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ABSTRACT

This report presents the State-of-the-Art with respect to the fast / instant release of safety relevant radionuclides from spent nuclear fuel. In the first part of the report basic information is described, such as the characterization of nuclear fuel, irradiation and temperature induced processes in UO$_2$ during its use in reactors, and disposal concepts for spent nuclear fuel in different countries. Secondly, the State-of-the-Art on fast release is documented by a summary of results obtained from more than 100 published experiments using different samples, experimental techniques, and duration of the experiments. All authors refer to a definition of the fast / instant release as a fraction of the inventory of radionuclides that may be rapidly released from the fuel and fuel assembly materials at the time of canister breaching. In the context of safety analysis, the time of mobilization of this fraction can be considered as an instantaneous release of some radionuclides at the containment failure time. The State-of-the-Art will be updated regularly according to the results obtained in the 7th Framework Programme Collaborative Project FIRST-Nuclides.

INTRODUCTION

The Fast Instant Release fraction of spent nuclear fuel is an operative definition arising from early spent nuclear fuel leaching experiments and it entails all those nuclides that as a result of the neutronic and thermal fields in the reactor have migrated towards the surface of the spent fuel and are easily accessible to water.

The definition of Fast Instant Release Fraction was initiated in the 80’s during the early days of the Spent Nuclear Fuel workshops where the US, Canadian and Swedish programs shared their experiences in informal annual meetings. In this context, the US and Canadian programs did a substantial amount of experimental work that will be analysed in the data section.

The Fast Instant Release Fraction was incorporated in the frame of the safety assessment of spent fuel disposal in the safety assessment exercises performed at the beginning of the 90’s, particularly in SKB91 (SKB, 1992). In this SA exercise the release of a number of fission products (I and Cs) was assumed to be, neither limited by the dissolution of the UO$_2$ matrix.
itself, nor by their own solubility and in this respect they were assumed to be released instantly when water was put in contact with the spent fuel.

Other national programs became members of the Spent Nuclear Workshop during the 90’s and they adopted this terminology in their approach to spent fuel dissolution. The operational basis of the Fast Instant Release Fraction bears in itself some intrinsic limitations in the way they are treated in PA related modelling. This is highly unsatisfactory from the scientific and technical perspective as it unreasonably penalizes spent fuel disposal from the radiological point of view. Under this perspective it is highly advisable that a more scientific and consistent approach is developed to characterize and model the potential release of these fission products.

Instant Release Fraction has been the objective of investigations since the early days of spent fuel investigations. In addition, the fast / instant release of radionuclides from spent nuclear fuel has been investigated in a series of previous European projects (such as SFS [1, 2], NF-Pro [3], MICADO [4]). Furthermore, mainly French research programs investigated and quantified the rapid release [5-7].

In this context, the 7th Framework Programme Topic Fission-2011-1.1.1 “research activities in support of implementation of geological disposal”, the Collaborative Project (CP) Fast / Instant Release of Safety Relevant Radionuclides from Spent Nuclear Fuel (FIRST-Nuclides) aims to bring about a more scientific basis to the understanding and the modelling of the release of these initially easily dissolving nuclides. The CP started in January 2012 and extends over 36 months. The objectives of the project are in line with the Vision Report and the Strategic Research Agenda (SRA) of the “Implementing Geological Disposal – Technology Platform (IGD-TP)”. Six experimental facilities having specialized installations and equipment to work with highly radioactive materials and four organizations having specific knowledge on this subject enter into an Inter-European collaboration. The experimental facilities will perform studies using high burn-up spent nuclear fuels in combination with advanced analytical methods.

The outcome of abundant research on instant release fraction characterization and dissolution has clearly put in evidence the relationship between, fuel burn-up, fission gas release and Cs content in the gap. However, there are still some open questions, as pointed out during the MICADO project. Among them the most critical would be:

- To define more realistic relationships between FGR and the presence of fission products in the gap, including C, Rb, Sr, I and Tc
- The quantification of the IRF in high burn-up fuels
- The role of grain boundaries in the potential retention of fission products.

A more recent publication, (Johnson et al, 2012) brought some additional data from higher burn-up fuels that indicated that the basic relationships between FGR and Cs release were maintained and that the I release was closely related to FGR. Nevertheless, not sufficient data
are available and significant correlations between sample preparation and IRF quantification seem to exist.

The project FIRST-Nuclide aims at contributing to find a satisfactory answer to the former questions. The present report summarises the state-of-the-art and demonstrate the rationale for the investigations undertaken within the project, based on expensive experiments using irradiated nuclear fuels in hot cell facilities and application of sophisticated analytical methods and modelling tools.

The beneficiaries of the consortium are the following organizations: Karlsruher Institut fuer Technologie (KIT) DE, Amphos 21 Consulting S.L. (AMPHOS21) ES, JOINT RESEARCH CENTRE – INSTITUTE FOR TRANSURANIUM ELEMENTS (JRC-ITU) EC, FORSCHUNGSZENTRUM JUELICH GMBH (JÜLICH) DE, PAUL SCHERRER INSTITUT (PSI) CH, Studiecentrum voor Kernenergie (SCK•CEN) BE, CENTRE NATIONAL DE LA RECHERCHE SCIENTIFIQUE (CNRS) FR, FUNDACIO CTM CENTRE TECNOLOGIC (CTM) ES, Magyar Tudományos Akadémia Energiatudományi Kutatóközpont (MTA EK) HU, STUDSVIK NUCLEAR AB (STUDSVIK) SE. Associated groups, contributing also to FIRST-Nuclides with their experiences and having access to the discussions within the project are: Commissariat à l’énergie atomique et aux energies alternatives (CEA) FR, Los Alamos National Laboratory (LANL) USA, Sandia National Laboratory (SNL) USA, National Decommissioning Authority (NDA) UK, National Nuclear Laboratory (NNL) UK, Posiva Oy FI, Teollisuuden Voima Oy (TVO) FI.

1. DESCRIPTION of the FUEL

The spent nuclear fuel considered in the CP FIRST-Nuclides consists of UO₂ fuel. MOX fuel is presently not within the scope of the project. The fuel burn-up covers the range between 48 and 71 GWd/tHM which is representative of the presently operative PWRs and BWRs. Defining an average burn-up of the fuel is difficult, because of the different enrichments and Gd₂O₃ content in fuel rods and the variation of the neutron flux in the reactor of the upper and lower part of a fuel rod. For this reason different fuels (PWR, BWR and THTR / VVER types) and burn-ups (from 48 to 71 GWd/tHM) will be used in the experimental program, as shown in Section 6.

UO₂ fuel

Presently, UO₂ is produced from UO₂/U₃O₈ or Ammonium di-Uranate (ADU) powders which are compacted together with sintering aids to an initial density of 4.5-5 g cm⁻³. (see Figure 1). The sintering is performed at ~1700 °C for 2 to 8 hours. The grain size of this material can be adjusted through the sintering process and by adding additives to the powders such as Cr₂O₃. The grain size influences both the mechanical properties and the fission gas release (FGR) of the fuel. Larger grains show a lower FGR. Additives produce a coating onto the outer surfaces
of the grains what reduces the FGR further. Additives up to 50,000 ppm of metal oxides can form oxygen defects, which can incorporate fission gases. During the sintering process, the remaining U(VI) is reduced. Present UO₂ fuels have grain sizes of 10-20 µm, a density of 10.0 to 10.8 g cm⁻³ and pore sizes in the range of 5 to 80 µm.

**Figure 1.** From powder to pellet. A) Powder: UO₂ grain size of 20 µm; B) Pellet: PWR 17×17 - Ø 8.17 (AREVA) - a length of 9.8 mm, BWR 10×10 - Ø 8.87 (Atrium 10XP) - a length of 10.5 mm.

**Relationship between fuel enrichment and burn-up**

The resulting fission product generation and consequently the composition and abundance of fission products is related to the fuel history and burn-up. In this respect, there is a correlation between initial fuel enrichment and burn-up. In the early days of nuclear power generation average burn-up values ranged between 30-50 GWd/tHM. In the last years, with the aim of increasing cost efficiency and decrease spent fuel volumes, the discharge burn-up has increased.

In 2003 the OECD/NEA Nuclear Science Committee initiated an Expert Group on Very High Burn-ups in LWRs, with the task of delivering a state-of-the-art report on high burn-ups in LWRs. Very high discharge burn-ups are defined by this group, as average burn-ups from 60-100 GWd/tHM. The 60-100 GWd/tHM range took the analysis largely beyond the usual range of current LWR experience (although there was experience of experimental test rods having been taken into this burn-up range) and looked very much to the future of LWRs. The report covered only conventional LWR fuel assembly designs, which conform to the assembly geometries used in present LWRs and that use conventional oxide fuels [9]. Figure 2 shows the initial ²³⁵U enrichment versus the average discharge burn-up.
The initial enrichment relation in Fig. 2 indicates that the maximum average discharge burn-up achievable within the 5.0 wt.% $^{235}$U fabrication limits is slightly above 60 GWD/tHM. This figure may be slightly pessimistic because of the gadolinium residual absorption penalty and because a five- or six-batch scheme or higher (instead of 4 cycles) are known to yield slightly higher average discharge burn-ups for the same initial $^{235}$U enrichment.

