Gamma Spectroscopy and Gamma Emission Tomography for Fuel Performance Characterization of Irradiated Nuclear Fuel Assemblies
Abstract


Gamma spectroscopy and gamma emission tomography are two non-destructive measurement techniques for assessing the performance of nuclear fuel which have been investigated in this thesis for existing and novel applications through theoretical studies and experimental demonstrations. For assessment of individual fuel rods using gamma spectroscopy, fuel assemblies are dismantled so that the fuel rods may be measured separately, which is time-consuming and may cause damage to the fuel. Gamma tomography is more seldom used, but its application on complete fuel assemblies would enable the assessment of individual fuel rods without the need to disassemble the fuel. Both techniques are based on recording gamma rays, emitted at characteristic energies from decaying radioactive products in the fuel.

The feasibility of measuring short-lived fission gasses in the gas plenum of fuel rods with short cooling time was experimentally investigated. Based on the feasibility demonstration, a method was proposed and experimentally demonstrated for determining the fission gas release fraction of $^{133}$Xe in fuel rods with short cooling time. Additionally, a method for investigating the origin of released fission gasses based on the measured ratio of $^{133}$Xe/$^{85}$Kr in the fuel rod gas plenum was demonstrated. These methods may be employed at research reactors, where fuel with short cooling time is available for measurement.

A gamma emission tomography instrument has been designed, constructed and experimentally demonstrated on a Halden Reactor fuel assembly. Simulation studies showed that the instrument and the tomographic reconstruction methods employed may be useful for: identifying a leaking fuel rod in an assembly by its lack of fission gas content; reconstruction of the rod-wise fission product distributions in the fuel stack and plenum regions of the assembly; and determining the rod-wise fission gas release fractions.

In the experimental demonstration, the rod-wise distributions of the fission products $^{137}$Cs and $^{85}$Kr in the fuel stack and plenum regions of the assembly were reconstructed, as well as the distributions of the activation products $^{60}$Co and $^{178m}$Hf in the plenum region, revealing the plenum springs and tie rods, respectively. The reconstructed data was in the form of images, useful for qualitative assessment of the fuel.

Keywords: Nuclear fuel performance, Nuclear fuel assemblies, Gamma emission tomography, Gamma spectroscopy

Scott Holcombe, Department of Physics and Astronomy, Applied Nuclear Physics, Box 516, Uppsala University, SE-751 20 Uppsala, Sweden.

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ISSN 1651-6214
ISBN 978-91-554-9099-7
urn:nbn:se:uu:diva-235124 (http://urn.kb.se/resolve?urn=urn:nbn:se:uu:diva-235124)
to Heather
Cover page:
Tomographic images of isotope distributions in a fuel assembly from the Halden research reactor.
- Top left: Distribution of $^{137}$Cs in the fuel stack, showing its migration to the pellet surfaces
- Top right: Distribution of the gaseous fission product $^{85}$Kr in the gas plenum region
- Bottom left: Distribution of $^{60}$Co in the plenum region, showing neutron activation of the plenum springs
- Bottom right: Distribution of $^{178m}$Hf in the plenum region, showing neutron activation of the tie rods
List of Papers

This thesis is based on the following papers, which are referred to in the text by their Roman numerals. Reprints were made with permission from the respective publishers.


This paper investigates the feasibility of measuring short-lived fission gas isotopes in the gas plena of individual fuel rods to determine the release fraction of the selected isotope. My contribution: I investigated physical properties of Krypton and Xenon isotopes, performed MCNP simulations of the measurements, and analyzed experimental gamma spectroscopy measurement data. I was the main author of the paper.


This paper describes simulations of gamma emission tomography measurements on fuel assemblies and reconstruction of the data in order to locate a failed fuel rod in the assembly, which is identified by its missing fission gas inventory. My contribution: I created the MCNP models and performed the simulations. I applied tomographic reconstruction techniques to generate images of the fuel from the simulated measurements. I was the main author of the paper.

This paper describes a method for investigating the origin of fission gasses released from the pellet stack of irradiated fuel rods based on non-destructive gamma spectroscopy measurements of long-lived and short-live fission products in the fuel stack and gas plenum regions. My contribution: I developed the method and applied it to experimental data. I was the main author of the paper.


This paper describes a method for determining the fission gas release fraction of a short-lived fission gas isotope based on gamma spectroscopy measurements, enabling the analysis of fission gas release over time. My contribution: I developed the method and applied it to experimental data. I was the main author of the paper.


This paper describes gamma-ray spectroscopy measurements of nine Halden reactor driver fuel rods. My contribution: I performed the measurements and analyzed the results. I was the main author of the paper.


This paper describes simulated gamma emission tomography measurements of the fuel-stack and gas plenum regions of a Halden reactor fuel assembly. My contribution: I created the MCNP models and performed the simulations. I applied tomographic reconstruction techniques to obtain rod-wise data from the simulated measurements. I was the main author of the paper.
This paper describes analytic and algebraic tomographic algorithms for reconstructing data from gamma tomography measurements on nuclear fuel assemblies. Methods for accounting for gamma-ray attenuation are also introduced into the analytic and algebraic methods. The algorithms were applied to simulated gamma-ray intensities. *My contribution:* I created the MCNP models and performed the simulations. I applied the analytic tomographic reconstruction techniques to generate images from the simulated measurements. I developed the method for attenuation compensation in the analytic reconstruction technique.

This paper describes the gamma emission tomography instrument developed at the Halden reactor, and initial measurement results are presented. *My contribution:* I specified the design of the equipment and the measurement sequence. I reconstructed the experimental data to generate images of the rod-wise distribution of selected fission products and activation products in the fuel-stack and plenum regions of the measured fuel assembly. I was the main author of the paper.

All conference papers (I, III, V and VI) have undergone and passed a peer-review procedure similar to the journal papers. The following are additional papers/reports by the author, which are not included in the thesis:

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Abbreviations

ART  Algebraic Reconstruction Techniques
ASIRT  Additive Simultaneous Iterative Reconstruction Technique
BWR  Boiling Water Reactor
FBP  Filtered Backprojection
FGR  Fission Gas Release
FWHM  Full Width at Half Maximum
GW(e)  Giga-watts electric
HBWR  Halden Boiling Water Reactor
HPGe  High-purity Germanium
HRP  Halden Reactor Project
IAEA  International Atomic Energy Agency
kEV  Kilo electron volt
LHR  Linear Heat Rate
LOCA  Loss of Coolant Accident
LVDT  Linear Variable Differential Transformer
MCA  Multi-channel analyzer
MCNP  Monte Carlo n-particle (simulation software)
MWd/kgUO₂  Megawatt days per kilogram Uranium dioxide
NPP  Nuclear Power Plant
OECD  Organization for Economically Developed Countries
PIE  Post Irradiation Examination
PWR  Pressurized Water Reactor
RIA  Reactivity Initiated Accident
SFSP  Spent Fuel Storage Pool
SPECT  Single-photon Emission Tomography
UO₂  Uranium dioxide
%FGR  Percent Fission Gas Release
1. Introduction

Currently, there are 434 nuclear power reactors operating in the world, with another 72 under construction [1]. Together, these Nuclear Power Plants (NPPs) produce 372 GW(e), which accounts for approximately 11% of the world’s electricity production [2]. In addition to these commercial NPPs, there are currently 266 operating research reactors (including 19 which are temporarily shut down) with 6 more under construction [3]. Rather than being used for electricity production, research reactors are instead used for a variety of other research, education, and industrial purposes.

The safe and efficient operation of NPPs is of continued importance for existing and future reactors. The operating limits of commercial NPPs as well as research reactors are, to a large extent, determined by the properties and behavior of the nuclear fuel. Furthermore, mitigation of the consequences from possible accidents requires additional knowledge with respect to beyond-design limits. As such, increased understanding of nuclear fuel behavior during both normal operation and accident scenarios leads to enhanced safety and more efficient use of the fuel. As a result of the latter, less amounts of high level waste are produced.

1.1. Operation of Nuclear Power Plants

The primary use of commercial NPPs is to produce electricity, whereas research reactors are used for a variety of purposes such as isotope production, training and education, neutron research, and materials testing. Of the world’s 266 operating research reactors, 63 are utilized for the latter purpose, which is largely in support of commercial NPPs.

Among existing commercial NPPs, as well as those planned or under construction, there are many variants of nuclear plant design [1], [2], [3], [4], and [5]; however, at the most basic level, the operation of all NPPs is based on fission reactions occurring in the nuclear fuel. When fission occurs, energy is released within the fuel which is imparted, in the form of heat, to the reactor coolant as it circulates through the reactor core.

In most NPPs (353) the heated coolant in the primary circuit (which contains the reactor core) passes its heat energy to water in a secondary coolant circuit via heat exchangers (called steam generators). The water coolant in the secondary loop is turned to steam and used to drive the steam turbines
and generators. Of the NPPs with multiple coolant circuits, 336 utilize water as the coolant, 15 are cooled with CO₂ gas, and two are cooled with liquid sodium. Figure 1 shows a schematic overview of a Pressurized Water Reactor (PWR) - the most common reactor type, of which there are 273 currently operating.

Figure 1. Schematic diagram of a Pressurized Water Reactor (PWR) – the most common NPP reactor type currently in operation. (NRC file image – public domain)

In the remaining 81 currently-operating reactors, there is only one coolant circuit, where the water which passes through the core, turns to steam, and drives the steam turbines and generator directly. These reactors are referred to as Boiling Water Reactors (BWRs). Figure 2 shows a schematic overview of a BWR – the second most common reactor type.

Figure 2. Schematic diagram of a Boiling Water Reactor (BWR) – the second most common NPP reactor type in operation. (NRC file image – public domain)
Understanding behavior of the fuel is of central importance for operating a nuclear plant since the fuel provides the heat that powers the plant, and since the fuel contains the radioactive fission products that result from fission. In fact, the operation of the plant is done such that the maximum amount of energy may be extracted from the fuel while ensuring that the fuel is maintained in a safe condition.

Research reactors are utilized to continuously validate the understanding of fuel performance and to evaluate fuel performance under various conditions. Results of investigations conducted at research reactors are carried over to operation of commercial NPPs in the form of validated or adjusted safety and operating margins. Research determines how the fuel may safely and efficiently be operated, and knowledge is continuously obtained which helps to improve fuel performance and efficiency.

The fuel and methods discussed in this work are primarily for PWRs and BWRs; however the methods are generally applicable to other reactor fuels which have similar geometries and materials compositions. Several of the methods that were developed have been demonstrated at the Halden Boiling Water Reactor (HBWR) – a research reactor located in Halden, Norway.

1.2. Utilizing Gamma Rays to Assess Nuclear Fuel

While there are many nuclear plant design variants, the fuel used in all currently operating NPPs is primarily Uranium Dioxide (UO₂). As a result, many of the fuel behavior characteristics which are of interest are common for all reactor types, regardless of the specific reactor design. Specifically, the heavy nuclei in UO₂ fuel undergo fission, producing stable as well as unstable isotopes, called fission products. Analyzing the content and spatial distribution of these fission products within selected regions of the fuel reveals information about the in-reactor operating conditions of the fuel.

The unstable fission products undergo radioactive decay, emitting gamma rays with energies characteristic of the specific isotopes from which they are emitted. These gamma rays may be detected outside the fuel where gamma rays are sorted according to their energy into a gamma-ray spectrum. Figure 3 shows an example gamma-ray spectrum from the gamma radiation field surrounding an irradiated nuclear fuel assembly. Peaks in the gamma-ray spectrum shown in Figure 3 correspond to specific isotopes in the fuel. As an example, the 662 keV peak emitted in the decay of ¹³⁷Cs is highlighted in the figure.
Figure 3. Example of a gamma-ray spectrum obtained when recording the gamma-radiation field surrounding an irradiated nuclear fuel assembly. Many fission products emit gamma radiation at characteristic energies upon decay. As an example, the 662 keV peak from $^{137}$Cs is identified.

The measured intensities in the characteristic peaks are used to determine the content of the gamma-ray emitting isotopes in the fuel, based on radioactive decay properties of the isotopes and the detection efficiency of the measurement system. When the content of selected fission products in the fuel is known, knowledge of these isotopes’ physical properties is exploited to deduce characteristics of the fuel and its performance in the reactor core. Some examples of fuel properties and isotopic markers are presented in section 4.3.
2. Nuclear Fuel

At a basic level, the nuclear fuel used in NPPs has many design characteristics in common, regardless of specific type NPP. The fuel typically consists of UO$_2$ pellets inside metal tubes, which are bundled together into a fuel assembly. The metal tubes – called fuel rod cladding – separate the fuel pellets from the reactor coolant/moderator. The fuel pellets and fuel rod cladding serve important purposes with regard to operating NPPs.

The ceramic UO$_2$ fuel pellets are chemically stable, can withstand high temperatures, and contain the radioactive fission products. The fuel cladding is chosen to have favorable neutronic properties, and must also withstand reactor temperatures and exposure to the reactor coolant. The fuel assemblies must also be maintained in the proper geometry in order to sustain the controlled nuclear reactions in the core.

Due to their critical function in maintaining the fuel in the proper geometry and acting as the first barriers to the release of radioactive fission products, the fuel pellets and cladding and their performance dictate many of the operating parameters and limitations of NPPs. If the fuel cladding integrity is not maintained, fuel and fission products may be released into the reactor coolant circuit, contaminating the coolant loop, and potentially exposing plant workers or the general public to unnecessary radiation doses. In the worst case, the fuel geometry may be degraded to the extent that the nuclear reactions are no longer controllable, possibly leading to fuel melting and unreparable damage to the NPP.

2.1. Mechanical Design

The UO$_2$ fuel pellets are approximately 1 cm in diameter and 1 cm in height, where the specific dimensions vary according to the reactor and fuel design. The fuel pellets are stacked inside metal cladding tubes which are pressurized with an inert gas (typically He) and sealed at both ends with end-plugs. Fuel rod design typically incorporates a gas plenum volume which collects gaseous fission products, called fission gasses, and allows the fuel rod internal pressure to remain at an acceptable level throughout the fuel rod’s lifetime. In the plenum, there is generally a plenum spring, which keeps the pellets in their vertical position while still allowing for swelling of the fuel pellet stack. The pellet stack in PWR and BWR reactors is approximately 4
m in length. Fuel rods used in fuel assemblies in the HBWR research reactor are similar to commercial fuel rods with the exception that HBWR fuel rods have a pellet stack length of approximately 1 m. Figure 4 (Left) shows a side, cutaway view of a typical fuel rod.

Fuel rods are bundled together into fuel assemblies, where the number and configuration of fuel rods varies depend on the reactor type, and fuel design. Schematic views of some PWR, BWR, and HBWR fuel assembly designs are shown in Figure 4 (Right).

