}+6 \text{ a} \]
\[ ^9\text{Be}(n,\alpha)^6\text{He} \rightarrow ^6\text{Li}(n,\alpha)^3\text{H} \quad \beta^- \quad 0.807 \text{ s} \]
\[ ^9\text{Be}(n,2n)^8\text{Be} \rightarrow ^{24}\text{He} \quad \beta^- \quad 7\text{e}-17 \text{ s} \]

Impurities (n,X)APs (\(^{14}\text{C}, ^{60}\text{Co}\))

<table>
<thead>
<tr>
<th>Institution</th>
<th>Amount of Be</th>
</tr>
</thead>
<tbody>
<tr>
<td>GKSS Geesthacht (FRG-1)</td>
<td>681.2 kg</td>
</tr>
<tr>
<td>HMI Berlin (BER-II)</td>
<td>1582.6 kg</td>
</tr>
<tr>
<td>TU Munich (FRM-I)</td>
<td>168.0 kg</td>
</tr>
<tr>
<td>HZDR Rossendorf (RFR)</td>
<td>329.0 kg</td>
</tr>
</tbody>
</table>

Disposal limit in KONRAD: 24.5 kg Be

➢ Safe management solutions for irradiated beryllium missing to date
Research perspectives for irradiated Be

- Development of management strategies for irradiated beryllium

- Radiological characterization of irradiated Be
- Nuclide vectors & scaling factors (as function of irradiation history and material composition)
- Evaluation of decontamination techniques aiming at clearance of decontaminated beryllium and reduction of waste volumes
- Development of encapsulation materials
- Material behaviour under disposal conditions
- Alternative applications for re-use of decontaminated low-active Be
- Identification of optimal conditions for storage and disposal
- Requirements for safe handling of irradiated Be
R&D for non-destructive analyses of wastes

- **Purpose:**
  - compliance of waste packages with specifications and acceptance criteria for interim storage and final disposal
  - inventories of radionuclides and chemotoxic elements
  - heterogeneity of waste packages
  - identification of shielding structures in waste packages
  - accurate and reliable characterisation of waste packages at industrial scale
  - characterisation of legacy wastes

- **Development/application of innovative passive and active non-destructive techniques assisted by modern computational simulation tools, e.g.**
  - Segmented Gamma Scanning (SGS)
  - Instrumental Neutron Activation Analysis
  - P&DGNAA
  - Fast Neutron Imaging
  - Digital Radiography (DR)
  - Transmission-Emission Computer Tomography
Segmented-Gamma-Scanning (SGS)

- Radiological characterization and quantification of gamma-emitting isotopes in 200-L waste drums
  - Collimated HPGe-Detector for gamma-ray measurement
  - 20 Segments ($\Delta h = 4$ cm) / 12 sectors ($\Delta \theta = 30^\circ$) – counting time 1 h per drum
  - Calibration based on homogeneous activity and density distribution
  - Radionuclide activity calculated from the sum-spectrum
  - Count rate distribution give qualitative information on waste heterogeneity (hot-spots)

- R&D:
  Improvement of reliability and accuracy of activity determination of radioactive waste drums with non-uniform isotope and matrix distribution including the presence of internal shielding structures
Non-radioactive substances in nuclear wastes

MEDINA:
Multi-Element Detection based on INstrumental Activation Analysis

- Assay of chemotoxic elements in 200-L waste drums

Development
- concept and set-up
- parameterization (neutron flux, detector efficiency)
- influence of activity (Cs-137 and Co-60)
- algorithms for quantification
- validation
- analysis time 1 to 4 hours
- accuracy: 7±4 % (homog.); 14±7 % (inhomog.)

R&D:
Improved identification and quantification of chemotoxic elements in mixed wastes

Non-radioactive substances in nuclear wastes

HPGe-detector

14 MeV Neutron Generator

6.5 T Graphite Reflector/Moderator

1 T Crane

Turntable inside the chamber

PGNAA

Compound Nucleus

Delayed Gamma

Electron (after Beta decay)

Stable Nucleus

DGNA
- Tiefengeologische Endlagerung ist die beste Option

- In Deutschland: Endlager Konrad ab Mitte 20er Jahre, Endlager für hochradioaktive Abfälle gegen Ende des Jahrhunderts

- Endlagerung in einigen EU Mitgliedsstaaten bereits weit fortgeschritten – erstes Endlager für hochradioaktive Abfälle Anfang der 20er Jahre (in Finnland)

- Wissenschaftliche Basis und Begleitung - langfristig bis zum Verschluss des Endlagers erforderlich