As example, the following information can be useful: In BWR from the Leibstadt powerplant (BWR-KKL) the fuel pellet enrichment varies between 4.46 and 3.71% $^{235}$U and density of 10.50 and 10.53 g cm$^{-3}$ respectively. The average grain size is 10–11 µm [10]. In PWR from the Gösgen plant (PWR-KKG), the initial $^{235}$U enrichment in UO$_2$ fuel rods is presently 4.5-5.0 % $^{235}$U. In the PWR from the Neckarwestheim-II reactor (PWR-GKN II), the enrichment is 4.4 wt.% for fresh UO$_2$ fuel and 4.6 wt.% for UO$_2$ fuel produced from reprocessed uranium.

**Fuel Rods and fuel elements**

Besides burn-up, another factor the affects fission product generation is the actual geometry of the fuel rods and fuel assemblies. The light water reactor (LWR) is a nuclear power reactor initially developed in the USA. The power reactor is filled with normal water. This water cools the power reactor and moderates the neutron energy to help facilitate nuclear fission. There are two main types of reactors: the pressurized water type (PWR) and the boiling water type (BWR). The LWR fuel element, both for PWR and BWR, consists of a quadratic arrangement of fuel and control rods (see Figure 3).

A **fuel rod** itself consists of a gas-tight Zircaloy/M5 cladding and has end caps in both ends. At the bottom there is a supporting sleeve. Above, an insulating pellet of Al$_2$O$_3$ separates the
supporting sleeve from the uranium dioxide pellets. The UO$_2$ pellets are stacked up to the heights of the reactive zone of the reactor. Also at the top of the fuel rod, an Al$_2$O$_3$ insulating pellet separates the UO$_2$ pellets from the fission gas plenum, which contains a pressure spring. The spring keeps the pellets in place while reserving some space inside the rod for volume expansion as the pellets expands due to heat and neutron irradiation, and because of formation of gaseous fission products. The fuel rods are filled with He gas to a pressure of 22 bar. At the top and bottom, the rods are fixed by an anchor grid, between top and bottom the rods distance between the rods is fixed by spacers. The rod material consist of Zry-2 partly Fe enhanced for BWRs and of M5, Optimised Zry-4, modified Zry-4 or Duplex [11] for PWRs.

A fuel element consists of the fuel rod, spacers and control rods. The nuclear fuels to be considered in the CP FIRST-Nuclides are originated from, mainly pressure water reactors (PWR), boiling water reactors (BWR), as well as VVER-440 reactors. An overview of relevant fuel element types is given by [11], where four main reactor types (PWR, VVER, BWR and heavy water) are represented. Illustrations and photographs show the representative designs for most of the manufacturers. AREVA produces PWR fuel elements arranged in a square, holding 14x14-(16+1) 15x15-(20+1) 16x16-(20) 17x17-(24+1) to 18x18-(24) fuel rods, for BWRs the ATRIUM fuel elements have 9x9 and 10x10 designs. A cross cut through a BWR fuel element (Olkiluoto, OL1/2, Findland) is given in Figure 4, showing the arrangement of the two water rods for improvement of moderation and the twelve gadolinium doped rods [12]. These rods are required for high burn-up, where higher initial $^{235}$U enrichment is used. For new fuel elements, a reduction of the power peaking factor and a compensation of the excess reactivity is achieved by the burnable neutron poison, Gd. Some fuel elements have radially zoned enrichment distributions, which reduce the power peaking factor and optimize the heat transfer.
Figure 3. Schemes of PWR and BWR fuel assemblies, fuel rods and pellet characteristics. Taken from http://www.nfi.co.jp/e/
Figure 4. Arrangement of radially zoned fuel rods, Gd$_2$O$_3$ doped rods and water rods in a 10×10 OL1/2 BWR fuel element.

The fuel rods may have many different levels of enrichment zones and burnable absorbers. It is not uncommon to have ten or more different fuel rod designs in a single assembly. It is very important that a fuel rod of a given design be located in its designed position in the assembly [13].

Optimization of nuclear fuel (UO$_2$)

As previously mentioned, with the aim of enhancing the economical efficiency of the nuclear generation cycle, and to reduce the amount of spent fuel, the fuel discharged burn-up has been increased in the last years. This is the reason why research on fuel pellets focuses more and more on the high burnup fuel pellets.

The utilities have put in place a number of improvements to optimize the performance of the UO$_2$ fuel during operation. Some of them have clear consequences in the fission gas release (FGR) and fission product generation. The optimization of the nuclear fuel covers a series of different processes:

- The initial enrichment with respect to increased burnup,
- The control of criticality,
- The burn-up behaviour, such as swelling
- The mechanical and thermal requirements,
- Pellet cladding interactions (PCI)
- Minimization of fission gas release by adjusting UO$_2$ grain sizes, grain pores, and grain coverage.

Most of these optimizations are only described in patents. For example, for criticality control, some rods in the fuel elements are doped with Gd. In BWR fuel elements between 1 and 18
rods are doped between 1 and 7 wt.%. In PWR fuel elements between 2 and 12 rods are doped with Gd$_2$O$_3$ in the concentration range between 1 and 7 wt. %. The manufacturing process of these binary oxide fuel was published by Assmann [17].

The mechanical and thermal behaviour of the fuel, the UO$_2$ grain sizes and grain pores are controlled through the sintering process. Various sintering processes have been described (mainly in patents). One example is the NIKUSI low temperature process. The "NIKUSI" process bases on a two steps low temperature sintering technique for UO$_2$ involving sintering at 1100 to 1200°C in CO$_2$ and later reduction in hydrogen.[18].

A higher burnup in the fuels, leads to an increase in the amount of fission gases such as xenon (Xe) and krypton (Kr). The fission gases in the pellet are continuously released out from the pellets during the reactor operation and increase the internal pressure of nuclear fuel rod. The increased internal pressure induced by the nuclear fission gases increase the stress working on the cladding tube, resultantly reduces the safety margin of the nuclear fuel rod. Thus, in order to solve such a problem, the nuclear fission gases generated due to nuclear fission must be released out as small amounts as possible to the outside of the sintered pellet.

The process of nuclear fission gas release to the outside of the nuclear fuel pellet is generally conceptualised in a way than when the fission gas is generated in a grain, it moves to the grain boundary through diffusion so as to exist as bubbles in the grain boundary, and when the bubbles are increased to reach a certain amount, a bubble tunnel is formed along the grain boundary, and the bubbles are then discharged to the outside of the nuclear fuel pellet (for a more complete description of the different concepts and models developed to account for FGR see deliverable D.4.2. of the FIRST-Nuclides project, which can be accessed in the project website). Therefore, the larger the grain size of the pellet, the longer the time needed for the gas to reach the grain boundary, thus to be released. This means that a high burnup fuel would require a larger grain size to minimize gas release.

Admixtures of various metal oxides control the evolution of mono-disperse grain size during sintering [19-21]. The pore structures and pore sizes of uranium dioxide fuel can also be varied by application of precursor liquids, for example, allylhydridopolycarbosilane (AHPCS) before sintering [22]. Present high burn-up fuel has grain sizes between 20 and 25 µm. Covering the UO$_2$ grains by various ceramic compounds described in some patents can also minimize FGR. If and to which extent such specially treated UO$_2$ is used by the utilities is regarded is uncertain as it belongs to the realm of utilities operational know-how.

A low density of the material, on another hand, may enhance fission gas release. This effect has been observed from experiments reported in Johnson et al. (2012), when comparing the results of FGR obtained from fuels of a density of 10.05-10.3 g/cm$^3$ instead of the usual denser fuels (10.5g/cm$^3$).
2. Irradiation induced processes in UO₂

The abundance and generation of fission products are the result of the irradiation processes in the fuel. Originally UO₂ fuel is fabricated by pressing and sintering the powder to a density less than 100% of the theoretical density due to pores in the bulk material. During a burn-up in the range of ~50 GWd/tHM, the FIMA (Number of fissions per initial metal atom) is about 5.5 atom-%. The fission and fission products cause expansion in the UO₂ crystal structure leading to swelling [14].

Initial porosity reduces pellet swelling. Swelling is a consequence of the fission process increasing the number of atoms thus affectig the physical/chemical properties of the fuel. A high initial porosity is helpful from this point of view but it has other consequences, for example:

- Porosity influences all the physical properties of the fuel, e.g. thermal conductivity, creep and strength, and elastic constants.
- Porosity influences moisture and residual gas content of the pellets.
- It may produce overcompensation of the swelling during the early reactor irradiation causing rapid shrinkage of small sized pores and a densification of the UO₂.

UO₂ spent fuel stoichiometry and composition.

In the case of the crystal structure, the face-centered cubic UO₂ shows an extended range of composition apart from the exact stoichiometry O/U = 2, so that one may write UO₂±x. The deviation x from the stoichiometric composition affects all physical and chemical properties, in particular those which depend on the atomic mobility (e.g. diffusion coefficients), and the oxygen partial pressure or chemical behaviour of the fission products.

The chemical stability of oxides of the fission products in equilibrium with UO₂±x, can be classified into three main groups:

(i) The rare earth elements and Y, Zr, Ba and Sr, whose oxides form either solid solutions with UO₂, or single phase precipitate.
(ii) Mo, Cs and Rb, which are either oxidized or not, depending on the O/U ratio; and elements like Ru, with unstable oxides which form metallic precipitates within the UO₂.

Fabrication imperfections

There are a number of potential fission processes that may arise as a result of defects and failures in the fabrication of the fuel pellets. The most interesting in relation to FIRST project are the so-called pellet to cladding interactions. Pellet to cladding mechanical interaction is associated with a defect in the pellet or cladding, such as a missing pellet surface (MPS),
which creates excess stress in the cladding. Rapid changes in local power cause pellet expansion, which, in the presence of these stress multipliers, can break the cladding. The other type of pellet cladding interaction is stress corrosion cracking. High power levels promote the release of fission product gases. Iodine in particular is very corrosive to Zircaloy. The presence of iodine near a preexisting cladding imperfection or a stress riser accelerates crack propagation through stress corrosion cracking. PCI can also occur during reactor startup due to rapid changes in core power and unconditioned cladding.