![Figure 4](image)

*Figure 4, Left: Side, cut-away view of a typical fuel rod; Right: schematic views of PWR, BWR, and HBWR fuel assemblies. Note that the fuel rod and fuel assembly images are not to scale. The axial length of the PWR and BWR assemblies are similar at about 4 m, whereas the fuel rods in HBWR assemblies are only about 1 m long. A PWR assembly contains about 200-300 rods, a BWR assembly contains about 100 rods and an HBWR assembly contains about 40 rods or less.*

In BWRs and PWRs, spacer grids, at several axial positions in the fuel assembly, maintain the as-designed spacing between the fuel rods. Tie plates are located at the top and bottom of PWR and BWR fuel assemblies and provide a means for positioning and handling the fuel. Special rods connect the upper and lower tie plates. In PWR assemblies these rods are hollow tubes called guide tubes, in BWR assemblies they are special fuel rods called tie-rods, and in HBWR fuel assemblies they are solid rods which are also called tie rods. BWR fuel assemblies are surrounded by fuel channels which direct the coolant flow through and around the assemblies, which is im-
important when boiling occurs in the coolant. A similar channel, called a shroud, is used in HBWR fuel assemblies.

PWR and BWR fuel assemblies utilize a square lattice pattern for the fuel rods, where PWR fuel assemblies measure approximately 21 cm x 21 cm, and BWR fuel assemblies measure approximately 14 cm x 14 cm. Figure 5 (Left) and Figure 5 (Right) show a 17x17 PWR fuel assembly with 264 fuel rods, and a 10x10 BWR fuel assembly with 96 fuel rods.

Figure 5, Left: Cross-section view of a 17x17 PWR fuel assembly with 264 fuel rods; Right: Cross-section view of a 10x10 SVEA-96 BWR fuel assembly with 96 fuel rods. In the BWR assembly, the rods are surrounded by a fuel channel, which contains the two-phase flow of water and steam in this reactor type.

Since the HBWR is a research reactor, the fuel assembly geometry may vary considerably depending on the intended use of the fuel. Fuel assemblies are generally in a circular lattice pattern, where one to 37 fuel rods are contained in a circular shroud that channels the two-phase (boiling) coolant flow. The maximum diameter of an HBWR fuel assembly (the shroud) is 7.3 cm. Figure 6 shows a cross section view of three HBWR fuel assembly designs comprised of 37 fuel rods and three tie rods (left), 13 fuel rods and four tie rods (middle) and 9 fuel rods and four tie rods (right). Fuel rods are illustrated as dark grey and tie rods are illustrated in light grey shade in Figure 6.
Figure 6. Cross-section view of three HBWR fuel assembly designs comprised of 37 fuel rods (left), 13 fuel rods (middle), and 9 fuel rods (right), surrounded by a circular shroud. To maintain assembly geometry these designs also contain 3, 4 and 4 metallic tie rods, respectively, illustrated in light grey.

Materials used in PWR, BWR and HBWR fuel assemblies have been chosen to optimize the in-reactor performance of the fuel. In order to reduce parasitic thermal neutron absorption, fuel cladding materials with a low neutron absorption cross section have been selected. Specifically, the fuel rod cladding and fuel assembly structural material are made primarily of zirconium alloys, called Zircaloy. Tie plates are typically made of stainless steel, and other components such as spacer grids and plenum springs may be made of Inconel alloys.

2.2. Irradiated Fuel Properties

The fuel rods are the source of energy in a nuclear reactor, where the energy is manifested in the form of heat. The rate of energy generation in the fuel is dependent on the local neutron flux and is expressed in terms of power per fuel rod length. Together with the thermal conductivity, the heat generation rate in the fuel governs the temperature in the fuel, and it is referred to as the Linear Heat Rate (LHR). LHR is commonly expressed in terms of kilowatts per meter (kW/m). The total amount of energy generated in the fuel over time is referred to as burnup and is commonly expressed in terms of megawatt-days per kilogram of UO₂ (MWd/kgUO₂).

When the fuel is manufactured, the fuel pellets and fuel rod cladding have carefully controlled compositions, mechanical properties and dimensions; however, these change as functions of burnup and LHR as the fuel is irradiated in a reactor. Within the reactor, the neutron flux is not uniform, and consequently the burnup and LHR are not uniform throughout the reactor. For example, the neutron flux is depressed at the boundaries of the core (due to neutron leakage), and near inserted control rods (due to neutron absorption). Figure 7 shows a representative axial distribution of fast and thermal neutron flux in the HBWR reactor core and Figure 8 shows a representative radial distribution of fast and thermal neutron flux in the HBWR reactor core.
Rapid changes in the neutron flux, so-called transients, may cause mechanical interaction between the fuel pellets and the cladding, which may damage the cladding as well as the pellets. Furthermore, the properties of the fuel pellets depend to a high degree on the burnup of the fuel. This is further described in the following sections.
2.2.1. Pellets

At their Beginning of Life (BOL), fuel pellets are solid, cylindrical, and composed of nearly pure UO₂. When the fuel is placed in a reactor and is irradiated, the uranium fuel absorbs neutrons. Some uranium nuclei undergo fission, resulting in the splitting of the uranium nuclei and the production of lighter isotopes called *fission products*. However, not all uranium nuclei fission at the event of neutron absorption, and thus the irradiation of neutrons in the reactor core also leads to the build-up of heavier elements, such as heavier isotopes of uranium and also heavier elements such as e.g. plutonium, which in turn may undergo fission when irradiated further. These heavier elements are called *transuranic elements*. Lighter elements present in the fuel materials may also absorb neutrons and form new nuclei. These are often radioactive and are called *activation products*. Ultimately, hundreds of other isotopes are produced, altering the chemical composition of the fuel [6].

The ceramic fuel pellets have relatively poor thermal conductivity, thus the power deposited from fission in the interior of the pellets is not conducted efficiently to the outer radius of the pellet and eventually to the reactor coolant. This leads to a large temperature gradient (which can be several hundred degrees) over the radius of the pellet (~5 mm) [7].

The temperature gradient across the pellets and the appearance of fission products and transuranic elements in the fuel has several important effects on the fuel properties and behavior [8], some of which are listed below:

- **Radioactivity**: Many transuranic elements, activation products, and fission products are radioactive, causing high levels of radiation around the fuel.
- **Density**: Initially, the density is increased during irradiation, but with increasing burnup, fission products cause the pellets to become less dense and swell in size.
- **Cracking**: The high temperature gradient across the radius of the pellets leads to cracking from thermal stress.
- **Fission Gas Release (FGR)**: Some fission products are gaseous and may escape the pellets. FGR is influenced by the inventory of these so-called fission gasses, burnup, pellet cracking, and in particular, the temperature plays an important role [9].
- **Varying spatial composition**: Transuranic elements are produced to a higher degree near the pellet surface and at high temperatures volatile fission products may migrate during irradiation, thus the chemical composition varies radially across the pellet. Figure 9 shows the radial distributions of Plutonium, Uranium, and Oxygen over the diameter of a fuel pellet. Note the elevated Pu content and depleted U content at the pellet outer surface compared to the interior of the pellet.
- **High burnup structure (HBS):** At high local burnups (above 50 MWd/kgUO₂), the HBS begins to form at the pellet outer surface. The HBS does not share the grain structure of the rest of the pellet, and is also characterized by the absence of fission gasses in the matrix (the UO₂ material), where the gasses are instead present in pores within the HBS [10]. This is conceptually illustrated in Figure 10.

*Figure 9.* Radial distribution of Plutonium, Uranium and Oxygen across the diameter of a fuel pellet. Note the elevated Pu content and depleted U content near the pellet outer surface. This figure is from [11] and used with permission.

*Figure 10.* Conceptual illustration of the radial distribution of Xenon within a fuel pellet. Note the drop off in Xe content in the matrix near the rim of the pellet, where the Xe is instead contained in the pores of the HBS. This artificial data conceptually shows the effect seen in Post Irradiation Examinations [12], [13]. Note also that the x-axis extends from the relative radius of 0.7 to 1.
2.2.2. Cladding

The fuel cladding mechanical properties are altered through exposure to the neutron flux, mechanical interaction with the fuel pellets, and through chemical interaction with the coolant and fission products in the fuel pellets. Some examples of how the cladding properties change with irradiation are:

- **Changes in diameter and length**: [14] At BOL, there is a gap between the pellet and the cladding, but the pressure difference between the reactor coolant and the internal pressure of the fuel rods causes the cladding to creep inwards toward the pellet stack. This creep continues until the cladding contacts the pellets, at which time the cladding swells outward and lengthwise with the swelling of the pellets in the radial and axial directions. Neutron irradiation of the cladding also induces damage to the grain structure of the metal, which additionally adds to the axial growth.

- **Corrosion and composition changes**: [15] Chemical interaction with the coolant water causes hydriding as well as oxidation and of the cladding’s outer surface, which continues over the lifetime of the fuel and consumes the base metal in the cladding. In addition, these processes induce stress to the cladding due to the difference in density between the chemical compounds and the base metal.

- **Lift off, ballooning and ruptures**: [16] In case of extensive release of fission gasses from the pellets, the rod internal pressure may increase enough to exceed the external pressure from the coolant, causing the cladding to lift-off from the pellet stack, balloon and even rupture. Such behavior is, however, connected to accidental conditions rather than normal operation.

Because the processes described in the first two points above continue throughout the cladding’s lifetime, the mechanical properties of the cladding are degraded with increasing burnup. Furthermore, the degradation accelerates with increasing temperatures, since increasing temperatures increase chemical reaction rates for oxidation and hydriding. The mechanical performance of the cladding is an important limitation on the reactor operating conditions (e.g. water chemistry, temperature) and ultimately on how long the fuel may be irradiated.
3. Investigating Nuclear Fuel Performance

As described in the previous section, the operating environment in the reactor core leads to many changes in the fuel properties, and in many respects the fuel performance is degraded as burnup increases. Since it is of general interest to use the fuel more efficiently, and thus increase burnup, it is necessary to investigate nuclear fuel performance in order to better understand it and find ways to mitigate the degradation processes. Accordingly, new fuel designs are periodically introduced. The ability of the fuel to perform its intended functions as well as the ability of designers and engineers to predict the fuel performance under various conditions must be verified, and updated according to changing operating conditions and/or fuel designs.

There are many computer codes used to design nuclear fuel and to predict its behavior in reactors for a wide range of operating conditions. The fuel design and fuel behavior codes attempt to account for all of the phenomena that have an effect on fuel performance. Specifically, the codes account for effects from e.g. neutron flux, LHR, burnup, isotopic content, FGR, pellet cracking, cladding oxidation, including the interdependence of all of these phenomena. The codes are continuously updated for new fuel designs and operating conditions, for which they need to be validated using experimental measurement data.

Many types of destructive and non-destructive examinations are performed on fuel irradiated in commercial as well as research reactors in order to investigate fuel behavior under various conditions. The results of these examinations are incorporated into the fuel design and fuel performance codes.

3.1. Assessment of Commercial Reactor Fuel

Investigations of nuclear fuel performance at commercial NPPs are limited in scope. While the fuel is being irradiated in the core it is only possible to evaluate the condition of the fuel through indirect means, such as monitoring the radiation monitor readings in the steam lines or examining the coolant chemistry. The radial and axial power distribution within the core is also periodically assessed using detectors that are moved through the core at fixed locations; however these measurements only reveal the global power distribution in the core and not the assembly or rod-wise power distributions.
The fuel may also be examined when it is removed from the core and placed in the spent fuel storage pool (SFSP). Examinations performed on the fuel in the SFSP are referred to as poolside investigations or poolside inspections.

The information that is obtained in the poolside examinations is somewhat limited for several reasons: first the investigations are desired to be nondestructive since storing and/or disposing of non-intact fuel is costly, second, for radiation shielding and fuel cooling purposes the fuel is inspected while it is located several meters below the surface of the water in the spent fuel pool, so the inspection methods must be able to be performed remotely underwater, and last, removal of fuel rods for individual inspection during a poolside investigation is time-consuming and entails the risk that the fuel will be damaged. Since poolside investigations are nondestructive in nature, they have the advantage that the fuel may be inspected and reinserted into the reactor core for further irradiation. Thus, the fuel may be inspected many times throughout its lifetime in order to investigate its properties as the burnup increases and/or as it is exposed to varying operating conditions. However, it is generally desired to avoid repeated dismantling of fuel for inspection of individual fuel rods due to the risk of damaging the fuel.

Many of the pool-side inspections are focused on assessing the condition of the cladding. The length and diameter of the fuel rods may be measured using conventional methods, such as visual inspection, and the oxide thickness may be determined using eddy-current techniques. Attributes of the fuel assembly may be determined, such as the amount of bowing present in BWR fuel channels, or the physical dimensions of PWR spacer grids. The surface of fuel rods may be also scraped for analysis of its chemical composition [17].

Other inspection techniques that may be used are gamma-ray spectroscopy and gamma tomography, which are the subjects for this thesis. Gamma-ray spectroscopy may be performed using dedicated underwater measurement equipment [18], in order to investigate the spatial variations of isotopic content in the fuel. These measurements may be performed on individual fuel rods or entire fuel assemblies. Gamma tomography is an advanced method of investigating the fuel through its emitted gamma-radiation field that has previously been experimentally performed to a very small extent at a commercial NPP [19]. Gamma-ray spectroscopy and gamma tomography methods and their applications are described in detail in section 4 and section 5, respectively.

There are also numerous methods for destructive analysis of fuel from commercial NPPs, which require the fuel to be sent to hot laboratories where it is cut into pieces and analyzed. Such methods can give very detailed information about the fuel, but are costly, time-consuming, and lead to waste management issues. Destructive analysis methods are beyond the scope of this thesis.
3.2. Experimental Investigations at Research Reactors

At research reactors, a myriad of investigations may be performed. Many fuel properties are investigated while the fuel is undergoing irradiation in the reactor, and the fuel may additionally be investigated under conditions that are not present in commercial reactors. Examples of the properties that may be investigated during irradiation are listed in Table 1.

Table 1, Fuel properties assessed during irradiation in research reactors.

<table>
<thead>
<tr>
<th>Fuel property</th>
<th>Means of assessment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel centerline temperature</td>
<td>Fuel thermocouples or expansion thermometer [20], [21]</td>
</tr>
<tr>
<td>Pellet stack elongation</td>
<td>Linear Variable Differential Transformer (LVDT) [20]</td>
</tr>
<tr>
<td>Rod-internal pressure</td>
<td>Pressure transducer [20], [21]</td>
</tr>
<tr>
<td>Fission gas composition</td>
<td>Online gamma measurement during irradiation [21]</td>
</tr>
<tr>
<td>Fuel rod diameter</td>
<td>Diameter gauge (LVDT-based) [20]</td>
</tr>
</tbody>
</table>

The operating conditions to which the experimental fuel is exposed may be adjusted with regard to e.g. power level (i.e. LHR), water chemistry, and pressure. The fuel may also be exposed to accident conditions such as power excursions (i.e. Reactivity Initiated Accidents (RIA) or loss of coolant accidents (LOCA)).

In addition to altering the conditions outside the rod during the test irradiation, the conditions inside the fuel rods may also be altered. For example, the composition of the fill gas may be changed, and the fuel rod cladding may intentionally be breached in order to study the effects of a fuel rod cladding failure.

Much of the research performed is classified as materials research, where the performance of fuel pellet material and fuel cladding material is investigated. Indeed, many of the experiments performed at research reactors are done in preparation for introducing new fuel types or new operating regimes at commercial NPPs.

Research reactors are also equipped with facilities to perform extensive destructive and nondestructive Post Irradiation Examination (PIE) on the fuel once it is removed from the reactor. Examples of PIE investigations performed within the OECD Halden Reactor Project (HRP) are shown in Table 2:
Table 2. Irradiated nuclear fuel properties and the corresponding PIE methods utilized within the OECD Halden Reactor Project [22].