**Thermal processes during operation**

The UO$_2$ pellets are subject to a high central temperature and a steep radial temperature gradient. Like other ceramic materials, UO$_2$ shows little thermal shock resistance and behaves in a brittle manner at low temperatures. The vapour pressure increases rapidly with temperature. These properties are responsible for the development of a typical crack pattern in the cool outer part of the pellets and micro-structural changes in the hot inner part of the pellets, up to a complete change in microstructure without melting.

The actual temperature, stresses and behaviour of fuel rods under irradiation are modeled by 2D or 3D codes [15]. For a rough estimation, an analytical equation may be used which relates the linear power to the temperature increase $\Delta T$ of a fuel rod.

$$\Delta T = \frac{\text{lin. power}}{4\pi \cdot \lambda}$$  \hspace{1cm} \text{eq. 1}

Using eq. 1 following temperatures are obtained for different reactors (Table 1) (The heat conductivity $\lambda_{\text{irrad fuel}} = 2.5 \text{ W m}^{-1} \text{ K}^{-1}$ was used [16]).

<table>
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<th>Pellet</th>
<th>KKG (PWR)</th>
<th>GKN II (PWR)</th>
<th>KKL (BWR)</th>
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<tr>
<td>$\varnothing$ [mm]</td>
<td>9.3</td>
<td>8.05</td>
<td>8.5</td>
</tr>
<tr>
<td>Lin power [W/cm]</td>
<td>228</td>
<td>167</td>
<td>184</td>
</tr>
<tr>
<td>$\Delta T$ [K] (eq.1)</td>
<td>725</td>
<td>528</td>
<td>585</td>
</tr>
<tr>
<td>$T_{\text{coolant}}$ [K]</td>
<td>325</td>
<td>305</td>
<td>263</td>
</tr>
<tr>
<td>$T_{\text{center}}$ [K]</td>
<td>1050</td>
<td>833</td>
<td>848</td>
</tr>
</tbody>
</table>

During the operation in a reactor, the temperature causes a radial thermal expansion of the fuel pellets, which is partly reversible when cooling.
3. Modeling tools for fuel performance

The morphology of the fuel pellet, the properties of uranium oxide, its structure, thermal expansion and thermal conductivity and heat capacity affect the performance of nuclear fuel. The properties of the cladding materials have to guarantee compatibility of the UO$_2$ fuel with respect to thermal properties, linear thermal expansion, thermal conductivity, specific heat capacity and the mechanical properties, including the elastic constants and the plastic deformation properties. They account also for irradiation effects, such as irradiation-induced growth and hardening, as well as irradiation-induced creep behaviour. The corrosion and hydrogen pickup of the cladding are also important parameters especially under the high temperature conditions in a reactor. All these processes are accounted for in modelling tools currently used by the utilities to simulate and predict fuel behaviour during reactor operation.

Modelling tools used by the utilities take into account basic phenomena for in-reactor performance, such as the neutronics of the fuel, the evolution of the nuclide content, the presence of absorbers. They also take into consideration all heat transfer processes and the thermal characteristics under consideration of the axial heat transport in the coolant, the heat transport in the pellet, the heat transport through the cladding. The effects of irradiation on gap conductance between the pellets and cladding are also taken into account. The modelling of the mechanical behaviour includes the calculation of strains in the pellet and cladding. Further modelling tools have been developed for studying the fission gas behaviour, for modelling the high burn-up structure, pellet-cladding interaction, irradiation-induced stress corrosion cracking and cracking events caused by power ramps. Additionally, a series of codes deal with accidental situations in a reactor core. A comprehensive summary of the relevant basic assumptions and equations are provided by Van Uffelen et al. in the recent published Handbook of Nuclear Engineering [23].

The utilities and manufacturers of nuclear fuel use a number of different codes. A special issue of the journal “Nuclear Engineering and Design” collected the papers from the IAEA Specialists’ Meeting on the Computer Modelling of Nuclear Reactor Fuel Elements, held March 13-17, 1978 at Blackpool, UK (Vol. 56 (1) 1980). Various specialists meetings and workshops have been organized, for example by IAEA [24] and NEA. A recent overview on Fuel Performance Codes was published by Stan [25]. Dion Sunderland; Nuclear Science and Technology Interaction Program (NSTIP), ORNL, have provided the list of important codes July 8, 2011). Codes are used for different purposes, e.g. regular fuel performance, but also for “loss-of-coolant accidents (LOCA)”. They consider items such as:

- thermal-mechanical behaviour of the fuel (pellet), thermal expansion, creep effects, thermal induced porosity effects and densification, cracking
- fission induced densification and swelling, pore formation
- pellet cladding interactions (PCI),
chemical properties such as diffusion processes for oxygen and fission products, crystal structure – defects, phase stability of nuclear fuels, especially during transient regimes,

- Fission gas release (FGR).
- A detailed description of effects relevant for FGR is given in the thesis of Peter Blair [26]. Actual FGR codes include:
  - Recoil and Knockout
  - Diffusion
  - Diffusion and trapping onto natural defects such as grain boundaries, dislocation lines, closed pores in as fabricated fuel and impurities in the solid. Trapping onto radiation produced defects, such as vacancy cluster, interstitial loops, fission gas bubbles, solid FP precipitates.
  - Gas accumulation in grain boundaries (in grain bubbles, in grain solution between grain boundaries)
  - Sweeping of gas bubbles by grain boundaries
  - Gas break away by gas bubble interaction
  - Bubble migration

Under the guidance of the Nuclear Science Committee (NSC) of the NEA, Multi-scale Modelling Methods are evaluated (Fig. 4). The expert group aims to provide an overview of the various methods and levels of models used for modelling materials for the nuclear industry (fuels and structural materials). The state of the art includes an overview of the methods but also the possibilities and limits of linking different scales. The report also covers the “Mixed-oxide (MOX) Fuel Performance Benchmark for the Halden reactor project MOX Rods [27]. (see http://www.oecd-nea.org/science/wpmm/expert_groups/m3.html).

**Figure 5.** Space and time scales involved in simulating phenomena relevant for nuclear materials. The modelling assumptions and approaches are shown in parenthesis [28].
Since 2003, annual Materials Modeling and Simulation for Nuclear Fuels (MMSNF) workshops are organized. The goal of the MMSNF workshops is to stimulate research and discussions on modeling and simulations of nuclear fuels, to assist the design of improved fuels and the evaluation of fuel performance. In addition to research focused on existing or improved types of LWR reactors, modeling programs, networks, and links have been created. Examples are organized in EURATOM 7\textsuperscript{th} FP, such as the F-BRIDGE project (Basic Research for Innovative Fuel Design for GEN IV systems (www.f-bridge.eu), the ACTINET network, the EURACT-NMR coordination and support action\textsuperscript{1} of to provide access to European nuclear licensed facilities that have recently invested in advanced nuclear magnetic resonance spectrometers.

A review of the available models developed to account for fission gas release can be found in deliverable D.4.2 of the CP FIRST-Nuclides project, available from the project website.

\section{Disposal of spent nuclear fuel}

While the abundance and composition of fission products and fission gas release is highly dependent on the fuel fabrication and reactor operational factors, the fast release fraction is only relevant in the frame of the disposition of the used fuel. It is a very important and critical component of the performance of spent fuel storage facilities, but it is also very relevant when assessing the performance of spent fuel geological repositories. In this context, we will give a brief account of the commonalities in European disposal concepts and how they affect potential dissolution behaviour of the SNF, and the fast release of the fission products.

\subsection*{Canister concepts}

All European disposal concepts for spent nuclear fuel consider thick-walled inner steel casks having various overpack or other protecting properties. The different/canister disposal concepts are described in Appendix II.

\subsection*{Water contact to the fuel in the casks}

Under disposal conditions, one may assume that after a certain period of time water penetrates into the inner steel cask and starts corroding the steel at the inner steel surfaces causing a hydrogen pressure build-up due to anoxic steel corrosion. Some corrosion mechanisms are shown in Appendix II.

\subsection*{Water access to the spent fuel}

The potential barrier function of the Zircaloy cladding of the fuel rods is not considered in current assessments but the behaviour of this material may affect the instant/fast release of

\footnotesize\textsuperscript{1} Nuclear magnetic resonance can provide unique atomic scale structural information in liquids and crystalline and amorphous solids. High resolution instruments operating at KIT-INE (liquids) and JRC-ITU (solids) offer the opportunity to apply this technique to actinide containing materials through the FP7 trans-national access programme EURACT-NMR (www.euract-nmr.eu).
radionuclides from the spent fuel. An irradiated cladding can contain local hydrogen concentrations higher than 1000 ppm. The embrittlement effect of hydrogen in Zircaloy is far less at 350°C than that at lower temperatures e.g. 100°C [33]. Under the assumption that water penetrates into the canister, anaerobic corrosion of the steel starts forming hydrogen gas. The gas dissolves in the contacting water until its solubility is exceeded. A gas phase can be formed afterward, and the gas pressure increases until an equilibrium pressure is achieved which depends on the depth of the repository. At 500 m depth, the maximum pressure is in the range of 5 MPa. This may cause further penetration of H₂ into the Zircaloy. Detailed studies of the H₂ effect on the Zircaloy embrittlement are available, e.g. [34-36]. Due to the homogeneous distribution of H₂ over the length of the fuel rods, embrittled material zone extends over the complete length of the rods. Therefore, it can be expected that the cladding of the fuel rods will not be damaged by single holes (such as pitting corrosion in stainless steel) but by ruptures, especially at positions where pellet cracking occurred. Maximal cladding hoop stress is located between pellets. Such defects have been investigated in the context of Pellet Cladding Interaction studies [37, 38]. A schematic view is given in Figure 6.