<table>
<thead>
<tr>
<th>Fuel Property</th>
<th>PIE method(s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>%FGR and isotopic content of gases</td>
<td>Gamma spectroscopy, puncturing and mass spectrometry</td>
</tr>
<tr>
<td>Axial burnup and power profiles</td>
<td>Gamma spectroscopy</td>
</tr>
<tr>
<td>Grain structures of the fuel pellets and cladding</td>
<td>Metallography/ceramography</td>
</tr>
<tr>
<td>Isotopic distribution within the fuel pellets</td>
<td>Gamma scanning</td>
</tr>
<tr>
<td>Fuel pellet integrity and cladding hydriding</td>
<td>Neutron radiography</td>
</tr>
</tbody>
</table>

As seen in Table 2, many of the PIE methods involve gamma spectroscopy. It can be noted that gamma spectroscopy and gamma tomography measurements applied on experimental fuel at research reactor facilities differ from applications at commercial NPPs in several ways:

- At research facilities, the instrumentation may be placed in a laboratory environment, as opposed to commercial NPPs where the equipment is underwater in an industrial environment.
- Measurements may be performed on fuel that has a very short cooling time such that the content and spatial distribution of short-lived isotopes may be characterized.
- The time available for measurements is greater at research facilities than at NPPs, thus longer measurements and/or a greater quantity of measurements may be performed at research facilities.

Gamma spectroscopy and gamma tomography characterization of research reactor fuel is the topic of this thesis and these methods are described in more detail in the following sections.
4. Gamma-ray Spectroscopy on Nuclear Fuel

4.1. General Methodology / Principles

When radioactive isotopes decay, many of them emit gamma-rays, where the energies of these gamma rays are specific to the isotopes from which they are emitted. The gamma-rays are furthermore emitted in a known fraction of radioactive decays for each isotope [23], referred to here as the gamma-ray yield. The presence of specific isotopes in a radioactive object and the amount of each isotope present may be determined by analyzing the intensity of their characteristic gamma rays in the radiation field emitted by object.

The rate of decay of a radioactive isotope at time, $t$, is referred to as activity, and is expressed in terms of the number of decays per second. The SI unit of activity is the Becquerel (Bq), where one Bq is equal to one decay per second. Equation 1 describes the activity of an isotope at time $t$.

$$a(t) = n(t)\lambda$$

Eq. 1

Where $a(t)$ is the activity at time $t$, $n(t)$ is the amount of the isotope at time $t$ and $\lambda$ is the so-called decay constant, particular to that isotope. As a consequence of the decay, the amount of an isolated isotope decreases with time according to Equation 2.

$$n(t) = n(0)e^{-\lambda t}$$

Eq. 2

Furthermore, the intensity of characteristic gamma rays emitted for a selected isotope is calculated according to Equation 3.

$$I(t) = a(t)\varepsilon_E$$

Eq. 3

Where the $I(t)$ is the intensity of emitted characteristic gamma rays at time $t$, $a(t)$ is activity at time $t$, and $\varepsilon_E$ is the gamma-ray yield for gamma rays of energy $E$ particular to that isotope.

The relationships in Equation 1, 2 and 3, may be used to determine the amount of an isotope present in a measured volume, based on the intensity in
of the isotopes characteristic gamma rays. However, one must also take into account the loss of gamma rays as they pass through various materials from the point of emission to the detector, which is referred to as attenuation, and which can be expressed according to Equation 4.

\[ I(x) = I(0)e^{-\mu x} \]  

Eq. 4

Where \( \mu \) is called the attenuation coefficient, which is specific for each material and varies with the gamma-ray energy, and \( x \) is the distance through the material travelled by the gamma ray. If the gamma ray passes through many different materials, which is often the case, the attenuation of each material must be taken into account.

In gamma-ray spectroscopy, the gamma radiation field surrounding a radioactive source is recorded by a detection system in which the detected gamma rays are sorted according to their energy. The detection system thus records the gamma-ray energy spectrum in terms of the gamma-ray intensity as a function of gamma-ray energy. An example gamma-ray spectrum recorded from a measurement of nuclear fuel is shown in Figure 11.

\[ \text{Counts/second} \quad \text{Energy [keV]} \]

\[ 662 \text{ keV} \]

*Figure 11, Example of a gamma-ray spectrum.*

The various peaks observed in the gamma-ray spectrum in Figure 11 are at energies characteristic to specific isotopes present in the nuclear fuel at the time of measurement, and the intensity in the peaks is proportional to the amount of the isotopes present. The massive background under the peaks at low energies is produced partly from the scattering of gamma-rays in the object and in nearby materials into the detector – a process in which the gamma rays lose energy – and partly from incomplete energy collection in the detector itself, which is also mainly due to scattering.
By analyzing a gamma-ray spectrum, the fission product and activation product isotopes present in the measured volume, as well as the amounts of these isotopes may be determined. Specifically, the content of a radioactive isotope within a measured volume in nuclear fuel is calculated according to Equation 5.

\[
N = \frac{A \cdot 1 \cdot 1 \cdot 4\pi R^2}{\lambda \cdot \eta \cdot \epsilon} \cdot \frac{1}{S} \cdot e^{\sum \mu_i d_i} \cdot e^{\lambda t_D}
\]  
Eq. 5

Where,

- \(N\): Number of the atoms of the selected isotope in the measured volume at the time of reactor shutdown [atoms/cm³]
- \(A\): Measured intensity in a characteristic gamma-ray peak at time, \(t_D\), after shutdown [counts/second]
- \(\lambda\): Decay constant of the selected isotope [seconds⁻¹]
- \(\eta\): Relative detector efficiency at the selected gamma-ray energy
- \(\epsilon\): Gamma emission yield for gamma rays of the selected energy per disintegration of the selected isotope
- \(R\): Distance between the fuel and the detector [cm]
- \(S\): Area of the detector exposed in the measurement [cm²]
- \(\mu_i\): Linear attenuation coefficient at the selected gamma-ray energy, for material, \(i\), between the fuel and detector [cm⁻¹]
- \(d_i\): Gamma-ray travel distance through material, \(i\), between the fuel and detector [cm]
- \(t_D\): Time since irradiation in the reactor [seconds]

Furthermore, the spatial distribution of the isotopes in the fuel may be determined by measuring several discrete volumes, e.g. the distribution of selected isotopes may be determined along the axial length of a fuel rod, or the relative content of isotopes in one fuel assembly may be compared to their content in another assembly. The absolute and relative content of the isotopes present reveals information about fuel performance. The instrumentation necessary for gamma-spectroscopy measurements on nuclear fuel and some measurement applications are discussed in the following sections.

The process of measuring the gamma-ray intensity along the axial length of a fuel rod is referred to as \textit{gamma scanning}; however, the term gamma scanning may also refer to gamma-spectroscopy measurements of nuclear fuel in general, even when the measurements do not comprise an axial scan.
4.2. Requirements on the Instrumentation

The instrumentation necessary to perform gamma spectroscopy measurements on nuclear fuel consists of two basic components; 1) a gamma-ray spectroscopy system, and 2) a physical system that enables measurement of selected, discrete volumes within the fuel. The gamma ray spectroscopy system consists of a gamma-ray detector and the associated electronics and software for processing the signal generated in the detector and analyzing pulse heights to create a gamma-ray energy spectrum of the type shown in Figure 11. The physical system consists of shielding and collimation which allows only gamma rays from selected regions of the fuel to reach the detector and fixtures for positioning the detector relative to the fuel.

4.2.1. Gamma-ray Spectroscopy System

An important property of a gamma-ray detector is its energy resolution, i.e. its capability to retain information on the energy deposited in the detector material by incoming gamma rays. Energy resolution is exhibited in terms of the width of each peak in the gamma-ray energy spectrum. The width of gamma ray peaks are expressed in terms of their Full Width at Half Maximum (FWHM) – the width of the peak at half of its maximum value. In general, high energy resolution (small FWHM) is beneficial since it allows for more details to be present in the gamma-ray spectrum where near-lying peaks are separated, and the background present in terms of the continuum under the peaks is limited.

The energy resolution is particular to the detector type and the specific physical dimensions and construction of each individual detector. The energy resolution of a detector is also dependent on the energy of the incoming gamma rays. Table 3 lists the energy resolution of some selected detector types (for gamma rays with energy of 662 keV).

Table 3. Examples of some typical gamma-ray detector types and representative energy resolutions at 662 keV.

<table>
<thead>
<tr>
<th>Detector type</th>
<th>Energy Resolution at 662 keV</th>
</tr>
</thead>
<tbody>
<tr>
<td>BGO (Bismuth Germanate)</td>
<td>6.5% (43 keV) [24]</td>
</tr>
<tr>
<td>LaBr₃ (Lanthanum Bromide)</td>
<td>2.9 – 4.1% (19 - 27 keV) [25]</td>
</tr>
<tr>
<td>HPGe (High Purity Germanium)</td>
<td>0.2% (1.3 keV) [26]</td>
</tr>
<tr>
<td>NaI (Sodium Iodide)</td>
<td>6.9 % (46 keV) [25]</td>
</tr>
</tbody>
</table>

As seen in Table 3, the measurement of the 662 keV peak from $^{137}$Cs using a detector with relatively large FWHM, such as a BGO detector, would
result in other eventual peaks within some tens of keV from the 662 keV peak being mixed with the 662 keV peak.

High Purity Germanium (HPGe) detectors offer the best commercially available energy resolution; however, they require cryogenic cooling (commonly through the use of liquid nitrogen). HPGe detectors are particularly useful for measurements when many gamma-ray peaks are present and this level of resolution is necessary to be able to observe two near-lying peaks in a gamma-ray spectrum, which is often the case for irradiated nuclear fuel. An example of a spectrum from a PWR fuel assembly 5 years after discharge from the reactor, collected using four different detector types is presented in Figure 13, where the data is from [27].

![Figure 12](image)

**Figure 12**, Gamma-ray energy spectra collected from a PWR fuel assembly 5 years after it was discharged from the reactor, using four different detector types to demonstrate differences in energy resolution. (data from [27])

The gamma-ray detector is coupled to a spectroscopy system that provides the high voltage for the detector and processes the signal generated in the detector. The main components of the signal processing system are the high-voltage supply, amplifiers and an analog to digital converter. The spectroscopy system may be composed of modular components, or it may consist of a single device that houses all components – a so-called Multi-Channel Analyzer (MCA).

The output of the spectroscopy system is coupled to a computer which utilizes gamma-spectroscopy software to record the measured gamma-ray spectra.
4.2.2. Shielding, Collimation, and Positioning Fixtures

In order to measure a selected discrete volume in the fuel it is necessary to shield the detector from undesirable sources of radiation, including radiation which may be present in the background, and radiation emitted from the parts of the fuel which are not being measured. Typical shielding materials are dense materials such as lead or tungsten, but adequate shielding may also be provided by water when gamma spectroscopy measurements are performed underwater.

A collimator is used to restrict the gamma rays emitted from the fuel into a beam which is directed at the detector. The collimator is effectively a shielding component that contains an opening, through which gamma-rays may pass without being (significantly) attenuated prior to reaching the detector. The collimator is positioned such that only gamma rays from the region of interest are allowed to reach the detector.

In order to measure gamma rays from a region of interest in the fuel it is necessary to position the detector, collimator, and fuel relative to each other as desired.

4.3. Measurement Applications

Non-destructive gamma-spectroscopy measurements can be applied to nuclear fuel at commercial NPPs as well as at research reactors. The non-destructive measurement applications are similar for measurements at both types of plants; however, due to fuel handling constraints, it is generally not possible to measure isotopes with very short half-lives at commercial NPPs. Some common applications that are important with regard to fuel performance are shown in Table 4 along with the measured isotopes.

<table>
<thead>
<tr>
<th>Fuel Property</th>
<th>Isotope</th>
</tr>
</thead>
<tbody>
<tr>
<td>Burnup</td>
<td>$^{137}\text{Cs}$</td>
</tr>
<tr>
<td>Power distribution</td>
<td>$^{140}\text{Ba}/^{140}\text{La}$</td>
</tr>
<tr>
<td>Fission Gas Release</td>
<td>$^{85}\text{Kr}$</td>
</tr>
</tbody>
</table>

To be useful for assessment by gamma scanning, the gamma-ray energy must be high enough for the gamma rays to escape the fuel and reach the detector. In addition, the half-life must be suitable for the measurement application in question. This is further discussed below.
4.3.1. Measuring Power and Burnup Distributions

The isotopes $^{137}$Cs and $^{140}$Ba have high fission yields and are produced proportionally to the neutron flux in the reactor. $^{137}$Cs has a long half-life and builds up in the fuel over the fuel’s lifetime. If corrected for decay after irradiation in the core, the content of $^{137}$Cs in the fuel is thus proportional to the total burnup of the fuel. The total burnup of the fuel also reveals the average power (LHR) over the lifetime of the fuel, and by scanning rods or assemblies axially, one can map the axial burnup distribution.

$^{140}$Ba has a much shorter half-life than $^{137}$Cs, so its content in the fuel is proportional to the burnup over a shorter period of reactor irradiation, proportional to its half-life (i.e. not the entire lifetime). As with $^{137}$Cs the burnup reached over this period of time also reveals the average power (LHR) over the same time period. Measurements of $^{140}$Ba are in-fact used to investigate the average power (i.e. average LHR) over the last weeks of reactor operation [28]. The cumulative fission yields for $^{137}$Cs and $^{140}$Ba for fission of $^{235}$U, as well as their half-lives are shown in Table 5.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life</th>
<th>Gamma-ray Energy [keV]</th>
<th>Gamma-ray Emission Yield [%]</th>
<th>Cumulative fission yield (from $^{235}$U fission) [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{137}$Cs</td>
<td>30.08 years</td>
<td>662</td>
<td>85</td>
<td>6.1883</td>
</tr>
<tr>
<td>$^{140}$Ba</td>
<td>12.75 days</td>
<td>1596*</td>
<td>95</td>
<td>6.2145</td>
</tr>
</tbody>
</table>

*emitted in the decay of the $^{140}$Ba decay product $^{140}$La

In order to for the measurements of the 1596 keV gamma rays emitted in the decay of $^{140}$La, which has a half-life of 1.678 days, to be representative of the content of $^{140}$Ba and thus the power distribution, the fuel should be allowed to decay for one to two weeks such that the $^{140}$La inventory present in the fuel at the time of shutdown vanishes. After allowing the fuel to decay sufficiently, the inventory of $^{140}$La in the fuel arises solely from the decay of $^{140}$Ba (since the neutron flux is no longer present).

While one should wait for the initial End of Life (EOL) inventory of $^{140}$La to decay, the measurements must also be performed before the $^{140}$Ba inventory itself vanishes through decay. The intensity in the 1596 keV gamma-ray peak also diminishes as the $^{140}$Ba inventory decreases in the fuel. Due to these reasons it is desirable to perform the measurements as soon as possible after the initial decay period of approximately one to two weeks. An example of a gamma-ray spectrum for a fuel assembly at 23 days cooling time is presented in Figure 13.
The distribution of $^{140}$Ba may be measured along the axial length of entire fuel assemblies or individual fuel rods may be extracted for measurements. Measuring the axial distribution yields the axial power distribution, and comparing the relative contents in several fuel assemblies or fuel rods at the same axial level yields the radial power distribution in the reactor core and/or in the fuel assembly, depending on which fuel assemblies and rods are measured.