5. Previous Investigations on fast/instant release

In the following section, previous investigations, results, and conclusions of the beneficiaries of the CP FIRST-Nuclides are summarized. The data generated after 2005 uses Johnson’s et al. definition on the fast/instant release [7] in order to select the data. Since SKB 91, the Spent Nuclear Fuel (SNF) source term in a water-saturated medium is described as the combination of two terms:

1. A fraction of the inventory of radionuclides that may be rapidly released from the fuel and fuel assembly materials at the time of canister breaching. In the context of safety
analysis, the time of mobilization of this fraction can be considered as an instantaneous release of some radionuclides at the containment failure time.

2. A slower long-term contribution corresponding to the dissolution of the uranium oxide matrix, induced by \( \alpha \)-radiolytical oxidation processes and impeded by the effect of hydrogen. The so-called hydrogen effect, which reduces the matrix dissolution extent, has been incorporated in a later stage.

The state-of-the-art report, surveys the published information. In a later step, each project beneficiary will provide information on his own investigations, including fuel material, kind of experiments (FGR, dissolution based release), sample sizes, duration of the experiments, solutions composition, pH, \( \text{pO}_2 \) (Eh), \( \text{pH}_{\text{CO}_2} \), …, and the obtained results.

In 2005, Johnson et al. generated a number of tables covering the fast release (leaching) data until 2005 [2, 7]. In the following, the different leaching experiments with SNF (UO\(_2\) and MOX) that have been performed during the last 30 years are given. In some cases, although the work was not focused on the study of the IRF, it is possible to calculate it. All the data are listed in Table 2 and Table 3. The experiments were carried out using, essentially, four kinds of samples:

- Bare fuel: consisting of a piece of fuel and cladding with an artificially produced defect.
- Pellet: this term will be used when referring to a cladded fuel segment.
- Fragments: pieces or portions of fuel without cladding.
- Powder: fuel material obtained after de-cladding, sieving and milling.

The earlier IRF data was generated within the US Yucca Mountain project. The publications cover the span: 1987 to 1990; Oversby and Shawn (1987)[39]; Wilson, 1987[40]; Wilson and Shawn, 1987[41]; Wilson, 1988[42]; Wilson, (1990 a, b)[43, 44]; Wilson and Gray, 1990 [44]. In this work they used two bare fuel specimens prepared from, HBR-type fuel (BU of 30 MWd/kgU) and TP-type fuel (BU of 27 MWd/kgU) PWR SNF’s. The tests were conducted in unsealed silica vessels under ambient hot cell air and temperature conditions. The initial weight was 83.10 g for HBR SNF and 27.21 g for TP SNF. The leaching solution was J-13 water, which corresponds to the chemical composition of the J-13 well in Yucca Mountain. The composition in mmol/L was: \( \text{Na}^+ \) 2.2; \( \text{K}^+ \) 0.13; \( \text{Ca}^{2+} \) 0.38; \( \text{Mg}^{2+} \) 0.08; \( \text{Si} \) 1.14; \( \text{F} \) 0.37; \( \text{NO}_3^- \) 0.15; \( \text{SO}_4^{2-} \) 0.2; \( \text{HCO}_3^- \) 2.0, pH 7.2. Two leaching cycles were performed consecutively. The experiments were carried at 25 and 85°C. The data reported in Table 1 correspond to the sum of the two cycles that means a total leaching time of 425 days for HBR SNF and 376 days for TP SNF.

Within the Yucca Mountain project additional IRF dissolution tests were performed; Gray and Wilson, (1995) [47]; Gray [48]: measured the gap and grain boundaries inventories from different PWR SNF’s: ATM-103 (BU of 30MWd/kgU), ATM-104 (BU of 44 MWd/kgU) and ATM-106 (BU of 43 and 46 MWd/kgU), and from BWR SNF’s: ATM-105 (BU of 31 and 34 MWd/kgU).
The fuel specimens were prepared from 12 to 25 mm long SNF-segments. To measure the gap inventories, the SNF was discharged from the cladding and placed in a glass vessel along with the empty cladding segment. A measured volume (200 to 250 mL) of distilled ionised water (DIW) was added to the vessel and allowed to stand for 1 week at hot cell temperature under ambient atmosphere. Grain boundary inventory measurements were performed on the SNF specimens after completion of the gap inventory measurements. After one week exposure to water, the SNF fragments were dried, crushed, and screened using screens with 20 to 30 µm openings, depending on the grain size of the SNF being prepared. The grain-boundary inventory measurements consisted of placing 0.5 g of the screened SNF grains and subgrains in a 50 mL beaker along with 20 mL of 0.1 M HCl. Periodically after 3 h the acid was removed, filtered, and replaced with fresh acid.

- The Swedish program performed characterization of fuel samples and dissolution experiments of a number of specimens from the Oskarsham and Ringhals reactors. Forsyth and Werme, (1992) [45]; Forsyth, (1997) [46]: experiments performed with 20mm long fuel/clad segments from Oskarsham-1 BWR SNF (BU of 42 MWd/kgU), Ringhals-1 BWR SNF (BU between 27.0 and 48.8 MWd/kgU), and from Ringhals-1 PWR SNF (BU of 43 MWd/kgU). The specimen, a fuel/clad segment suspended in a spiral of platinum wire, was immersed in 200 ml of the leaching solution in a 250 ml Pyrex flask. All tests were performed at 20-25°C, ambient temperature of the hot cell and under oxidizing conditions. The solution compositions used were DIW (De-Ionized water) and GW (GroundWater), the composition in mmol/L of the GW used was: Na⁺ 2.8; K⁺ 0.1; Mg²⁺ 0.2; Ca²⁺ 0.45; Si 0.2 HCO₃⁻ 2.0, Cl⁻ 2.0; SO₄²⁻ 0.1. The experiments were performed at room temperature and contact times were of the order of 7 days.

- Within the Spanish programme, Serrano et al., 1998 [49] conducted experiments with irradiated fuel samples prepared from pins of two SNF’s: UO₂ (BU of 54 MWd/kgU) and MOX (BU of 30MWd/kgU). Sequential batch leaching experiments in DIW at room temperature under ambient atmosphere were made. On completion of the selected contact period, the samples were transferred to clean vessels containing fresh solution. The leaching times encompassed an interval between 24 and 1300 hours. The vessels were, after used, rinsed with 1M HNO₃ for 1hour at room temperature. During the experiment, the vessels were kept closed tight; after the longest leaching time, no significant loss of leachate from the bottles was observed. The leaching experiments with irradiated fuel were performed in a hot cell at an ambient hot cell temperature of 25±2 °C. The data given in Table 2 and Table 3 correspond to a leaching time of 31 days. The matrix contribution was subtracted.

- IRF characterization and dissolution data generated in the Transuranium Institute was reported by Glatz et al., 1999 [50]. In this work, three MOX SNF’s (BU of 12, 20 and 25 MWd/kgU) and three UO₂ SNF’s (one with 30 MWd/kgU and two with 50 MWd/kgU) rods, each of them about 6 cm long were used. Both ends of each rodlets were closed by
means of tight stainless steel end-caps. One UO₂ fuel rod with a burn-up of 50 GWd/tHM was machined to produce two defects (in each case, 3 holes of 1 mm diameter each), one at the top and in contact with vapour and the other at the bottom of the rod and in contact with the leaching solution. In all the other samples, the defects were placed in the centre of the rod and the autoclave was filled completely with the leaching solution. MOX fuels were fabricated following the MIMAS blend process. The leaching experiments were carried out in autoclaves equipped with Ti-liners, using DIW at 100°C under anoxic or reducing conditions. The data given in Table 2 and Table 3 correspond to a leaching time of 365 days without considering the matrix contribution.

- The German programme reports work on spent fuel dissolution since the early 90’s. Loida et al., 1999 [51] reported ¹³⁷Cs the initial release of ¹³⁷Cs from various spent fuel materials measured in MgCl₂-rich and concentrated NaCl solutions at 25, 90, 100, 150 and 200°C. The materials, sample sizes, duration of the experiments, and solution compositions are summarized in in Table 2 and Table 3. These experiments cover mainly fuels of a burn-up between 36 and 50 GWd/tHM. Most of the ¹³⁷Cs IRF are below or in the range of the pessimistic values compiled by Poinssot and Gras (2009) [52]. An IRF of 3.7% of the total ¹³⁷Cs was measured in a leaching experiment with a spent nuclear fuel sample from the NPP Biblis (KW, discharged June 1979; burn-up of 36.6 GWd/tHM) in concentrated NaCl solution at 200°C. This value is slightly higher than the pessimistic ¹³⁷Cs IRF estimate for fuel with a burn-up of 41 GWd/tHM. The experiments were performed partly at FZK (today KIT) and partly at KWU.