These measurements are performed both at commercial NPPs, and at research reactors [22], [28]. It should be noted that removal of individual fuel rods for measurement is time consuming and entails the risk of damaging the fuel as mentioned in section 3.1.

With respect to $^{137}$Cs, its distribution may also be measured along the axial length of entire fuel assemblies or individual fuel rods may be extracted for measurements. Measuring the axial distribution of $^{137}$Cs yields the axial burnup distribution. Comparing the relative contents in fuel rods within the same assembly yields the radial burnup distribution within that assembly and the relative burnup between different fuel assemblies may be determined by comparing their relative $^{137}$Cs contents.

It should be noted that $^{137}$Cs is volatile and may, under certain conditions, migrate within the fuel from its point of production. $^{137}$Cs migrates from hot areas of the fuel to cooler areas of the fuel. This means that $^{137}$Cs migrates away from the hot pellet central region to the outer pellet surface and to the pellet-pellet gaps. $^{137}$Cs may also migrate axially from the axial center of the fuel rod (where the peak LHR usually occurs) to the upper or lower parts of the rod where the LHR is lower than the axial center region. It has been

Figure 13, Gamma-ray spectrum recorded from irradiated fuel with a cooling time of twenty-three days where a strong 1596 keV peak is observable.
shown that the migration of Cesium occurs at LHRs greater than 30 kW/m\cite{30, 31, 32}.

Due to its long half-life, $^{137}$Cs may be measured for many decades after the fuel has been discharged from the reactor. An example of a gamma-ray spectrum from fuel with a cooling time of 21 years is presented in Figure 14.

![Gamma-ray spectrum from irradiated fuel with cooling time of 21 years](image)

*Figure 14, Gamma-ray spectrum from irradiated fuel with cooling time of 21 years where a strong 662 keV peak is observable.*

### 4.3.2. Measuring Fission Gas Release

As discussed in section 2, some of the fission products are gaseous, and at high temperatures, these fission gasses may be released from the fuel pellets and cause the internal pressure in the fuels rods to rise, which eventually may cause cladding lift-off or even ballooning. It is therefore of high relevance to gain better understanding of the mechanisms behind FGR and thus measurements are often carried out to assess to which degree the fission gasses have escaped the fuel pellets. When performing non-destructive assessment of FGR, the fuel is typically dismantled and gamma-spectroscopy measurements are carried out one rod at a time in the gas plenum region of the fuel rod, where the gasses collect and where there is no fuel material.

Most fission gas isotopes are short lived (half-lives < 1 week), and due to the fuel handling constraints at commercial NPPs, it is not practical to measure these isotopes since they have vanished through decay by the time the fuel may be dismantled and measurements can be performed. One fission gas isotope, $^{85}$Kr, however, has a long half-life of 10.78 years, meaning that its content in the fuel corresponds to the entire irradiation lifetime. It emits gamma rays at an energy of 514 keV, which is often measured during poolside gamma spectroscopy measurements that may be performed many years after the fuel’s EOL\cite{33, 34, 35}. Two particular challenges when
measuring this peak are its weakness ($^{85}$Kr has a low fission yield and the branching ratio of gamma-ray emission at 514 keV in its decay is as low as 0.434%) and the presence of two nearby peaks, which are often relatively strong in comparison to the 514 keV peak, namely the 511 keV annihilation peak and the 512 keV peak from the decay of $^{106}$Rh. Figure 15 (top) is an example of a gamma-ray spectrum collected in the gas plenum of a fuel rod with 9 months cooling time, where Figure 15 (bottom) shows the energy range around the 514 keV peak, where the 511 keV peak is also observed.

Figure 15, top: Gamma-ray spectrum from the gas plenum of irradiated fuel with a cooling time of 9 months, where the 514 keV peak is labeled; bottom: the energy range around the 514 keV peak where the 514 keV peak and the overlapping peak at 511 keV are labeled.

At research reactors, where the fuel handling constraints do not prevent gamma spectroscopy measurements from taking place a short time after
shutdown, more short-lived isotopes may be measured in the gas plenum. Possible isotopes that may be assessed have been investigated in Paper I, based on theoretical considerations and on simulations. Three short-lived isotopes of interest were identified; $^{133}$Xe, $^{133m}$Xe and $^{135}$Xe. Some selected properties of these isotopes are presented in Table 6, together with the properties of the more established isotope $^{85}$Kr.

Table 6, Properties of the gaseous gamma-emitting fission products $^{85}$Kr, $^{133}$Xe, $^{133m}$Xe and $^{135}$Xe and the energies of the gamma rays emitted in their decay that have been studied in this thesis [29].

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life</th>
<th>Cumulative fission yield (from $^{235}$U fission) [%]</th>
<th>Prominent gamma-ray energies [keV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{85}$Kr</td>
<td>10.78 years</td>
<td>0.283</td>
<td>514</td>
</tr>
<tr>
<td>$^{133}$Xe</td>
<td>5.24 days</td>
<td>6.699</td>
<td>81, 161</td>
</tr>
<tr>
<td>$^{133m}$Xe</td>
<td>2.2 days</td>
<td>0.195</td>
<td>233</td>
</tr>
<tr>
<td>$^{135}$Xe</td>
<td>9.14 hours</td>
<td>6.539</td>
<td>250, 608</td>
</tr>
</tbody>
</table>

Of the short-lived isotopes in Table 6, an experimental assessment presented in Paper I showed the best results for $^{133}$Xe, with a measurable peak at 81 keV, emitted at a branching ratio of 38%. However, the relatively low energy requires special considerations to limit the gamma-ray attenuation between the fuel and the detector. The investigations made are further described in section 7.1, including experimental gamma-ray spectra from fuel rods with short decay time.

4.3.3. Determining Fission Gas Release Fraction

The fission gas release fraction, %FGR, is conventionally calculated according to Equation 6 [35].

$$%FGR = 100 \cdot \frac{N_{released}}{N_{produced}}$$

Eq. 6

Where, $N_{released}$ is the amount of the fission gas isotope that has been released from the fuel pellets, and $N_{produced}$ is the total amount of the fission gas isotope produced in the fuel. $N_{released}$ and $N_{produced}$ may be determined through gamma spectroscopy measurements on the fuel rod gas plenum and fuel stack regions, respectively.

$N_{released}$ is determined by measuring the concentration of a radioactive fission gas isotope in the gas plenum region of a fuel rod; however, a direct measurement of the fission gas content in the fuel, $N_{produced}$, is generally not possible due to the high levels of gamma emission from the fuel material.
Instead, $N_{\text{produced}}$ is measured indirectly using an abundant gamma-emitting fission product with similar production pathway and half-life as those of the measured fission gas isotope.

The established isotope for measurement of $\%\text{FGR}$ is $^{85}\text{Kr}$, and the corresponding isotope measured in the fuel stack is $^{137}\text{Cs}$. The half-lives of $^{85}\text{Kr}$ and $^{137}\text{Cs}$ are similar, in that they are both longer than the typical irradiation time of a nuclear fuel assembly, meaning that their inventory at the fuel’s EOL corresponds to the entire lifetime of irradiation. Furthermore, they are both produced predominantly by direct fission and only to a very small extent by neutron absorption (and consequent decay).

Figure 16 illustrates how the measurements are performed. Specifically, the gamma rays emitted in the decay of $^{85}\text{Kr}$ are measured in the fuel rod gas plenum (Figure 16 left), and the gamma rays emitted in the decay of $^{137}\text{Cs}$ are measured in the fuel stack (Figure 16 right). While Figure 16 illustrates the scenario at commercial NPPs where the measurements are performed underwater, the scenario is the same at research reactors (e.g. the HBWR) except that the measurements may be performed in air instead of underwater. At commercial power plants, there is often no collimator opening available in the pool wall, and then a submersible device may be deployed [18].

![Figure 16](image)

*Figure 16.* Illustration of measurement scenario for determining the fission gas release fraction. The image at left depicts the gas plenum measurements, while the image at right depicts the fuel stack measurement.

If the measurement geometry is identical for the fuel and plenum measurements, Equation 7 may be used to calculate the fission gas release fraction.
\[
\%FGR = 100 \cdot \frac{A_5 \tau_7}{A_7 \tau_5} \cdot F(Z) \cdot \frac{\ell \cdot k_k}{L} \cdot \frac{Y_7}{Y_5} \cdot \frac{\varepsilon_7}{\varepsilon_5} \cdot \frac{1 - e^{-\lambda_7 t_B}}{1 - e^{-\lambda_5 t_B}} \cdot \frac{e^{-\lambda_7 t_D}}{e^{-\lambda_5 t_D}} \cdot \frac{\alpha_{\mu}(662)}{\alpha_{\mu}(514)} \cdot \frac{\eta(662)}{\eta(514)} \cdot P_{\mu} \cdot Q_{\mu} \cdot K_e
\]

Eq. 7

Where the index 5 and 7 correspond to \(^{85}\text{Kr}\) and \(^{137}\text{Cs}\), respectively, and:

- \%FGR: Fraction of \(^{85}\text{Kr}\) released from the fuel
- A: Net counts in the gamma-ray peak at time, \(t_D\), after reactor shutdown [counts/second].
- \(\tau\): Accumulation (live) time of the measurement
- \(F(Z)\): Axial burnup form factor at the position of the \(^{137}\text{Cs}\) measurement
- \(\ell\): Plenum length [mm]
- \(L\): Fuel column length [mm]
- \(k_k\): Compression factor taking into account the rod internal volume distribution.
- \(Y\): Fission yield [atoms per fission]
- \(\varepsilon\): Gamma-ray emission yield [gamma-rays per disintegration]
- \(\lambda\): Decay constant of the selected isotope [seconds\(^{-1}\)]
- \(t_B\): Burnup time [seconds]
- \(t_D\): Decay time [seconds]
- \(\alpha_{\mu}(E)\): Gamma-ray attenuation from fuel rod to detector for energy \(E\)
- \(\eta(E)\): Relative detector efficiency at energy \(E\)
- \(P_{\mu}\): Attenuation of 662 keV gamma rays in the fuel stack
- \(Q_{\mu}\): Attenuation of 662 keV gamma rays in the additional absorbers (if present)
- \(K_e\): Calibration factor

It should be noted here that the measurement geometry in the plenum and fuel stack regions are identical except for the possible addition of an absorber in the collimator opening to limit the count rate in the detector during the fuel measurement. Due to this, the geometrical arrangement of the measurements does not need to be accounted for in the calculation of the fission gas release fraction.

In order to determine fission gas release fraction using this method, fuel rods must be removed from their assemblies for individual measurement, effectively limiting the number of fuel rods which are characterized in one measurement campaign.

**Determining %FGR based on measurement of short-lived isotopes**

The measurement of \%FGR based on \(^{85}\text{Kr}\) and \(^{137}\text{Cs}\) is useful for determining the integrated fission gas release which has occurred over the fuel’s lifetime, thus providing information about the fuel performance over its life-
time. Determining %FGR based on short-lived isotopes, however, gives information about the FGR behavior over short periods of irradiation (corresponding to the half-lives of the measured isotopes). Information on the FGR which occurs over shorter time periods may be useful in investigations of the mechanisms that contribute to FGR. A novel method for assessing time-dependent FGR based on measurements of $^{133}$Xe and $^{140}$Ba has been proposed in Paper IV. It has also been demonstrated experimentally, as further discussed in section 7.

The half-lives of $^{133}$Xe and $^{140}$Ba are on the order of days, such that their content in the fuel at the end of irradiation corresponds to the last weeks of irradiation. It should be noted that these short-lived isotopes may only practically be measureable at research reactors, and that the irradiation cycles at these reactors may be shorter than one or two half-lives of $^{133}$Xe and $^{140}$Ba, meaning that their content in the research reactor fuel corresponds to the entire experimental irradiation.

As mentioned in section 4.3.1, $^{140}$Ba content is determined by measuring the 1596 keV gamma rays emitted by its decay product $^{140}$La, which is also produced separately from $^{140}$Ba during reactor operation. The method described above (based on $^{85}$Kr and $^{137}$Cs) may be applied directly to measurements of $^{133}$Xe and $^{140}$Ba in the case that $^{140}$La is allowed to decay prior to the measurements (as described in section 4.3.1). However, if the measurements are performed a short time after shutdown such that the $^{140}$La inventory present at the time of reactor shutdown contributes to the measurements, then the calculation of %FGR based on $^{133}$Xe must take this inventory into account. The proposed method for accounting for the EOL inventory of $^{140}$La is described in Paper IV and based on using core physics calculations, using the assumption that the core physics code accurately calculates the contents of $^{133}$Xe, $^{140}$Ba, and $^{140}$La.

Specifically, %FGR for $^{133}$Xe can be calculated according to Equation 8. One may note that the ratio of calculated contents of $^{140}$La and $^{133}$Xe is used here instead of the ratio of fission yields to relate the amount of $^{133}$Xe and $^{140}$La produced.

$$FGR_{Xe} = 100 \cdot \frac{\sum_{i} A_{Xe} \cdot \frac{e}{\eta_{Xe}} \cdot \frac{4\pi R^2}{S_{detector}} \cdot \frac{V_{gas}}{V_{meas}} \cdot \frac{V_{fuel}}{V_{fuel-code}}}{N_{Xe}^{fuel-code} \cdot N_{Xe}^{fuel-meas}} \cdot e^{\lambda_{Xe} t_{D}} \cdot \frac{N_{La}^{fuel-code}}{N_{La}^{fuel-meas}} \cdot \frac{N_{La}^{Xe}}{N_{La}^{Xe}} \cdot \frac{N_{La}^{fuel-code}}{N_{La}^{fuel-meas}} \quad \text{Eq. 8}$$
Where,

\[ N_{\text{fuel-code}}^{La}(t_D) = N_{SD}^{La} e^{-\lambda_{La} t_D} + \frac{N_{SD}^{Ba} \lambda_{Ba}}{\lambda_{La} - \lambda_{Ba}} (e^{-\lambda_{Ba} t_D} - e^{-\lambda_{La} t_D}) \]  \hspace{1cm} \text{Eq. 9} \\

and,

\[ N_{\text{fuel-meas}}^{La} = \frac{A}{\lambda_{La}} \cdot \frac{e}{\eta_{La}} \cdot \frac{4\pi R^2}{S_{\text{detector}}} \cdot \frac{F}{V_{\text{meas}}} \]  \hspace{1cm} \text{Eq. 10} \\

and where,

\%FGR: Fraction of \(^{133}\)Xe released from the fuel  \\
\(A\): Net counts in the gamma-ray peak at time, \(t_D\), after reactor shutdown [counts/second].  \\
\(\lambda\): Decay constant of the selected isotope [seconds\(^{-1}\)].  \\
\(\mu_i\): Linear attenuation coefficient at the selected gamma-ray energy, for material, \(i\), between the fuel and detector [cm\(^{-1}\)].  \\
\(d_i\): Gamma-ray travel distance through material, \(i\), between the fuel and detector [cm].  \\
\(\eta(E)\): Relative detector efficiency at energy \(E\).  \\
\(\varepsilon\): Gamma-ray emission yield [gamma-rays per disintegration].  \\
\(R\): Distance between the fuel and the detector [cm].  \\
\(t_D\): Decay time [seconds].  \\
\(F\): Axial form factor at the position of the \(^{140}\)La measurement.  \\
\(S_{\text{plenum}}\): Area of the detector exposed to the fuel in the plenum measurement [cm\(^2\)].  \\
\(V_{\text{gas}}\): Total free volume inside the fuel rod [cm\(^3\)].  \\
\(V_{\text{meas}}\): The measured volume (defined by the collimator) [cm\(^3\)].  \\
\(V_{\text{fuel}}\): Total volume of the fuel pellets inside the fuel rod [cm\(^3\)].  \\
\(N_{\text{fuel-code}}^{Xe}\): Concentration of \(^{133}\)Xe present in the fuel at shutdown from the core physics code (assuming 0\% FGR) [atoms/cm\(^3\)].  \\
\(N_{\text{fuel-meas}}^{La}\): Concentration of \(^{140}\)La in the fuel at time of measurement [atoms/cm\(^3\)].  \\
\(N_{\text{fuel-code}}^{La}\): Concentration of \(^{140}\)La in the fuel at time of measurement as obtained using input from the core physics code [atoms/cm\(^3\)].  \\
\(N_{SD}^{La}\): Concentration of \(^{140}\)La in the fuel at time of shutdown from the core physics code [atoms/cm\(^3\)].  \\
\(N_{SD}^{Ba}\): Concentration of \(^{140}\)Ba in the fuel at time of shutdown from the core physics code [atoms/cm\(^3\)].

Experimental measurements to investigate the feasibility of these measurements and calculations are described in Paper IV and in section 7.
4.3.4. Determining Radial Origin of Released Fission Gasses

As described in section 2.2.1, the spatial distribution of fission products and transuranic elements is not uniform within the fuel, and in the HBS in the radial periphery of the fuel pellets, fission gasses are depleted in the UO$_2$ material, and concentrated in bubbles. Furthermore, the temperature in the fuel is higher in the center of the pellet than at the outer pellet surface. When investigating the FGR phenomenon, it is of interest to understand the location in the fuel where the released fission gasses originated, especially under transient conditions such as may be investigated at research reactors. Paper III describes a method based on measurement of the two fission gasses, $^{133}$Xe and $^{85}$Kr which can be utilized to investigate the radial origin of the released gasses.

$^{133}$Xe has a similar fission yield for fissions occurring in U and Pu, while $^{85}$Kr has different fission yields for fissions in U compared to Pu. Table 7 shows the cumulative fission yields for $^{133}$Xe and $^{85}$Kr for fission in $^{235}$U and $^{239}$Pu. Note that the fission yield of $^{85}$Kr is more than double in $^{235}$U fission as compared to $^{239}$Pu fissions.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Cumulative yield for fission in $^{235}$U [%]</th>
<th>Cumulative yield for fission in $^{239}$Pu [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{133}$Xe</td>
<td>6.6991</td>
<td>7.0163</td>
</tr>
<tr>
<td>$^{85}$Kr</td>
<td>0.2834</td>
<td>0.1227</td>
</tr>
</tbody>
</table>

Since the fission yield of $^{133}$Xe is approximately the same for fission in $^{235}$U and $^{239}$Pu, its production rate across the pellet follows the radial burnup profile of the pellet; however, since less $^{85}$Kr is produced in fission of $^{239}$Pu as compared to fission of $^{235}$U, its rate of production will be relatively lower near the outer surface of the pellet where there is relatively more Pu and less U as compared to the central region of the pellet. As a result, the ratio of $^{133}$Xe/$^{85}$Kr will be higher near the pellet outer surface than in the center part of the pellet.

The method described in Paper III consists of performing a gamma spectroscopy measurement on the gas plenum of an experimental fuel rod, determining the ratio of $^{133}$Xe/$^{85}$Kr in the released fission gasses, and comparing that ratio to the pellet-average ratio of $^{133}$Xe/$^{85}$Kr produced in the fuel as determined using a core physics code. If the ratio in the released gasses is higher than the ratio in the fuel, it indicates that fission gasses have been released from a region of the pellet with a relatively lower production rate of $^{85}$Kr, e.g. the pellet outer surface, or HBS.
When the gamma-ray peaks from $^{133}$Xe and $^{85}$Kr can be observed in the same gamma ray spectrum collected from the gas plenum of a fuel rod, the ratio of $^{133}$Xe/$^{85}$Kr is calculated according to Equation 11.

$$\frac{N_{\text{Xe}}}{N_{\text{Kr}}} = \frac{I_{\text{Xe}} \lambda_{\text{Kr}}}{I_{\text{Kr}} \lambda_{\text{Xe}}} \cdot \frac{\sum e^{-\mu_i X e d_i}}{\sum e^{-\mu_i Kr d_i}} \cdot \frac{\eta_{\text{Kr}} \varepsilon_{\text{Kr}}}{\eta_{\text{Xe}} \varepsilon_{\text{Xe}}} \cdot \frac{e^{\lambda_{\text{Xe}} t_D}}{e^{\lambda_{\text{Kr}} t_D}}$$

Eq. 11

and,

$N$: Number of atoms of the isotope in the measured volume
$I$: Measured intensity in the peak [counts/second]
$\lambda$: Decay constant of the selected isotope [seconds$^{-1}$]
$\mu_i$: Linear attenuation coefficient at the selected gamma-ray energy, for material, $i$, between the fuel and detector [cm$^{-1}$]
$d_i$: Gamma-ray travel distance through material, $i$, between the fuel and detector [cm]
$\eta$: Relative detector efficiency for gamma rays from the respective isotope
$\varepsilon$: Gamma-ray emission branching ratio [gamma-rays per disintegration]
$t_D$: Decay time [seconds]

Experimental results are presented in Paper III, section 7.1.3 and [36].
5. Gamma Emission Tomography

Tomographic techniques are applied in order to nondestructively investigate the interior of objects. A typical application of this technique is in the field of medicine; where internal images of the human body are obtained through external measurements. In transmission tomography, an external source transmits a signal through an object, and is measured on the opposite side of the object. The measured signal is used to reconstruct information about the object’s interior in terms of its gamma-ray attenuating properties. In emission tomography, the source distributed inside the object itself is being measured and investigated. The type of emission tomography applied here is also referred to as Single Photon Emission Computed Tomography (SPECT) and the experiments described in this work are based on measuring the radiation source from nuclear fuel rods in a complete nuclear fuel assembly. This technique is referred to here as gamma-ray emission tomography or simply gamma tomography.

Gamma tomography is based on gamma spectroscopy and tomographic reconstruction techniques, whereby gamma spectroscopy measurements record the radiation field surrounding a nuclear fuel assembly and the recorded intensities are used to reconstruct the spatial distribution of the gamma-ray source within the assembly. With this method, the rod-wise distribution of fission products may be determined without the need to remove the fuel rods for individual measurements. Since it is not necessary to dismantle the fuel, the time-consuming process of removing individual fuel rods, and the risk of damaging the fuel are avoided.

At commercial NPPs, the number of fuel rods that are characterized using single-rod gamma spectroscopy measurements is limited due to the substantial time required for dismantling the fuel. There are also constraints for single-rod measurements at research reactors. At the HBWR, experimental fuel assemblies, which are typically fitted with instrumentation for monitoring their in-core performance, do not permit easy removal of individual fuel rods prior to the assemblies’ EOL in order for them to be individually characterized using gamma spectroscopy. Since gamma tomography characterizes all fuel rods in an assembly without the need to dismantle the fuel, this technique enables faster rod-wise characterization of commercial nuclear fuel assemblies with less risk of damaging the fuel. At the HBWR, experimental fuel assemblies may be characterized on a rod-wise basis as desired during the fuel’s lifetime, without the need to remove individual fuel rods.
5.1. General Methodology

Gamma emission tomography is based on gamma-ray spectroscopy and tomographic reconstruction techniques. Gamma tomography characterization of nuclear fuel is a two-step process:

**Step 1:** Gamma spectroscopy measurements are performed at selected points surrounding the fuel assembly in order to record the gamma-radiation field.

**Step 2:** The information from the recorded gamma spectra is reconstructed to obtain the rod-wise gamma-ray source distribution within the fuel assembly.

Step 1 is illustrated in Figure 17 for an HBWR driver fuel assembly where the measurement sequence consists of rotating the fuel assembly relative to the detector and collimator, and translating the detector and collimator laterally relative to the fuel. In this way, a lateral gamma scan of the assembly is obtained for each rotational position of the fuel assembly where the measured intensity corresponds to the volume defined by the collimator at that measurement position. The result of the lateral scan is shown at right in Figure 17, where the measured intensity in a selected gamma-ray peak is plotted versus the lateral measurement position.

**Figure 17**, Illustration of gamma tomography measurement sequence on an HBWR nine-rod driver fuel assembly. The fuel rotates relative to the detector and collimator, and the detector and collimator translate laterally relative to the fuel. A gamma peak is selected in the energy spectrum using gamma spectroscopy, and the intensity in the peak is measured at a large number of detector and fuel positions. The result for each rotational position of the fuel is the intensity in the selected gamma-ray peak versus lateral position (shown at right in the illustration).

Step 2, the tomographic reconstruction process, may be accomplished using a variety of techniques. Gamma tomography measurements on nuclear
fuel assemblies, where highly heterogeneous gamma-ray attenuation is encountered within the fuel assembly, pose specific challenges on the algorithms used in this step, as further discussed in section 5.3.

5.2. Requirements on the Instrumentation

Gamma tomography instrumentation is designed to accomplish the first step in the measurement sequence, i.e. to record gamma spectra from selected points surrounding the fuel assembly that correspond to well-defined volumes of the fuel assembly. This task implies several constraints on the equipment:

- The overall dimensions of a gamma tomography device must accommodate the fuel to be measured, and be able to precisely position the fuel and detector relative to each other.
- The gamma tomography instrumentation must utilize a collimator which precisely defines the measured volume of the fuel.
- The gamma spectroscopy system must be capable of resolving the gamma-ray peaks of interest in the measured gamma spectra, and the gamma spectroscopy system must be able to accommodate the gamma-ray intensities expected from the fuel to be measured.
- Shielding of the detector against background radiation must be considered, and biological shielding may be necessary, depending on where the instrument is to be placed.
- Consideration must be given to assembly and handling constraints for the equipment.
- Serviceability and decontamination should be considered in the design.

The device used for the tomographic experiments of this work was designed to fulfill these requirements, and it is presented in section 6.2.

5.3. Reconstruction Techniques

There are various ways in which the recorded gamma-ray intensities can be used to reconstruct the internal properties of the objects. Two main categories of tomographic reconstruction techniques are 1) analytic and 2) model-based or algebraic. For nuclear fuel assemblies with their highly inhomogeneous mix of highly attenuating uranium dioxide and surrounding water or air (depending on whether the measurements are performed under wet or dry conditions), reconstruction poses a challenge and algebraic reconstruction techniques have previously been shown to perform better than analytic ones with regard to precision of the reconstructed activities (see ref. [19] and Pa-
per VII). In this work, both types of techniques have been explored. The algorithms used and the applications foreseen for their use are described in Paper VII. Summaries of the two techniques are presented in the following sections.

5.3.1. Analytic Techniques

The Radon transform, presented in 1917 by Johann Radon, forms the basis for analytic reconstructions algorithms. The Radon transform of an object, also referred to as a Projection, is formed from a series of line integrals of some property (e.g. the activity distribution) through an object. The formation of a projection from a set of line integrals is illustrated in Figure 18. Here, two systems of coordinates are used; the Cartesian system \((x,y)\) and the polar system \((s,\theta)\).

![Figure 18, Illustration of the formation of a projection (or Radon Transform) of an object from a series of line integrals through the object.]

In SPECT imaging, the projections, \(p(s,\theta)\), are the measured gamma-ray intensities while the object function, \(a(x,y)\), is the source distribution. As accounted for in Paper VII, there is a variety of analytic algorithms for reconstructing \(a(x,y)\) from \(p(s,\theta)\). For the images presented in section 8 of this thesis, a basic Filtered Back-Projection (FBP) algorithm has been used, based on the Fourier transform, as accounted for in Equation 12.

\[
a(x, y) = \int_0^{2\pi} \left[ \int_{-\infty}^\infty \left[ \int_{-\infty}^\infty p(s', \theta) e^{-j2\pi ws'} ds' \right] F(w) e^{j2\pi ws} dw \right] d\theta \quad \text{Eq. 12}
\]

Where \(F(w)\) represents a filtering function and \(w\) denotes the Fourier coordinate associated to the spatial variable \(s\).
With this reconstruction method is assumed that only activity along the line of sight contributes to the detected intensity (i.e. perfect collimation), when actually the measured intensity comes from a finite volume defined by the collimator. Additionally, in the Back-projection process, isotropic gamma ray emission is not considered, so that the contributions to the measured intensities are back-projected with a uniform distribution along the line of sight.

Equation 12 is applicable to the case of continuous sampling, while the measurement sequence actually defines a finite set of measurement coordinates; therefore, the FBP method is implemented using a discrete Fourier transform. The pattern of pixel elements, pixels, of the reconstructed image also requires interpolation between data points during the back-projection process. Linear interpolation has been applied in this work.

Filtering may either be performed in the spatial or the frequency domain; however, due to the simplicity of its application in the frequency domain, it is usually applied there. There are many filter functions which may be applied, including the windowed ramp, Hamming, Hann, or the Notched ramp filter [37], [38]. In this work the ramp and windowed-ramp filters have been applied.

In the analytic reconstructions presented in section 8 and Paper VIII, the attenuation in the object has not been taken into account. This assumption is justified by the fact that the objects under study are relatively small with a relatively small number of fuel rods in a sparse configuration. Furthermore, the Equation 12 has only been used to present qualitative images and has not been utilized for quantification of the activity contents in the fuel.

5.3.2. Algebraic Techniques

Using Algebraic Reconstruction Techniques, it is possible to take the measurement geometry and the materials composition of the measured object into account when reconstructing the activity distribution within the measured area. Two techniques are presented in Paper VII, where one technique assumes the homogenized fuel object geometry when reconstructing simulated fuel measurements, and the second technique models the actual fuel geometry in detail. For the quantitative results presented in section 8 of this thesis, the method where the actual fuel geometry is modeled has been applied, and an overview of this technique is presented below. More detailed descriptions of both techniques and their application are presented in Paper VII.