- In a later stage of the Spanish programme, Quiñones et al., (2006) [53]: investigated three UO₂ PWR SNF’s samples: U-568 (BU of 29.5 MWd/kgU), B4 (BU of 53.1 MWd/kgU) and AF-02 (BU of 62.8 MWd/kgU). Discs containing cladding and fuel with an approximately thickness of 2 mm were cut from the fuel rods. The weight of the SNF contained in these discs was about 2 g. Two samples of each specimen were prepared. Static batch leaching experiments were performed in 70 ml volume borosilicate glass vessels. All experiments were performed at room temperature, in a hot cell with air atmosphere. The leaching solution consisted of 50 ml of simulated groundwater, of the following composition (in mol/kgH₂O): Na⁺ 4.09·10⁻⁴; K⁺ 1.46·10⁻⁴; Mg²⁺ 2.51·10⁻⁴; Ca²⁺ 2.47·10⁻⁴; Cl⁻ 2.37·10⁻⁴; Si 4.99·10⁻⁵; SO₄²⁻ 7.19·10⁻⁵; HCO₃⁻ 1.07·10⁻³; F⁻ 1.05·10⁻⁵; PO₄³⁻ 1.04·10⁻⁷; Al³⁺ 1.85·10⁻⁷; U_total 2.32·10⁻⁹; pH around 7.0. The solution was de-aerated by purging it with inert gas for several hours prior to the start of the leaching. During each contact period, the vessels remained sealed.

Data reported in Table 2 were obtained from the graphics and represent average values of repeated measurements, the values referred to Rb are quite high in comparison with the values reported by other authors. The leaching time was 500 days for U-568 SNF, and 600 days for B4 and AF-02 SNF’s.
Although spent fuel is not a priority in the French waste management program, there has been substantial work performed by the CEA groups with respect to IRF characterization and dissolution. Roudil et al., (2007) [54], Roudil et al., (2009) [55]: investigated five PWR SNF’s samples, four UOX with a BU of 22, 37, 47 and 60 MWd/kgU; one MOX with a BU of 40 MWd/kgU. Two types of samples were used: i) 20 mm segments with cladding and ii) powder samples with a particle size of 20-50 µm. Powder samples were only prepared for the 60 MWd/kgU BU SNF. The experiments were carried out under oxidizing conditions at room temperature (25 °C).

The gap inventories were determined by static mode leaching experiments in carbonate water (10^{-3} M HCO_3^-) with 20 mm clad segments of the five SNF’s. The tests, under air atmosphere, lasted for 62 days with leachates sampled at the following intervals: 3, 10, 24 and 62 days.

Experiments to determine the inventory at the grain boundaries were carried out only on UOX PWR fuel with a BU of 60 MWd/kgU. Leaching experiments were carried out on SNF powder according to a protocol similar to the one developed and validated by Gray [47].

SNF fragments were sampled from the centre of a clad 35 mm segment previously leached for one week in carbonated water to eliminate the gap inventory. Sampling the fragments at the centre of the segment also eliminated the contribution of the rim. Powder samples with a particle size fraction of 20–50 µm were therefore prepared by grinding and sieving in a hot cell. The number of grains in each particle was estimated to be about forty.

Pseudo-dynamic leach tests, under air atmosphere in hot cell, were carried out on 567 mg of powder in 25 mL of carbonated water (10^{-2} M NaHCO_3) to prevent any precipitation of uranium used as a matrix alteration tracer. After each cycle the solutions were filtered and analysed. Fresh water was added to the leaching reactors. Thirty cycles were carried out, initially of short duration (1–2 h) to avoid any precipitation resulting from leaching of the oxidized UO_{2+x} layer, then longer (24–48 h). When the ratio of the released fractions was equal to 1 it was assumed that the complete inventory at the grain boundaries was leached during the preceding cycles.

Roudil et al., 2009 [55]: also studied the IRF coming from fragments and grains from the pellet peripheral zone coupon, near or in contact with the cladding. They used a PWR SNF’s with a BU of 60 MWd/kgU.

Each fuel segment was first slotted to obtain two cylindrical portions separated from the core and consisting of cladding and peripheral fuel. The cladding was then separated from the fuel with a mortar and pestle. The resulting fragments were separated by sieving using 20 and 50 microns screens. To increase the proportion of small grains, such as those in the rim, the powder used for the leaching experiments was sampled from the particle size fraction below 20 microns.

The leach tests were carried out on 282 mg of powder in about 25ml of bicarbonate water to prevent any precipitation of uranium used as a matrix alteration tracer. After each cycle a solution sample of about 17 mL was removed to avoid carrying away powder, filtered to contact with the fuel powder for the next cycle. Twenty cycles were performed in all.
initial cycles were shorter (lasting only few hours) to avoid uranium precipitation due to leaching of the oxidized \( \text{UO}_2^{x+} \) layer. The subsequent cycles lasted 24 hours each.

- Within the Korean waste management programme, Kim et al., (2007) [56] measured gap and grain boundaries in three different PWR fuel rods: SFR1 (BU of 39.6 MWd/kgU), SFR2 (BU of 39.6 MWd/kgU), SFR3-a (BU of 45.8 MWd/kgU) and SFR3-b (BU of 65.9 MWd/kgU). The fuel specimens were prepared by cutting fuels rods thereby obtaining discs with 2 mm thickness. The gap inventory was measured on a SNF pellet without cladding. The SNF and cladding were put into a bottle filled with 100 ml of distilled water. The experiment was run under hot cell conditions. Subsamples consisting of 5 ml of the leachate were sampled after leaching time intervals longer than 7 days.

Two types of SNF powder, sawdust produced during the cutting of fuel rod SFR1 and crushed powder produced by crushing the specimens after the gap inventory experiment, were used for a measurement of the radionuclide inventories in the grain boundaries. In the case of the sawdust, the inventory of the nuclides in the grain boundaries was calculated from the IRF, which is the combined inventories of the gap and the grain boundaries. The powder was leached in 50 ml of 0.1M HCl for about 20 minutes and the solution was sampled by filtering it through a 0.2 \( \mu \text{m} \) filter. The solution was replaced with a fresh acid and subsequently sampled.

- Within the Swedish programme, Fors, 2009 [57]; Fors et al., 2009 [58]. A commercial \( \text{UO}_2 \) SNF with a BU of 59.1 MWd/kgU and an average power line of 250kW/m was used. A 10 mm long segment was cut from a position of the fuel rod. The segment was core drilled to separate the fuel centre from its peripheral 725 \( \mu \text{m} \) thick rim part. The fuel containing the high burn-up structure (HBS) material was detached from the Zircaloy cladding by use of external stress in a screw clamp. The de-cladded fuel fragments contained about 15 wt.% HBS. The millimetre-sized fragments were stored under dry \( \text{N}_2 \) atmosphere (<2 vol.% \( \text{O}_2 \)) for one year before the start of the corrosion experiment. The leachant contained 10 mM NaCl and 2 mM NaHCO\(_3\). The pH of the initial solution was 8.1. After the leachant filling, the autoclave was pressurized to 4.1 MPa with hydrogen. This pressure was kept throughout the experiment. The experiment was carried out at ambient hot cell temperature at 23 ± 4°C. The leachate was not stirred. The results given in Table 1 refer to a leaching time of 7 days.

- Recently within the Swedish programme, Johnson et al., 2012 [8]: published two works performed at Studsvik and Paul Scherrer Institute (PSI). The work at Studsvik consisted of four commercials \( \text{UO}_2 \) SNF’s: Ringhals 3 (PWR, BU of 58.2 MWd/kgU), Ringhals 4 (PWR, BU of 61.4 MWd/kgU, Ringhals 3 (PWR, BU of 66.5 MWd/kgU and North Anna (PWR at North Anna NPP in Virginia, USA, BU of 75.4 MWd/kgU).
Two samples were cut from near the middle of each of the four fuel rods. A fuel corrosion sample consisting of a 20 mm segment was cut at mid-pellet height and contained one complete and two half pellets. These samples are referred to as closed rod samples. In another set of tests, referred to as open rod samples, adjacent fuel rod segments of 20 mm length were cut from each of the four rods and were weighed. The cladding was carefully sawn on both sides of the segment periphery and force was applied to the halves until the fuel broke away from the cladding. The two cladding halves, together with detached fuel fragments were collected in a glass vessel with glass filter bottom (100–160 µm pores) and weighed again. Then they were leached according to the same procedure as the closed rod samples. An initial solution sampling 2 h after test start was also carried out for all samples of this test series. The samples, kept in position by a platinum wire spiral, were exposed to 200 ml of synthetic Allard groundwater in a Pyrex flask. The pH was stable around 8.3 and carbonate concentration remained constant during all the experiments. The composition of the Allard water in mmol/L was: 0.45 Ca\(^+\), 0.18 Mg\(^+\), 0.1 K\(^+\), 2.84 Na\(^+\), 0.21 Si, 2.01 HCO\(_3\)\(^-\), 0.1 SO\(_4\)\(^2-\), 1.97 Cl\(^-\), 0.2 F\(^-\), 0.001 PO\(_4\)\(^3-\). The contact periods were 2 h, 7, 21 and 63 days. More information about these experiments can be found in [59].

The PSI work consisted of three commercial SNF specimens: Leibstadt (BWR, UO\(_2\) BU of 65.3 MWd/kgU) Gösgen (PWR, UO\(_2\) BU of 64 MWd/kgU), Gösgen (PWR, MOX BU of 63 MWd/kgU). The length of the fuel rod segments was 20 mm (two pellets) for each leaching experiments. For the rim samples the inner part of the fuel was removed mechanically by drilling (unintentionally somewhat off-center), leaving an asymmetric ring of fuel bonded to the cladding. One rim sample was left open, whereas a tight-fitting PVC plug was placed through the entire length of the other rim sample. In order to investigate if the fuel surface available for the attack of the leachant had a significant impact on the leach rate, a number of samples were broken into two halves, by cutting the cladding on opposite sides. Glass columns (total volume approx. 250 ml) with a sealed outlet tap for sampling and an implemented glass filter in order to retain solid particles were used. Approximately 200 ml of buffer solution (28 mM borate buffer, pH 8.5, containing 20 µg/g NaI as iodine carrier) were used per sample for the leaching experiments. As the objective of the measurements was to obtain the rapid release fraction of certain radionuclides, the experiments were performed in air-saturated buffer solutions. After filling the columns with the sample and buffer solution the supernatant air volume was removed through a hole in the piston cylinder which was closed afterwards to avoid additional air intake. Subsamples of 20 ml each were taken after 7, 14, 21, 28 and 56 days, whereas the last sampling of 30 ml per leach solution was performed after 98 days.