General Methodology

With the algebraic reconstruction technique the measured area is described as an area composed pixels, and the gamma-ray intensity measured in a specific detector position can then be expressed as in Equation 13.
\[ I_m = \sum_{n=1}^{N} \omega_{mn} A_n \quad \text{Eq. 13} \]

Where, \( I_m \) is the measured intensity at detector position \( m \), \( A_n \) is the activity in pixel \( n \), and \( \omega_{mn} \) is the probability that gamma rays emitted from pixel \( n \) will be transmitted to the detector in position \( m \). The values, \( \omega_{mn} \) are referred to as *contribution coefficients* and may be estimated or calculated using known information about the measurement geometry and the measured object (i.e. the nuclear fuel assembly). For \( M \) detector positions and \( N \) pixels, a system of equations can be constructed according to Equations 14a and 14b:

\[
\begin{align*}
I_1 &= \omega_{11} A_1 + \omega_{12} A_2 + \cdots + \omega_{1N} A_N \\
I_2 &= \omega_{21} A_1 + \omega_{22} A_2 + \cdots + \omega_{2N} A_N \\
\vdots \\
I_M &= \omega_{M1} A_1 + \omega_{M2} A_2 + \cdots + \omega_{MN} A_N \\
\end{align*}
\quad \text{Eq. 14a}
\]

or equivalently;

\[ \bar{I} = \bar{W} \cdot \bar{A} \quad \text{Eq. 14b} \]

Once the values of \( I_m \) are known and the contribution coefficients \( \omega_{mn} \) are calculated, the system of equations can be solved to reconstruct the activity distribution in the pixels. In this work, an iterative solution technique called ASIRT (Additive Simultaneous Iterative Reconstruction Technique) [39], [40] has been utilized, where the algorithm for updating the activities from iterative step, \( k \), to step \( k+1 \) is shown in Equation 15.

\[ A_{n}^{k+1} = A_{n}^{k} + \frac{1}{\sum_{m=1}^{M} \omega_{mn}} \sum_{m=1}^{M} \left( I_m - \sum_{n=1}^{N} \omega_{mn} A_{n}^{k} \right) \omega_{mn} \quad \text{Eq. 15} \]

Although ASIRT has been used here, there are other techniques available for solving the system of equations [41]. The reconstructed activities are not considered to be sensitive to the solution method used; however, for the method to return an accurate reconstruction of the activity distribution, the values of the contribution coefficients \( \omega_{mn} \) must be reasonable.

**Contribution Coefficients**

In order to allow for a simplified calculation of the contribution coefficients, only full-energy transport of mono-energetic gamma rays is considered in this work, and elastic scattering is neglected. These assumptions are valid
since the measurement equipment presented in section 6.2 and Paper VIII utilizes an HPGe detector that has a high level of energy resolution.

The methods used to perform detailed modeling of the gamma-ray transport through the fuel assembly and for modeling the detector response are described in detail in Paper VII, resulting in the following expression for calculation of the contribution coefficients:

\[ \omega_{mn} = \frac{1}{S_n} \left( \frac{h_c^2}{4\pi l_c} \right) \cdot \int_{x,y \in \mathbb{R}} \frac{1}{R(x,y)} \left[ \frac{l_c}{h_c^2 \cdot R(x,y)} \cdot \omega_{coll}(x,y) + b(x,y) \right] \cdot e^{-\mu_i d_i(x,y)} dydx \quad \text{Eq. 16a} \]

where

\[ \omega_{coll}(x,y) = \int_{z} \left( \int_{y_{det}}^{x_{det}} e^{-\mu_{coll} d_{coll}(x,y,x_{det},y_{det})} dx_{det} dy_{det} \right) dz \quad \text{Eq. 16b} \]

and where:

- \( S_n \): the area of pixel \( n \)
- \( R(x,y) \): the distance from point \((x,y)\) in the fuel to the detector in position \( m \)
- \( \mu_i \): the attenuation coefficient for material \( i \)
- \( d_i(x,y) \): the gamma-ray travel distance in material \( i \) from point \((x,y)\) in the fuel to the detector in position \( m \)
- \( h_c \): height of collimator slit
- \( l_c \): length of collimator slit
- \( b(x,y) \): exposed width of the detector in position \( m \) from position \((x,y)\) in the fuel
- \( \mu_{coll} \): attenuation coefficient of the collimator material

The double integral in Equation 16.b corresponds to the detector area that is not directly exposed through the collimator slit, and the outer integral corresponds to the axial direction in the fuel. Equation 16.b can be calculated separately since it does not include any properties of the measured object.

In practice, the integrations of Equations 16.a and 16.b are performed numerically over a large number of points in the pixels and on the detector. For practical reasons, the gamma-ray transport distances \( d_i \) through the fuel material are only modeled for one gamma-ray per emission point, even though one may argue that gamma rays that hit different parts of the exposed detector surface have travelled along different paths. This simplification is valid if the detector is situated far from the fuel and the collimator slit is
narrow, as is the case with the gamma tomography measurements and instrument presented here.

A prerequisite for the reconstructions to incorporate detailed modeling of the gamma-ray transport through the fuel to the detectors is that the fuel geometry is known. In addition to requiring the known fuel geometry and materials, the position of the fuel in the measurement system (i.e. relative to the detector and collimator) must also be known in order to perform this detailed modeling. The fuel position may be determined through dedicated measurements for this purpose; however, the position may also be deduced through image analysis of the reconstructed images [42].

Another feature of this technique is that the pixels can be assigned exclusively to regions in the fuel assembly which contain gamma-ray emitting materials, i.e. the fuel pellet regions in the nominal case. Figure 19 (right) illustrates gamma-ray transport through the fuel cross section of a SVEA96 fuel assembly, and Figure 19 (left) illustrates the pixel pattern imposed on this fuel geometry for obtaining the results presented in section 8.1, where five pixels are placed in each rod – with one central circular pixel and four peripheral pixels. No pixels are assigned to non-fuel rod areas in the fuel cross section.

Figure 19, Left: The pixel pattern imposed on the SVEA-96 fuel geometry, where each rod has five pixels – one central circular pixel, and four peripheral pixels. No pixels are assigned to non-fuel areas. Right: illustration of gamma-ray transport through the fuel cross section of the SVEA-96 fuel assembly.

5.4. Measurement Applications

Gamma tomography is based on gamma spectroscopy and, therefore, it may be used for many of the same fuel characterization applications as single-rod gamma spectroscopy. While gamma tomography may be used for analyzing the internal distribution of fission products in measurements on individual fuel rods [41], [43], [44], this thesis relates to measurements on complete fuel assemblies. Since such application of gamma tomography may be used to characterize all rods in an assembly simultaneously, additional applica-
tions are made possible, in addition to what is possible or practical using single-rod gamma spectroscopy. Some applications which have been demonstrated for gamma tomography on nuclear fuel assemblies are shown in Table 8 with their level of maturity, the measured isotopes and gamma-ray energies, as well as the fuel type for which the application was demonstrated.

Table 8, Nuclear fuel performance applications of gamma tomography on nuclear fuel assemblies, their level of maturity, the isotopes and gamma rays which are measured, and the fuel type for which the application has been demonstrated/investigated.

<table>
<thead>
<tr>
<th>Measurement application</th>
<th>Level of maturity</th>
<th>Isotope(s)</th>
<th>Gamma-ray energy [keV]</th>
<th>Fuel type</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rod-wise power distribution</td>
<td>Experimentally demonstrated [19]</td>
<td>$^{140}$Ba/$^{140}$La</td>
<td>1596</td>
<td>BWR</td>
</tr>
<tr>
<td>Rod-wise burnup distribution</td>
<td>Experimental data under evaluation*</td>
<td>$^{137}$Cs</td>
<td>662</td>
<td>BWR / HBWR</td>
</tr>
<tr>
<td>Rod-wise %FGR</td>
<td>Experimental data under evaluation*</td>
<td>$^{85}$Kr ($&amp;$ $^{137}$Cs)</td>
<td>514</td>
<td>HBWR</td>
</tr>
<tr>
<td>Leaker rod identification</td>
<td>Simulation Study*</td>
<td>$^{135}$Xe, $^{133}$Xe, $^{85}$Kr</td>
<td>250, 81, 514</td>
<td>HBWR</td>
</tr>
<tr>
<td>Nuclear Safeguards, detection of missing fuel rods</td>
<td>Experimentally demonstrated [45], [46], [47]</td>
<td>$^{137}$Cs, Gross gamma</td>
<td>662</td>
<td>BWR, PWR, VVER</td>
</tr>
<tr>
<td>Damaged Fuel Investigation</td>
<td>Experimentally demonstrated [48], [49]</td>
<td>$^{137}$Cs</td>
<td>662</td>
<td>Experimental</td>
</tr>
</tbody>
</table>

* Described in this work

Experimental gamma tomography measurements using the HBWR gamma tomography system (see Paper VIII and section 6.2) have been performed to demonstrate the applications of determining rod-wise burnup distribution, and rod-wise %FGR. Preliminary evaluations of these measurements are presented in Paper VIII and summarized in section 8.2. The leaker rod identification application has been investigated through a simulation study where the simulated fuel and measurement setup corresponded to HBWR fuel and the HBWR gamma tomography instrument. The leaker rod identification investigation is presented in Paper II and summarized in section 8.1.
6. Instrumentation for Gamma Scanning and Gamma Tomography at the Halden Research Reactor

Gamma spectroscopy measurements on individual fuel rods are performed at the HBWR as part of the routine PIE on experimental fuel and a gamma tomography measurement system has recently been used to perform experimental measurements on an HBWR driver fuel assembly. The gamma spectroscopy instrumentation for characterizing individual fuel rods, and the gamma tomography instrumentation for characterizing entire assemblies are described in the following sections.

6.1. Gamma Scanning Instrumentation

Gamma scanning of irradiated nuclear fuel rods is performed in a shielded compartment at the HBWR, which is used for handling and inspection of irradiated materials. Inside the compartment, the measured fuel rod is placed into a gamma scanning fixture that is capable of positioning the fuel rod both axially and laterally. The compartment features a through-wall opening into which a collimator is inserted. The detector is located outside the compartment and may be inserted into a recessed opening in the collimator.

The collimator which is inserted into the compartment wall is referred to as the wall-collimator, and it has an opening size of 2 mm x 20 mm x 483 mm in length. Additional collimators, called head-collimators, may be attached to the end of the wall-collimator, extending towards the measured fuel rod. The head collimators have various sized openings where most are equipped with rectangular slits and where the smallest is 0.4 mm x 12 mm. If necessary to reduce dead-time, additional absorbers may be placed into the recess of the wall-collimator between the detector and the slit opening.

Figure 20 shows a side, cutaway, schematic view of the collimator and Figure 21 shows a photographic image of the internals of the compartment during a gamma spectroscopy measurement. The wall-collimator is observed in the lower left of the photo with no head-collimator attached, and the gamma scanning rig is observed to the right. The fuel rod is attached in the gamma scanning rig and is located in the center of the photo.
Figure 20, Side, cutaway, schematic view of the collimator used in the single-rod gamma-spectroscopy measurements. Measurements in the figure are in [cm].

Figure 21, view inside the shielded compartment where the collimator is visible to the lower left, and where the gamma scanning rig is at the right. The fuel rod is in the center of the photo.
The gamma scanning fuel rod fixture holds one fuel rod at-a-time and is positioned using motors controlled by the gamma spectroscopy measurement software. The fuel rod position may be selected axially and laterally in steps of 0.5 mm. Rotation of the fuel rod, if desired, must be done by hand.

The detector used for gamma scanning at the HBWR is a Canberra model GC2018, coaxial, P-type, High Purity Germanium (HPGe) detector. The Multichannel Analyzer (MCA) is currently an Ortec D-Spec-50.

The gamma spectroscopy software in use at the HBWR was developed in-house, and includes integrated controls of the MCA and the fuel fixture.

6.2. Gamma Tomography Device

The gamma tomography device, which was developed as part of this PhD project for measurements on complete fuel assemblies at the HBWR, was designed according to the considerations described in section 5.2, considering the fuel geometries in use at the HBWR, the intended measurement applications, and the placement and handling constraints imposed by the facility. The device is described in Paper VIII.

6.2.1. General Considerations and Specifications

The fuel that will be assessed using the tomographic device consists of fuel assemblies which have been irradiated in the HBWR, which have a maximum diameter of 73 mm, and a total length of up to 6 m, including about 40 fuel rods or less, where the active fuel region is up to 120 cm in length, and is located near the lower end of the assemblies (see Figure 6 in section 2.1). Since some of the intended measurement applications require high-resolution gamma-ray spectroscopy, an HPGe detector was selected as the detector type. Based on the properties of the fuel to be measured and the time available for fuel characterization measurements at the HBWR, a single detector was judged to be adequate.

The instrument is placed in an auxiliary building outside the reactor hall, where the measurements are performed in air. The instrument was specified to be modular to enable storage of the instrument when not in use, and also to enable installation of the instrument at other locations at the facility, e.g. in the reactor hall.

The requirements on positioning capabilities were specified to be at least as good as the capabilities of the PLUTO device [50]. Specifically, the fixtures in the PLUTO device could position the detectors/collimators axially and laterally within 0.1 mm, and rotationally, within 0.1°. Optical systems with linear encoders were used to read out the axial and lateral positions within 0.01 mm and the rotational position within 0.01°.
6.2.2. Mechanical Systems

The HBWR gamma tomography system consists of 3 basic mechanical components: 1) the fuel fixture for positioning the fuel; 2) the detector/collimator positioning fixture, for positioning the detector and collimator laterally; and 3) the radiation shielding components. In addition, a collimator is used as an integrated part of the shielding to define the fuel volume measured by the detector. Figure 22 shows a schematic overview of the HBWR gamma tomography system with a cutaway view showing the system internal components.

![Figure 22, Overview of the HBWR gamma tomography system with cutaway views showing components housed inside the radiation shielding.](image)

The system was designed around a mobile shielded compartment, which was available at the HBWR. The compartment has many features which made it suitable for use including: adequate shielding, a through-wall opening in the side through which the collimator is inserted, an opening in the top through which the fuel is inserted, a shielded glass window for viewing inside the compartment, internal lighting, and manipulators.

In addition to providing shielding, the mobile compartment is a main structural component for the system, located at the floor level. The detector/collimator fixture is located at the floor level outside the compartment. The fuel fixture is below the floor level in a shielded well under the mobile compartment. A fuel transport cask docks to the top of the mobile compartment, from which the fuel is placed into the fuel fixture. The transport cask
remains in place to provide shielding while fuel is in the instrument. The system is supported at the floor level on a 60 mm thick steel plate which rests on a pair of H-beams which in-turn rest on the floor.

Details about the mechanical components as well as photographs of the device are provided in Paper VIII. The functions of the mechanical components are summarized below.

**Fuel Fixture**
The fuel fixture rotates the fuel and positions it axially by means of a rotation fixture which is fixed to an axial linear drive. The range of movement of the axial linear drive is 145 cm, which is greater than the active fuel length of a typical HBWR assembly. A separate linear encoder was used to verify the accuracy of axial positioning and it was determined to be accurate within ± 15 μm. The rotation fixture is positioned using a dedicated servo motor with a digital encoder, where the accuracy of rotational positioning was verified to be within ± 0.01°. The precision of positioning for the axial linear drive and the rotation fixture are better than the specified precisions in section 6.2.1.

**Lateral Positioning Fixture**
The lateral positioning fixture translates the detector and collimator to the desired lateral position. It has a lateral range of 16 cm and positioning is performed using a dedicated servo motor with digital encoder. A separate linear encoder is used to read out the position where lateral the positioning is performed with a precision of ± 1.0 μm, which is better than the precision specified section 6.2.1.

**Radiation Shielding**
The radiation shielding ensures that the radiation levels remain within regulated limits for personnel working near the system.

**Collimator**
The collimator defines the volume of the fuel assemblies that contribute to the intensity in the detector. The system was designed to utilize interchangeable collimators since different slit sizes may be called for depending on the measurement application and the fuel being investigated. The collimators and the collimator shielding also feature an opening into which additional absorbers may be inserted, thereby covering the slit and providing attenuation to adjust the count rate in the detector as desired. The collimators are made of a tungsten alloy and stainless steel.
6.2.3. Data Acquisition System and Software

As mentioned above, an HPGe detector was selected for the device, due to its capability to provide high energy resolution, enabling it to resolve weak gamma peaks in the presence of stronger peaks and high background. For the measurements presented in section 8.2, an ORTEC DSPEC digital data acquisition system was employed and in-house gamma spectroscopy software was used to collect and store the gamma-ray spectra.