- Also within the Spanish programme, Clarens et al., 2009 [60], González-Robles, 2011 [61], Serrano-Purroy et al., 2012 [62]: used four commercial UO\(_2\) SNF’s: three from PWR with a BU of 48, 52, 60, MWd/kgU and one from a BWR with a BU of 53MWd/kgU. The temperature was (24 ± 6)°C.

From the SNF’s with a BU of 48 and 60MWd/kgU, three different SNF samples corresponding to the central axial position (labelled CORE), the periphery of the SNF
pellet (labelled OUT) and to an emptied cladding segment with small amounts of SNF attached to the inner wall called CLAD were prepared. In order to remove fines attached to the grain surface, the CORE and OUT powder SNF fractions were washed several times with acetone.

Static leaching experiments of powder samples, CORE, OUT and CLAD, were carried out in (50 ± 0.1) mL borosilicate glass test tubes with dimensions of length of 150 mm and a diameter of 25 mm with thread and a plastic screw cap (Schütt Labortechnik GmbH, Göttingen, Germany). The tubes were placed on a rotating stirrer (nominal speed of 30 rpm) to avoid concentration gradients that could influence the dissolution rate. Static leaching experiments were carried out with two synthetic leaching solutions: bicarbonate (BIC), which composition in mM was: 19 Cl⁻; 20 Na⁺; 1 HCO₃⁻, pH 7.4; and Bentonitic Granitic Groundwater (BGW), with following composition in mM: Cl⁻ 93.9; SO₄²⁻ 45.2; HCO₃⁻ 0.9; Na⁺ 117.9; K⁺ 1.1; Ca²⁺ 15.4; Mg²⁺ 17.3; pH of 7.6. The experiments were carried out under oxidising conditions and with about 0.25 g of SNF. [60-62].

Static experiments of pellet samples of the four SNF’s were performed in a (50 ± 0.1) mL flask and daily shaken for 5 to 10 minutes to avoid the risk of breaking the flask through the manual shaking [61].

The head space of the gas phase was (3 ± 1) mL for the powder sample experiments and (10 ± 1) mL for the pellet samples. To avoid initial U saturation and secondary phase formation, the solution was completely replenished two times at the beginning of each experiment.

The key conditions and results of all the reported experiments are given in Table 2 and Table 3.
Table 2. IRF showing the gap and the grain boundary (gb) contribution, in %, from different PWR SNF’s.

<table>
<thead>
<tr>
<th>SNF Id.</th>
<th>BU (MWd/kgU)</th>
<th>FG (%)</th>
<th>Sample</th>
<th>T (°C)</th>
<th>Solution</th>
<th>Time (days)</th>
<th>Rb gap</th>
<th>Rb gb</th>
<th>Cs gap</th>
<th>Cs gb</th>
<th>Sr gap</th>
<th>Sr gb</th>
<th>I gap</th>
<th>I gb</th>
<th>Mo gap</th>
<th>Mo gb</th>
<th>Tc gap</th>
<th>Tc gb</th>
</tr>
</thead>
<tbody>
<tr>
<td>PWR-HBR&lt;sup&gt;a,b&lt;/sup&gt;</td>
<td>31</td>
<td>0.2</td>
<td>bare fuel</td>
<td>25°</td>
<td>J-13</td>
<td>425</td>
<td>0.76</td>
<td>0.020</td>
<td>0.025</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.015</td>
<td>0.009</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>85°</td>
<td>J-13</td>
<td>425</td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.284</td>
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<td></td>
</tr>
<tr>
<td>PWR-TP&lt;sup&gt;a,b&lt;/sup&gt;</td>
<td>27</td>
<td>0.3</td>
<td>bare fuel</td>
<td>25°</td>
<td>J-13</td>
<td>376</td>
<td>0.29</td>
<td>0.016</td>
<td>0.012</td>
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<td>0.024</td>
<td>0.008</td>
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<td></td>
<td></td>
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<td>0.076</td>
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<tr>
<td>Ringhals-2&lt;sup&gt;c&lt;/sup&gt;</td>
<td>43</td>
<td>1.06</td>
<td>pellet</td>
<td>20-25</td>
<td>GW</td>
<td>7</td>
<td>0.85</td>
<td>0.04</td>
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<tr>
<td>ATM-103&lt;sup&gt;d&lt;/sup&gt;</td>
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<td>0.25</td>
<td>pellet</td>
<td>25</td>
<td>DIW</td>
<td>7</td>
<td>0.2</td>
<td>0.01</td>
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<td></td>
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<td></td>
<td></td>
<td>powder</td>
<td>25</td>
<td>0.1MHCl</td>
<td>60min</td>
<td>0.48</td>
<td>0.11</td>
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<tr>
<td>ATM-104&lt;sup&gt;d&lt;/sup&gt;</td>
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<td>1.1</td>
<td>pellet</td>
<td>25</td>
<td>DIW</td>
<td>7</td>
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| 48BU<sup>n,o</sup> | 48 | | pellet<sup>g</sup> | 24 ± 6 | HCO<sub>3</sub><sup>−</sup> 1mM | 10 | 1.59 | 6.25 | 0.05 | 0.05 | 0.005 |
| | | | powder<sup>g</sup> | 24 ± 6 | HCO<sub>3</sub><sup>−</sup> 1mM | 10 | 4.14<sup>i</sup> | 6.06<sup>i</sup> | 1.64<sup>i</sup> | 1.02<sup>i</sup> | 0.10<sup>i</sup> |
| | | | powder<sup>g</sup> | 24 ± 6 | HCO<sub>3</sub><sup>−</sup> 1mM | 10 | 1.31<sup>j</sup> | 4.45<sup>j</sup> | 0.44<sup>j</sup> | 0.04<sup>j</sup> | 0.002<sup>j</sup> |
| | | | cladding<sup>g</sup> | 24 ± 6 | HCO<sub>3</sub><sup>−</sup> 1mM | 21 | 0.01 | 0.001 | | | 0.009 |
| | | | powder<sup>g</sup> | 24 ± 6 | BGW | 10 | 2.91 | 5.23 | 1.26 | 1.06 | 0.09 |
| | | | powder<sup>g</sup> | 24 ± 6 | BGW | 10 | 0.37 | 4.24 | 0.31 | 0.01 | 0.01 |
| | | | cladding<sup>g</sup> | 24 ± 6 | BGW | 21 | 0.012 | 0.005 | | | 0.01 |
| 52BU<sup>n</sup> | 52 | | pellet | 24 ± 6 | HCO<sub>3</sub><sup>−</sup> 1mM | 10 | 0.85 | 3.20 | 0.08 | 0.03 | 0.01 |
| | | | powder<sup>g</sup> | 24 ± 6 | HCO<sub>3</sub><sup>−</sup> 1mM | 10 | 0.34 | 3.20 | 0.12 | 0.008 | 0.007 |
| 60BU<sup>n,o,p</sup> | 60 | 15 | pellet<sup>g</sup> | 24 ± 6 | HCO<sub>3</sub><sup>−</sup> 1mM | 10 | 1.98<sup>i</sup> | 2.45<sup>i</sup> | 1.96<sup>i</sup> | 0.93<sup>i</sup> | 0.31<sup>i</sup> |
| | | | powder<sup>g</sup> | 24 ± 6 | HCO<sub>3</sub><sup>−</sup> 1mM | 10 | 2.88<sup>i</sup> | 3.59<sup>i</sup> | 1.52<sup>i</sup> | 0.32<sup>i</sup> | 0.10<sup>i</sup> |
| | | | powder<sup>g</sup> | 24 ± 6 | BGW | 10 | 0.007 | 0.38 | 0.008 | | | 0.41 |
| | | | powder<sup>g</sup> | 24 ± 6 | BGW | 10 | 2.48 | 2.15 | 1.98 | 1.11 | 0.40 |
| | | | cladding<sup>g</sup> | 24 ± 6 | BGW | 21 | 2.56 | 4.26 | 1.89 | 0.51 | 0.03 |

<sup>a</sup>[39]; [42]<br><sup>b</sup>[41]; [44]<br><sup>c</sup>[45];[46]<br><sup>d</sup>[47]<br><sup>e</sup>[49]<br><sup>f</sup>[50]

(D-N°:5.1) – State of the art
Dissemination level : PU
Date of issue of this report : 31/08/2013
D-N°: 5.1 – State of the art
Dissemination level: PU
Date of issue of this report: 31/08/2013
Table 3. IRF (%) showing the gap and the grain boundary (gb) contribution from different BWR UO$_2$ SNF’s.

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<th>Fuel Id.</th>
<th>BU (MWD/kg)</th>
<th>FG</th>
<th>Sample</th>
<th>T (°C)</th>
<th>Solution</th>
<th>Time (days)</th>
<th>Rb gap</th>
<th>Rb gb</th>
<th>Cs gap</th>
<th>Cs gb</th>
<th>Sr gap</th>
<th>Sr gb</th>
<th>I gap</th>
<th>I gb</th>
<th>Mo gap</th>
<th>Mo gb</th>
<th>Tc gap</th>
<th>Tc gb</th>
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<td>pellet</td>
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<td>0.006</td>
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$^a$[45];$^b$[46]
$^c$[47]
$^d$[61]
$^e$[8]
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<th>Time (days)</th>
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<th>Cs gap</th>
<th>Cs gb</th>
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<th>Sr gb</th>
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*D-[49]  
*b-[50]  
*c-[51]  
*d-[54]  
*e-[8]*
Selected values from tables Table 2 and Table 3 are shown in figures 6 and 7. The data shown are selected taking into account the experimental conditions such as sample preparation, aqueous phase, the use of pellets, fragments or powders, etc.

Figure 7. Fast Cs release as function of burn-up.

Figure 8. Fission gas release as function of burn-up.

Figure 7 shows the Cs release and Figure 8 the fission gas release as function of burn-up for PWR and BWR fuels. FRG or Cs release of zero means that these data are not given in the related publications. Most of the experiments were performed with fuel having a burn-up below 50 GWD/tHM. In almost all cases, the Cs release is below 5%, only two measurements (not shown) reveal significantly higher values.

In total, the data show some scatter and especially in Fig. 7 two trends can be observed: one increasing at 40 GWD/tHM and another at 60 GWD/tHM. However, considering only those measurements where both FGR and Cs release have been determined, a different picture appears (Figure 9).
Neglecting all data points in the preceding tables where only FGR or Cs release is given, a linear correlation between the FGR and the release of Cs in the aqueous phase can be observed.

Variations in FGR and correlations with FP release can be important depending on the irradiation history of the fuel, the sample preparation and the leaching conditions, what makes difficult to extract generalized numbers accounting for IRF. What is clear is that complete information on burn-up and linear power rate, temperature reached, and evolution of the porosity of the fuel during the irradiation stages are important, among other factors, to derive reliable correlations and to understand apparent variations in the SNF IRF and FGR behaviour.

6. Selection of materials within FIRST-Nuclides

One of the first activities in the CP includes collection of fuel characterisation data from the beneficiaries. The fuel characterisation covers the type of reactor, its electrical power, types of fuel assemblies, manufacturer of the fuels and the discharge date of the fuel to be investigated. With respect to the cladding, the characterisation includes the material, diameter of rods, material thickness and the initial radial gap, the distance between pellet and cladding. The information for the pellet addresses the initial enrichment, geometry, grain size, density and specifics of the production process. The irradiation history covers the burn-up, the irradiation time and the number of cycles as well as the maximum and average linear power rate. Finally information on the fission gas release (FGR) was asked for. This information addressed several categories: (i) essential information representing the minimum data and information that should be available for the fuel chosen for the study, (ii) parameters and data which are not directly measured, but are derived from calculations, and (iii) supplemental information referring to characteristics that may be needed depending on the studies to be performed.
Spent fuel rods are owned mainly by the reactor operating utilities. KIT, JRC-ITU, JÜLICH, PSI, SCK-CEN, AEKI and Studsvik confirmed the access to the spent fuel material to be used in the project; they have the full rights to perform investigations and to publish the results. A summary of fuel material used during the CP FIRST-Nuclides is provided in Table 4 [52]. More information of the characteristics data of the fuel under investigation in CP FIRST-Nuclides can be found in the deliverable D1.1 of the project (it is available at the project website: www.firstnuclides.eu).

Table 4. Characteristic data of fuel under investigation in CP FIRST-Nuclides.

<table>
<thead>
<tr>
<th>Discharge Manufacturer</th>
<th>PWR</th>
<th>BWR</th>
<th>THTR / VVER</th>
</tr>
</thead>
<tbody>
<tr>
<td>AREVA</td>
<td>AREVA/Westinghouse</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cladding Material</td>
<td>Zry-4 – M5</td>
<td>Zry-2</td>
<td>Graphite / Zr1%Nb</td>
</tr>
<tr>
<td>Diameter Thickness</td>
<td>9.50 - 10.75 mm</td>
<td>9.84 - 10.2 mm</td>
<td></td>
</tr>
<tr>
<td></td>
<td>0.62 - 0.73 mm</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pellet Enrichment</td>
<td>3.80 – 4.94 %</td>
<td>3.30 -4.25 %</td>
<td>2.4 -16.8%</td>
</tr>
<tr>
<td>Grain size</td>
<td>5-40 µm</td>
<td>6 ≤ x ≤ 25 µm</td>
<td>20 -80 µm</td>
</tr>
<tr>
<td>Density</td>
<td>10.41 g cm⁻³</td>
<td>10.52 g cm⁻³</td>
<td>10.8 g cm⁻³</td>
</tr>
<tr>
<td>Specifics</td>
<td>standard,</td>
<td>standard and Al/Cr addition to UO₂</td>
<td></td>
</tr>
<tr>
<td></td>
<td>NIKUSI production</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Irradiation Burn-up</td>
<td>50.4 – 70.2 GWD/THM</td>
<td>48.3 – 57.5 GWD/THM</td>
<td></td>
</tr>
<tr>
<td>Nº cycles</td>
<td>2 - 14</td>
<td>5 – 7</td>
<td></td>
</tr>
<tr>
<td>Linear power average</td>
<td>186 -330 W/cm</td>
<td>160 W/cm</td>
<td>130 – 228 W/cm</td>
</tr>
<tr>
<td>FGR</td>
<td>4.9 – 23 %</td>
<td>1.2 – 3.1 %</td>
<td></td>
</tr>
</tbody>
</table>

Table 4 shows that selected materials fit into the range of present high burn-up fuels which need to be disposed of in Europe. Some non-standard materials are included, such as a fuel produced by a low temperature sintering process (NIKUSI), a fuel having Al and Cr additions and one high burn-up fuel kept for 14 cycles in a reactor. The other types of materials cover irradiated and unirradiated TRISO particles and the determination of the activity release from damaged and leaking VVER fuel rods.

Fuel elements studied in FIRST-Nuclides

BWR fuel from KKL Leibstadt (CH) and OL1 and OL2 from Olkiluoto (Finland) are used. KKL is a General Electric, Type BWR-6 reactor of 1245 MWel electric power. In this plant, fuel elements type Optima2 (Westinghouse) and Atrium XM (AREVA) are used. In
total, KKL needs 648 fuel elements consisting of 96 fuel rods in a $10 \times 10$ – 4 grid. Each fuel element contains 180 kg uranium. The OL1 and OL2 units at the Olkiluoto nuclear power plant are operated by TVO. They are two identical BWRs with a net electrical output 880 MW and 860 MW, respectively. The reactors in the OL1 and OL2 power plant units each contain 500 fuel assemblies GE-14 type in a $10 \times 10$ raster. Each fuel element contains 91–96 fuel rods including 12 rods doped with Gd. The mass of uranium per fuel assembly amounts to 175 kg [12].

**PWR fuel** from KKG Gösgen, CH: KKG uses both UO$_2$ and MOX fuel. In the CP FIRST-Nuclides, only UO$_2$ fuel will be considered in this project. KKG utilises 177 fuel assemblies, with an overall weight of 666 kg per assembly. Each assembly consists of 205 fuel rods. Presently, the initial $^{235}$U enrichment is 4.3 wt. %.

**VVER fuel**: VVER-440 power reactors are pressurized water reactor using water as coolant and neutron moderator. The Loviisa nuclear power plant is operated by Fortum Oy and consists of two VVER-440 units, Loviisa 1 and Loviisa 2 of 488 MW. In the German Democratic Republic several VVER-440 reactors were in operation until 1990. In total, 5050 fuel assemblies of VVER-440 reactors have to be disposed of in Germany. The Hungarian power plants Paks 1-4 are also of the VVER-440 type. Nuclear fuel for VVER-440 reactors is manufactured and delivered in the form of fuel assemblies. The core of VVER-440 reactor is loaded with fuel assemblies and control fuel assemblies consisting of the fuel follower of control rod and absorbing extension. Compared to the 4-year fuel cycle, 5-year fuel cycles provide for an additional decrease in natural uranium consumption and number of manufactured, transported and stored fuel assemblies. U-Gd fuel is widely used for this fuel cycle (http://www.elemash.ru/en/production/Products/NFCP/VVER440/).

7. **Objectives of FIRST-Nuclides in the context of previous investigations**

The objectives of the CP FIRST-Nuclides are defined in order to answer to open questions raised in the previous projects and publications [1, 2], [3], [4], [5-7] and [8]. The objectives of the CP are directed towards an improved understanding the fast / instant release of long-lived radionuclides from high burn-up spent UO$_2$ fuels from LWRs in geological disposals. A very important aim is the elaboration of a common definition of “First / Instant Release”. As already mentioned at the onset of this report the “fast and instant release fraction” is an operational definition that has its direct translation within the safety analysis of a disposal concept. The long-term radionuclide release from spent nuclear fuel under reducing disposal conditions, however, is promoted by $\alpha$-radiolytically induced oxidation processes and impeded by the effect of hydrogen. The definition to be developed should include (i) needs of the project, (ii) delineation to the long-term dissolution processes and (iii) conversion of the short-term experiments and modelling to a “fast and instant release fraction” relevant for long-term safety.
WP 1 includes a discussion of fuel samples and sample preparation selected for the experiments. The sample sizes fragments or pellets or presence or in absence of cladding have significant impact on the results. It must be also considered, that the fuel rods of several fuel elements including different burn-up or Gd$_2$O$_3$ doped material may be disposed together.