The measurements and movement of the system components are controlled by a LabView program, which automates the entire measurement sequence, including acquisition of the gamma spectra, and it performs some pre-processing of the data (i.e. background subtraction and peak analysis).

Tomographic reconstruction of the measured data is performed using in-house software that applies the analytical and algebraic techniques described in section 5.3 and Paper VII. The software for the algebraic reconstructions was similar to that utilized in [51].
7. Experimental Results from Gamma Scanning

7.1. Measurement of Short-lived Fission Gas Isotopes

7.1.1. Experimental Assessment of Measurable Fission Gas Isotopes

The feasibility of measuring fission gas isotopes in fuel rods with short cooling time was investigated in Paper I. Calculations were performed with the ORIGEN depletion code [52] to determine the expected quantities of xenon and krypton isotopes at several burnup steps. The calculated quantities in the fuel were evaluated together with the decay properties of the most abundant isotopes to determine which gamma-rays were expected to be emitted with the highest intensities within a short time after reactor shutdown. Based on these properties, MCNP calculations were performed to determine the effect of attenuation on a few of the highest-intensity gamma rays as they travelled from the fuel to the detector. These evaluations resulted in five candidate gamma-ray peaks emitted from three isotopes which could be expected to be present in experimental gamma-spectroscopy measurements on the gas plenum of fuel rods with short cooling time. The three isotopes and their cumulative fission yields are listed in Table 6 in section 4.3.2. The five gamma-ray energies of interest are given in Table 9 below together with their respective branching ratios.

Table 9. Five gamma peaks from short-lived gaseous fission products identified for possible measurement and the half-lives of the respective isotopes [29].

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life</th>
<th>Gamma-ray energy [keV]</th>
<th>Gamma-ray emission yield [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{133}$Xe</td>
<td>5.24 days</td>
<td>81</td>
<td>38</td>
</tr>
<tr>
<td>$^{133}$Xe</td>
<td>5.24 days</td>
<td>161</td>
<td>0.07</td>
</tr>
<tr>
<td>$^{133m}$Xe</td>
<td>2.2 days</td>
<td>233</td>
<td>10.2</td>
</tr>
<tr>
<td>$^{135}$Xe</td>
<td>9.1 hours</td>
<td>250</td>
<td>90</td>
</tr>
<tr>
<td>$^{135}$Xe</td>
<td>9.1 hours</td>
<td>608</td>
<td>2.9</td>
</tr>
</tbody>
</table>
Experimental gamma-spectroscopy measurements were performed on two fuel rods with short cooling times, of two days and 23 days, respectively, the latter using a longer measurement time. The measured gamma-ray spectra for these rods are shown in Figure 23 and Figure 24, respectively.

*Figure 23*, Gamma-ray spectrum measured in the gas plenum of a fuel rod with two days decay time. The energy range shown includes the identified gamma-ray energies. The 81 keV, 233 keV and 250 keV peaks are observable.

*Figure 24*, Gamma-ray spectrum measured in the gas plenum of a fuel rod with 23 days decay time. The energy range shown includes the identified gamma-ray energies. The 81 keV and 161 keV peaks are observable, albeit the latter at very low intensity.
In the measurement of the fuel rod with two days decay time, the 81 keV, 233 keV and 250 keV peaks were observed, and in the measurement of the rod with 23 days decay time, the 81 keV and 161 keV peaks were observed. Table 10 lists the count rates in the five candidate gamma-ray peaks for each of the measured rods. It should be noted that a wider collimator and a shorter detector-to-fuel-rod distance was used for the measurement of the rod with the longer cooling time, so the count rates are not directly comparable between the two measured rods.

Table 10, The measured count rate in each of the five gamma-ray peaks for the two measured fuel rods. The collimator was changed between the two measurements, so the count rates are not comparable between the two measured rods.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Gamma-ray energy [keV]</th>
<th>Count rate in rod with 2 days decay time [counts/second]</th>
<th>Count rate in rod with 23 days decay time [counts/second]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{133}\text{Xe}$</td>
<td>81</td>
<td>28</td>
<td>350</td>
</tr>
<tr>
<td>$^{135}\text{Xe}$</td>
<td>161</td>
<td>-</td>
<td>1</td>
</tr>
<tr>
<td>$^{133m}\text{Xe}$</td>
<td>233</td>
<td>2.5</td>
<td>-</td>
</tr>
<tr>
<td>$^{135}\text{Xe}$</td>
<td>250</td>
<td>12</td>
<td>-</td>
</tr>
<tr>
<td>$^{135}\text{Xe}$</td>
<td>608</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Based on the investigation, which is further described in Paper I, it was concluded that it was feasible to determine FGR based on a short-lived isotope in fuel rods with short cooling time, where the 81 keV gamma-rays emitted in the decay of $^{133}\text{Xe}$ were identified as the strongest candidate for measurement. The data from the 23-days cooled fuel rod was also used to determine the %FGR and to draw conclusions on the radial origin of the fission gasses, as further described in sections 7.1.2 and 7.1.3 below.

7.1.2. Measurement of Fission Gas Release Fraction in Short-cooled Fuel

Based on the feasibility investigation described in the preceding section and in Paper I, a method was proposed for determining %FGR in fuel with short cooling time based on short-lived fission products. The method is summarized in section 4.3.3 and described in more detail in Paper IV. The feasibility of the method was investigated using gamma-ray spectra previously recorded on the fuel rod mentioned in the preceding section, which had decayed for 23 days at the time of measurement. The 81 keV peak from $^{133}\text{Xe}$ is shown in Figure 25 with the X-rays from the lead shielding which surround the peak labeled in the plot. The gamma-ray spectrum measured in the
fuelled region of the fuel rod is shown in Figure 26 where the 1596 keV peak from $^{140}$Ba/$^{140}$La is observed.

Figure 25, Selected energy range around the $^{133}$Xe 81 keV peak in a gamma-ray spectrum collected from the gas plenum of a fuel rod with twenty-three days cooling time. X-rays emitted from the lead shielding surrounding the detector are labelled.

Figure 26, Gamma-ray spectrum from the fuelled region of a fuel rod with twenty-three days cooling time. Note the strong 1596 keV peak.

%FGR for $^{133}$Xe was calculated according to the method described in section 4.3.3 and was determined to be approximately 3%. The uncertainty was estimated to be approximately ±15%, based on typical uncertainties when determining %FGR based on $^{85}$Kr and $^{137}$Cs. However, the measurements were not originally performed for the purpose of investigating this method, and as such several sources of uncertainty existed, which are described in Paper IV. One particular challenge is the large difference in gamma-ray attenuation between the two isotopes used for the analysis, which requires a well-characterized detection system. One example of inconsistency in the
analysis was the ratio of \( N_{\text{fuel-code}}^{140}\text{La} / N_{\text{fuel-meas}}^{140}\text{La} \), i.e. the calculated content of \(^{140}\text{La}\) in the fuel as compared to the measured content, which was 0.15, implying a bias in the measurements or the calculation of the \(^{140}\text{La}\) content in the fuel. In spite of these difficulties and inconsistencies, the results were considered highly promising for this method to be applied in future experimental work at the HBWR.

7.1.3. Measurement of the Radial Origin of Released Fission Gasses

The feasibility of the method for determining the radial origin of released fission gasses presented in Paper III and summarized in section 4.3.4 was investigated using gamma-ray spectroscopy measurement results from the rod with 23 days decay time which is mentioned in the preceding sections. The 81 keV peak from \(^{133}\text{Xe}\) (shown in Figure 25) and the 514 keV peak from \(^{85}\text{Kr}\) (shown in Figure 27, partially obscured by the 511 keV peak) were both observed in the gas plenum measurement of this fuel rod. Using the measurement data the ratio of \(^{133}\text{Xe} / ^{85}\text{Kr}\) in the gas was calculated to be 0.033025.

![Figure 27](image-url)

*Figure 27, The 514 keV peak (partially obscured by the 511 keV peak) from a gamma-ray spectroscopy measurement of a gas plenum region of a fuel rod irradiated in the HBWR and with 23 days cooling time.*

The expected ratio of \(^{133}\text{Xe} / ^{85}\text{Kr}\) produced in the fuel was determined through detailed calculations and determined to be 0.115. Since the ratio of \(^{133}\text{Xe} / ^{85}\text{Kr}\) in the gas was found to be lower than the ratio produced in the fuel, the results indicate that the gasses were released from the central regions of the pellet, which is consistent with the fuel properties and experimental conditions. More details are presented in Paper III.
7.2. Scanning of Long-cooled Fuel Which Had Operated at High Power

Gamma spectroscopy measurements were performed on the fuel stack and gas plenum regions of nine HBWR driver fuel rods using the gamma spectroscopy setup described in section 6.1. The experimental results are described in Paper V.

The nine fuel rods were selected from two parent fuel assemblies. Five rods were selected from a parent assembly with a burnup of 50.4 MWd/kgUO₂ and four rods were selected from a parent assembly with a burnup of 24.6 MWd/kgUO₂. Both fuel assemblies had operated at high LHRs, where the higher-burnup assembly and the lower-burnup assembly operated at a maximum assembly-average LHRs of 58 kW/m and 68 kW/m, respectively. This high LHR is much higher than what is typically encountered in commercial fuel, implying that significant FGR and migration of volatile fission products such as Cs can be expected. (See section 2.2.1.)

Examples of the gamma ray spectra measured in the fuel stack and plenum regions of a high burnup rod are shown in Figure 28 and Figure 29, respectively, where the 662 keV peak from $^{137}$Cs is observed in the fuel spectrum and the 514 keV peak from $^{85}$Kr is labeled in the plenum spectrum. Figure 30 shows the energy range around the 514 keV peak from the spectrum in Figure 29.

![Gamma-ray spectrum from the fuelled region of an HBWR driver fuel rod which had operated at a high LHR to a burnup of 50.4 MWd/kgUO₂ and had decayed for approximately 21 years at the time of measurement. Note the prominent peak from $^{137}$Cs at 662 keV.](image)

*Figure 28,* Gamma-ray spectrum from the fuelled region of an HBWR driver fuel rod which had operated at a high LHR to a burnup of 50.4 MWd/kgUO₂ and had decayed for approximately 21 years at the time of measurement. Note the prominent peak from $^{137}$Cs at 662 keV.
Figure 29. Gamma-ray spectrum from the gas plenum region of an HBWR driver fuel rod which had operated at a high LHR to a burnup of 50.4 MWd/kgUO₂ and had decayed for approximately 21 years at the time of measurement. The ⁸⁵Kr peak at 514 keV is labeled.

Figure 30. Energy range around the 514 keV peak from the spectrum in Figure 29.

The axial and azimuthal distributions of ¹³⁷Cs were characterized in selected regions of all fuel rods. Figure 31 shows an example of an axial scan of ¹³⁷Cs from 220 mm to 265 mm height on a fuel rod where two different measurement setups were used. The results were similar for all rods and indicate that significant ¹³⁷Cs migration had occurred, consistent with operation at the high LHRs of these rods. Figure 32 shows an example of the azimuthal variation in ¹³⁷Cs for the same rod at the 244 mm elevation, indicating that the radial migration of ¹³⁷Cs was most significant towards the 270° orientation of the fuel rod.
Figure 31, Axial scan of $^{137}$Cs in the 220 mm – 265 mm axial range using two different measurement setups; one with a narrow collimator slit (lower curve) and one with a wider collimator slit. The data from the narrow slit shows that significant migration of Cs has occurred to the pellet-pellet gaps, and thus radial migration can also be expected to have occurred.

Figure 32, Azimuthal variation of $^{137}$Cs in one fuel rod at the 244 mm elevation.

%FGR was calculated for each of the nine rods using $^{85}$Kr and $^{137}$Cs based method described in section 4.3.3. %FGR was calculated to be approximately 24% in each of the five higher-burnup rods and approximately 18% in each of the four lower-burnup rods. This is also consistent with the operation at high LHRs. The results are discussed in more detail in Paper V.

The measurement of FGR and Cs migration from research reactor fuel operated at very high LHR enhances the range of operating conditions for which this type of data is available, and thus it is valuable for understanding the physical processes behind these phenomena. Furthermore, this particular set of data is useful for the interpretation of the tomographic measurements presented in section 8, since these nine rods were assembled into a fuel assembly configuration and measured tomographically.
8. Results from Gamma Tomography

As discussed in section 5, gamma tomography may theoretically provide similar information on individual fuel rods as gamma scanning, albeit without the need to dismantle the fuel assemblies. In order to evaluate which fuel properties that are practically assessable using gamma tomography, investigations have been performed by means of computer simulations and also by experimental investigations using the device presented in section 6.2. The results from simulation studies have been presented in Papers II and VI, whereas initial experimental results have been presented in Paper VIII. The results from these papers are summarized below.

8.1. Simulations

8.1.1. Simulation Tools and Setups

Simulations of different gamma tomography measurement applications have been performed using the particle-transport code MCNP [53] for two HBWR fuel designs, representing the gas plenum region of an 18-rod assembly, as well as the fuel stack and gas plenum regions of a 9-rod assembly. The 18-rod assembly was simulated in a study of the capability to identify a leaking fuel rod, which is presented in Paper II. The 9-rod assembly was simulated in a study of the capability to determine distributions of $^{137}$Cs in the fuel stack and $^{85}$Kr in the gas plenum, and thus of the capability to tomographically determine the %FGR in the individual fuel rods, as presented in Paper VI. The measurement geometry simulated corresponds to the measuring device described in section 6.2 and Paper VIII.

In order to save simulation time, the MCNP simulations incorporated several simplifications including the following:

- The fuel assemblies were truncated to only a few cm in the axial direction.
- An artificial collimator package with tightly stacked slit openings was simulated to obtain information from a large number of lateral collimator positions simultaneously.
- Source biasing was applied to emit the gamma rays preferentially towards the collimator openings.
• Energy cutoffs were used such that gamma rays with energies 2-3 keV below the desired energy were not tracked in the simulations – a simplification justified by the use of an HPGe detector with high energy resolution in the simulated measuring device.

• In all models, the fuel rods were positioned in their nominal positions and the fuel assembly was positioned with its center at the center of rotation in the measurement system. Accordingly, only a subset of the angular measurement positions needed to be simulated and symmetry was used to construct the remaining simulated projections.

8.1.2. Rod-wise Fission Product Distributions

The simulations presented in Paper VI corresponded to measuring the $^{137}$Cs distribution in the fuel stack region and the $^{85}$Kr distribution in the plenum region of a nine-rod HBWR driver fuel assembly. The simulated fuel stack measurements utilized a collimator slit of 1 mm in width, 22 mm in height, and 755 mm in length. The simulated lateral step size was 2 mm, and the angular step size was 3°. The simulated source was monoenergetic 662 keV gamma rays from $^{137}$Cs. The simulated plenum measurements of 514 keV gamma rays from $^{85}$Kr utilized a collimator slit of 3 mm in width, 30 mm in height and 755 mm in length with a simulated lateral step size of 4 mm, and an angular step size of 3°. The plenum of each fuel rod was modeled as containing He and a series of slanted Inconel rings in order to approximate the plenum springs.