The experimental investigations include the release of gaseous and non-gaseous fission products from high burn-up UO$_2$ fuel (~ 60 GWd/tHM) and establishing correlations between the fission gas release (FGR) and the release of non-gaseous fission products. A recent study by Johnson et al. [8] demonstrates the importance of the linear power on the FGR. For this reason, it is beneficial for the project to investigate SF from different reactors and different operation strategies, such as the number of irradiation cycles.

The investigations aim at reducing uncertainties with respect to $^{129}$I, $^{135}$Cs, $^{79}$Se and $^{14}$C release and speciation, and will provide for improved data for these isotopes. The chemical form of the relevant elements requires also specific investigations, as the migration and retention behaviour depends strongly on their cationic or anionic character.

The experimental work is accompanied by modelling. The prime aim of the modelling is to improve the database for the fast/instant release fraction of the long-lived fission and activation products from high burn-up spent UO$_2$ fuel. Modelling will contribute by up-front assessment of the experimental boundary conditions, the coupling of experimental results with model development and the impact of different experimental findings on the refinement of fast/instant release prediction capability. Thereby, up-scaling from the analytical and modelling micro-scale to the experimental bulk observations and the release on a fuel-rod scale is a key challenge.

As indicated, the compilation of new data within the project will contribute to obtain a more complete picture on the IRF of SNF, especially on high BU fuels for which only in the most recent years data have been obtained. In the following section, the selection of materials to test in the project is presented. At the time of preparation of this updated version of the report, data produced within the project had not been reported yet. A new update of this report is foreseen after the 2$^{nd}$ Annual Workshop, where some results will be presented that will be put in context with the existing information available and will set the basis for the construction of a more complete IRF database aimed at obtaining empirical relationships between FGR and “instantly released” FPs, contributing to improving the conceptual model for the explanation of this behaviour and provide for the possibility of model testing and development.

8. References


[12] TVO, Nuclear power plant units Olkiluoto 1 and Olkiluoto 2, in, TVO Nuclear Services Oy, Olkiluoto, , FI-27160 EURAJOKI, FINLAND.


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APPENDIX I: MANUFACTURERS OF UO₂ FUEL AND FUEL ELEMENTS

Nuclear fuel elements are produced for energy production in nuclear power plants. The fuels to be considered in the CP FIRST-Nuclides are uranium oxide fuels having initial ²³⁵U enrichments up to 4.3 wt.%. An overview of fuel elements for the different types of reactors was published by [11]. The nuclear fuel used in Europe is mainly produced by subsidiaries of AREVA or Westinghouse.

**FBFC** (French acronym for Franco-Belgian Fuel Fabrication), a subsidiary of AREVA comprises three sites at Romans (F), Pierrelatte, Tricastin (F), and Dessel (B). The **Romans site** transforms uranium hexafluoride supplied by EURODIF into uranium oxide powder (UO₂). It also fabricates uranium pellets, rods, nozzles, and fuel assemblies for pressurized water reactors (PWRs). FBFC Romans employs 830 persons. The **Pierrelatte site** manufactures support grids for assemblies for PWR fuels and Harmoni™ control clusters. These products are then supplied to the Romans and Dessel plants, which fabricate fuel assemblies. It also produces spacers for MELOX's MOX fuel assemblies. **FBFC International** located in **Dessel**, Belgium, produces fuel assemblies for pressurized water reactors. The plant also fabricates pellets with gadolinium, rods, plugs, and spring packs for fuel assemblies for pressurized water reactors and boiling water reactors. It also assembles the various components for MOX fuel assemblies. At the Tricastin site, EURODIF Production operates the Georges Besse plant performing uranium enrichment by gaseous diffusion. With the new Georges Besse II plant in operation since 2009, AREVA uses the centrifugation technology for enrichment.

**Advanced Nuclear Fuels GmbH** (ANF) is a full subsidiary of AREVA. Its headquarters are in Lingen (Germany) and its operations are distributed over three sites in Germany. The plant in **Lingen** produces UO₂ powder, as well as pellets, rods, and fuel assemblies for pressurized and boiling water reactors. Since the site's commissioning in 1977, it has produced more than 20,000 fuel elements. Following stages in assembly fabrication are performed at the Lingen plant: uranium hexafluoride (UF₆) is transformed into uranium oxide (UO₂). This oxide is compacted into cylindrical pellets and baked in ultra-high-temperature furnaces. The pellets are then stacked into tubes of around 4 meters in length, called "rods". These are then sealed off at the ends. The zirconium-alloy rod cladding is subjected to rigorous testing. The rods are grouped into assemblies with the appropriate dimensions.

**Westinghouse Sweden Nuclear Fuel Factory**, situated in Västerås, Sweden. The fuel factory manufactures fuel assemblies for PWRs and BWRs, and fuel channels and control rods for BWRs. The factory in Västerås is responsible for the entire chain from research and development to manufacturing of nuclear fuel, as well as for control rods and fuel channels for BWR plants including codes for core surveillance. The present fuel factory has been in operation since 1971 and was continuously expanded and modernized. The factory produces approximately 400 tons of UO₂ fuel for BWRs and PWRs per year. In the conversion of UF₆ into UO₂ powder, the capacity as well as the plant license is limited to 600 tons UO₂.
British Nuclear Fuels Ltd (BNFL) was the manufacturer fuel elements feeding the British Energy’s (BE) AGR reactors as well as BNFL’s own Magnox Reactors. Advanced gas-cooled reactor fuel (AGR) comprises of 36 stainless steel rods each containing 64 pellets, grouped together inside a graphite ‘sleeve’ to form a ‘fuel assembly’. BNFL also made fuel for the older Magnox Reactors which consists of a natural uranium metal bar with a magnesium alloy casing. BNFL’s core expertise for fuel manufacture was based on fabrication of the uranium pellets, and the final assembly of the fuel elements. BNFL was finally abolished in 2010.

APPENDIX II: CANISTER/DISPOSAL CONCEPTS

Germany
In Germany, the disposal concept for spent nuclear fuel, the so called "direct disposal of spent fuel" was developed and examined with respect to safety aspects. The reference concept is based on the triple purpose cask POLLUX for transport, storage and final disposal as well as a conditioning technique that separates fuel rods from the structural parts of the fuel assemblies. Another most promising option is called BSK 3-concept. Both concepts are described in the literature [29]. The POLLUX canister consists of a shielding cask with a screwed lid and an inner cask with bolted primary and welded secondary lid. The inner cask consists of fine-grained steel 15 MnNi 6.3, the thickness of the cylindrical wall is 160 mm according the mechanical and shielding requirements. The outer cask provides shielding. Its thickness is 265 mm and it consists of cast iron GGG 40 (DIN 1693-1/2 steel grade). The weight of the inner cask (including spent fuel is 31 ton, the weight of the outer cask is 34 ton. Ten complete fuel elements can be packed into a POLLUX cask. Another possibility is the accommodation of consolidated fuel rods (5.4 tHM). The BSK 3 cask was designed for accommodation of consolidated fuel rods. The capacity of a BSK 3 is designed for three PWR fuel elements or nine BWR fuel elements. The wall thickness of the BSK 3 is 50 mm.

Sweden
The Swedish/Finnish Canister concept bases on an insert of nodular iron (a kind of cast iron) which is inserted into a copper tube (FigA 1). The copper lids are welded by friction stir welding. The canisters are about 5 meters long and have a diameter of more than one meter. When the canister is filled with 12 spent fuel elements, it weighs between 25 and 27 metric tons. (SKB Brochure “Encapsulation, When, where, how and why?”).

Belgium
In Belgium, the Supercontainer was developed which surrounds the steel cask containing the fuel elements by a thick concrete overpack [30]. The supercontainer is intended for the disposal of (vitrified) high level heat-emitting waste and for the disposal of spent fuel assemblies. In this concept, the spent fuel assemblies are enclosed in a carbon steel overpack
of about 30 mm thick. This overpack has to prevent contact of the waste with the water coming from the host formation during the thermal phase of several 1000 years for the spent fuels assemblies. For corrosion protection, the overpack is enveloped by a concrete buffer of about 70 cm thickness. The concrete is surrounded by a stainless steel cylindrical envelope (called liner). The outside radius of the supercontainer for spent fuel assemblies is about 1.9 m, and a length of about 6 m, it’s mass is about 54 tons [31].

![Image](image.png)

**FigA 1.** Components of the Swedish canister concept at SKB’s Canister Laboratory in Oskarshamn.

In all three concepts, corrosion of the fine-grained steel 15 MnNi 6.3 (Germany), nodular iron (Sweden, Finland) or carbon steel (Belgium) is a prerequisite before groundwater may come into contact with the fuel. Different corrosion processes may therefore be relevant. The three examples listed above all show a general corrosion due to an active corrosion mechanism. Only in the Belgian case, a passivation of the steel may occur. Under the reducing conditions of a deep underground disposal, anaerobic corrosion of the steel takes place. This active corrosion process normally shows a non-uniform reduction of the canister thickness, especially if there are gradients in material or mineralogical composition or in the welding material. The degree of non-uniformity in thickness reduction of the actively corroding steels was observed to be a factor of 2, in comparison to the general corrosion rate [32]. However, in the case of a heat-affected zone close to a welding, the corrosion rate may increase. For example, FigA 2 shows the corrosion of the welding of fine-grained steel in MgCl₂ brine at 150°C under γ-irradiation (10 Gy/h).
FigA 2. Corrosion of the welding of fine-grained steel 1.0566 (FStE 355) in MgCl₂ brine at 150°C under γ-irradiation (10 Gy/h) [32].