The rod-wise source activities of 662 keV and 514 keV gamma rays corresponded to the measured activities in the nine fuel rods which were experimentally characterized using gamma spectroscopy measurements as described in section 7.2 and Paper V. However, since no reliable information was available on the internal distribution of fission products in the rods, a homogeneous distribution of the source in each fuel rod was simulated. This is realistic for the gaseous $^{85}$Kr distribution, whereas the gamma scanning data presented in section 7.2 indicated that significant migration of $^{137}$Cs had occurred, making the homogeneous distribution of $^{137}$Cs less realistic. The uncertainty in the simulated tally results was less than 1% for the $^{137}$Cs and $^{85}$Kr simulations in the measurement positions with the highest intensity.

The simulated measurement results were reconstructed using the analytic reconstruction technique described in section 5.3.1 and Paper VII. The resulting images of the simulated 662 keV and 514 keV source distributions in the fuel and gas plenum, respectively, are shown in Figure 33. Qualitatively, the images correspond well to the simulated source distributions. The wider collimator slit simulated for the $^{85}$Kr measurements implies lower spatial resolution in the right image.
The simulated measurement results were also reconstructed using the algebraic rod-activity reconstruction method described in section 5.3.2 and Paper VII. The reconstructed rod-wise contents of the 662 keV source in the fuel and the 514 keV source in the gas plenum matched the simulated source contents within 0.23 % (1σ), and 0.13 % (1σ), respectively.

8.1.3. Rod-wise Fission Gas Release Fraction

Based on the reconstructed rod-wise distributions of 85Kr and 137Cs from the simulated measurements in the previous section, the rod-wise fission gas release fraction of 85Kr was calculated for each rod according to the method described in section 4.3.3. A modified version of Equation 7 was used since some of the terms are accounted for in the reconstruction when using the Algebraic Reconstruction Technique. The calculations are described in more detail in Paper VI.

The rod-wise %FGR calculated based on the simulated gamma tomography measurements was systematically a factor of approximately 2.7% lower than the rod-wise %FGR calculated for the same rods in the experimental measurements described in section 7.2 and Paper V. The deviation may arise due to inconsistent consideration of the plenum and fuel collimator dimensions in the MCNP simulations and the reconstruction software, e.g. with respect to the plenum spring. It is believed that such systematic differences may be taken into account in a real measurement by means of calibration based on a benchmark using e.g. destructive assay.

8.1.4. Leaker Rod Identification

In Paper II, a novel application of gamma tomography for locating failed fuel rods in a fuel assembly was investigated. The investigation consisted of
simulated measurements of an 18-rod HBWR-type fuel assembly, where one of the fuel rods contained much less fission gas than the other rods in order to represent the case where a failed rod had expelled most of its fission gas inventory. Figure 34 shows an illustration of the modeled fuel assembly where the shading represents the source activity in the rods and the leaking rod is labeled.

![Illustration of the simulated 18-rod fuel assembly](image)

*Figure 34, Illustration of the simulated 18-rod fuel assembly where the shading in each rod represents the source activity and where the leaking rod is labeled.*

The simulated measurement setup was such that the collimator slits were 1 mm wide, 20 mm in height, and 500 mm in length. 41 lateral positions were simulated at a lateral step size of 2 mm. The angular step size was 3°. Simulations were performed where the fuel was filled with water and alternately with air in order to investigate the measurement applicability for each of those conditions. Three gamma-ray source energies were used: 81 keV gamma rays from $^{133}$Xe, 250 keV gamma rays from $^{135}$Xe and 514 keV gamma rays from $^{85}$Kr. These gamma-ray energies were selected based on the feasibility study described in section 7.1.1 and Paper I.

The simulated results for measurements in air were reconstructed using the FBP method described in section 5.3.1 and Paper VII. The resulting images are shown in Figure 35 for 81 keV gamma-ray source (left), the 250 keV gamma-ray source (middle) and the 514 keV gamma-ray source (right).
The results indicate that it is feasible to utilize gamma tomography to locate failed fuel rods in a nuclear fuel assembly; however, the following considerations concerning the isotopes and gamma-rays available for measurement must be considered when planning this type of measurement:

- The 81 keV gamma rays from $^{133}\text{Xe}$ are strongly attenuated in the construction materials, so its usefulness for leaker rod identification in fuel assemblies with dense rod-patterns is questionable.
- $^{135}\text{Xe}$ has a short half-life ($T_{1/2} = 9.14$ hours), so it must be measured very quickly after the fuel is removed from the reactor, and its decay during the measurements must be accounted for.
- The 514 keV gamma rays are emitted in only a small fraction of $^{85}\text{Kr}$ decays, and $^{85}\text{Kr}$ has a small fission yield, resulting in a low intensity in the 514 keV peak. Additionally, interfering peaks at 511 keV and 512 keV may obscure the 514 keV peak.

Furthermore, an actual fuel assembly will contain a variation of concentrations of fission gasses in each fuel rod, which will complicate the interpretation of the measured images. In Paper II, it is suggested to base the interpretation on predicted rod-by-rod gas contents.

8.2. Experimental Measurements

Paper VIII describes experimental gamma tomography measurements that have been performed on a nine-rod HBWR driver fuel assembly. The gamma tomography instrument used is described in Paper VIII and section 6.2.

The nine-rod fuel assembly that was measured consisted of the nine fuel rods previously characterized using single-rod gamma spectroscopy (see section 7.2 and Paper V). Two gamma tomography measurements were per-
formed, which were similar to the simulated measurements presented in section 8.1.2 and Paper VI. Specifically, the measurements consisted of one measurement on the fuel stack region and one measurement on the plenum region.

As opposed to the simulations, the fuel stack and plenum region measurements utilized the same collimator, which has a slit size of 1 mm in width and 22 mm in height and with a length of 755 mm. In the fuel measurements, a 20 mm stainless steel absorber was inserted into the collimator slit in order to reduce the count rate in the detector. For the plenum measurements a 1 mm thick copper absorber was inserted into the collimator slit in order to reduce the count rate in the low-energy region of the measured spectra.

The fuel and plenum measurements were performed with a lateral step size of 1 mm over a lateral range of 72 mm. An angular step size of 3° was used and measurements were performed at 120 angular positions. The live time at each measurement position for the fuel stack measurements was 7 seconds. For the plenum measurements the live time was 100 seconds at each position.

Tomographic reconstruction was performed on the data using four characteristic gamma-ray peaks observed in the measured spectra. The four peaks and their emitting isotopes are listed in Table 11. Examples of typical spectra from the fuel stack and plenum regions are shown in Figure 36 and Figure 37, respectively.

Table 11, Gamma-ray peaks observed in the measured spectra and for which tomographic reconstructions were performed.

<table>
<thead>
<tr>
<th>Gamma ray [keV]</th>
<th>Isotope</th>
<th>Plenum/Fuel measurement</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>426</td>
<td>$^{178m}$Hf</td>
<td>Plenum</td>
<td>Activation product in construction material</td>
</tr>
<tr>
<td>514</td>
<td>$^{85}$Kr</td>
<td>Plenum</td>
<td>Gaseous fission product</td>
</tr>
<tr>
<td>662</td>
<td>$^{137}$Cs</td>
<td>Fuel</td>
<td>Fission product that migrates at high temperatures</td>
</tr>
<tr>
<td>1173</td>
<td>$^{60}$Co</td>
<td>Plenum</td>
<td>Activation product in plenum springs.</td>
</tr>
</tbody>
</table>
Figure 36, Gamma-ray spectrum recorded in the fuel-stack region of the nine-rod HBWR fuel assembly measured tomographically using the device described in Paper VIII. Note the 662 keV peak from $^{137}$Cs.

Figure 37, Gamma-ray spectrum recorded in the plenum region of the nine-rod HBWR fuel assembly measured tomographically using the device described in Paper VIII. Note the two strong $^{60}$Co peaks at 1173 keV and 1332 keV. The 514 keV peak from $^{85}$Kr, and the weak 426 keV peak from $^{178m}$Hf are labeled.

8.2.1. Fission Product Distributions

The measurement data from the two fission products; $^{137}$Cs in the fuel stack region of the assembly, and $^{85}$Kr in the gas plenum region of the assembly were reconstructed using the FBP method described in section 5.3.1 and Paper VII. Figure 38 shows the reconstructed distributions of $^{137}$Cs in the fuel stack region (left) and of $^{85}$Kr in the gas plenum region (right). In these images, white corresponds to the maximum activity and black corresponds to the minimum activity.
From Figure 38 (left) it can be noted that the $^{137}\text{Cs}$ has migrated to the outer region of the fuel pellets, which is consistent with the fuel rods’ operation at high LHRs and consistent with the observations from the gamma scanning measurements (see section 7.2). The uniform distribution of the $^{85}\text{Kr}$ gas within each fuel rod’s gas plenum is observed in Figure 38 (right). In both images in Figure 38, the five high-burnup rods can be easily identified, having a higher content of $^{137}\text{Cs}$ as well as $^{85}\text{Kr}$.

8.2.2. Activation Product Distributions

The data from the two activation products $^{60}\text{Co}$ and $^{178m}\text{Hf}$ in the plenum measurements was reconstructed using the FBP method described in section 5.3.1 and Paper VII. Figure 39 shows the reconstructed distributions of $^{60}\text{Co}$ (left) and of $^{178m}\text{Hf}$ (right). In these images, white corresponds to the maximum activity and black corresponds to the minimum activity. The $^{60}\text{Co}$ distribution reveals the plenum springs in the gas plena of the fuel rods, while the $^{178m}\text{Hf}$ distribution reveals the tie-rods.

Figure 39. Reconstructed images of the rod-wise $^{60}\text{Co}$ distribution in the plenum region (left) and the rod-wise $^{178m}\text{Hf}$ distribution in the plenum region (right).
Some interesting features can be identified in Figure 39: First, the diameter of the plenum springs in the $^{60}$Co image is smaller than the rods in Figure 38 (left) and the gas plenum diameter in Figure 38 (right), and the primary interpretation is that the plenum springs have a smaller diameter than the fuel pellets and the inner radius of the cladding. Second, there are seven highly activated plenum springs in the image of the $^{60}$Co distribution and not five as one would have expected. Accordingly, the springs in two of the four low-burnup rods have apparently been almost as strongly activated as the springs in the high-burnup rods. The reason for this is so far unknown, but it may likely be found in somewhat different material compositions in the springs. Third, the only feature visible in the $^{178m}$Hf image is the tie rods, indicating that this material is dominating in terms of this type of activation, which may also be deferred to the material composition of the construction material and an irradiation history of the tie-rods which differs from that of the fuel rods (the rods and fuel assembly structure measured here were previously not irradiated together).

In conclusion, these images show that this type of data can bring highly valuable information on fuel performance and material properties. In this context, one should note that one tomographic measurement can return a multiple of isotopic distributions, which in-combination brings additional information.
9. Conclusions

9.1. Gamma Spectroscopy

The feasibility of measuring and analyzing fission products using single-rod gamma spectroscopy has been investigated theoretically and experimentally. The novel applications investigated are based on measuring gamma-rays emitted in the decay of short-lived fission gas isotopes in the gas plena of fuel rods with short cooling time:

- Experimental measurements were used to investigate which gamma-rays from a selection of short-lived fission gas isotopes were feasible for non-destructive gamma-ray spectroscopy measurements. Gamma-rays emitted in the decay of $^{133}$Xe, $^{135}$Xe and $^{133m}$Xe were experimentally shown to be available for measurement at 2 days after the measured fuel was discharged from the reactor. $^{133}$Xe was also shown to be feasible for measurement in fuel 23 days after discharge.

- A method for determining %FGR based on measurement of short-lived fission products was developed and demonstrated using experimental measurements. Specifically, the 81 keV gamma rays emitted in the decay of $^{133}$Xe were measured in the gas plenum and, together with a measurement of the 1596 keV gamma rays emitted in the decay of $^{140}$La in the fuel stack, and core physics code results, the %FGR for $^{133}$Xe was determined.

- A method for investigating the origin of released fission gasses based on non-destructive single-rod gamma-ray spectroscopy measurements of two fission gas isotopes, $^{133}$Xe and $^{85}$Kr was proposed and experimentally demonstrated.

The measurement applications which are listed above are foreseen to be most applicable in research reactor settings where fuel handling constraints and laboratory measurement conditions permit disassembly of the fuel and measurement of the short-lived isotopes in individual fuel rods.

In addition to applications based on measuring short-lived isotopes, experimental measurements were performed in order to characterize HBWR driver fuel rods which had operated at high LHRs and which had decayed for approximately 21 years at the time of measurement. Significant axial redistribution of $^{137}$Cs as well as high %FGR was observed in each rod, consistent with the fuel rods’ operation at high LHRs.
9.2. Gamma Tomography

Gamma tomography combines gamma spectroscopy with tomographic measurement schemes and reconstruction of cross-sectional source distribution in the measured objects. In this thesis, the tomographic assessment of complete fuel assemblies in order to nondestructively extract data on the individual fuel rod level has been investigated, and tomographic reconstruction algorithms developed specifically for the application on nuclear fuel assemblies have been presented.

Gamma tomography measurements were simulated using MCNP to investigate several measurement applications, including locating a failed rod in an assembly and determining the rod-wise distribution of fission products within fuel assemblies. Leaker rod identification measurements were simulated based on measuring the 81 keV gamma rays from $^{133}$Xe, the 250 keV gamma rays from $^{135}$Xe, as well as the 514 keV gamma rays from $^{85}$Kr. The simulated measurement data was reconstructed using the analytic FBP method and indicate that measurements of the two latter could be feasible, although some experimental challenges have been identified, which have to be addressed in an actual measurement.

Gamma tomography measurements of the fuel and gas plenum regions of an HBWR fuel assembly were also simulated and the data was reconstructed using both analytic and algebraic reconstruction techniques. The analytic reconstructions qualitatively represent the simulated source distributions in terms of cross-sectional images and the algebraic reconstructions result in quantitative rod-by-rod data. The latter type of reconstruction showed good agreement between the reconstructed and simulated source distributions, where the deviations were 0.23 % ($1\sigma$) for the rod-wise $^{137}$Cs content in the fuel stack and 0.13 % ($1\sigma$) for the $^{85}$Kr content in the gas plenum.

A gamma tomography instrument was designed, constructed and experimentally demonstrated at the HBWR, where the measured fuel consisted of a nine-rod HBWR driver fuel assembly. The measurement data for two fission products and two activation products was reconstructed using the analytic FBP technique to produced images of the source distribution in the fuel.

The reconstructed images of the fission product distribution revealed that $^{137}$Cs had migrated to the outer pellet surfaces, consistent with operation at high LHRs, and the distribution of $^{85}$Kr in the plenum region of the fuel was qualitatively consistent with the rod-wise distribution determined from previously-performed single-rod gamma-ray spectroscopy measurements on the same fuel rods. The reconstructed images of the activation products in the plenum region of the fuel assembly revealed the locations of the plenum springs and the structural tie rods. These measurements showed that one tomographic measurement can return a multiple of isotopic distributions, which in combination can brings valuable information on fuel performance and material properties.
10. Outlook

10.1. Gamma Spectroscopy

The gamma spectroscopy applications investigated and experimentally demonstrated in this work, have been shown to be feasible and may be applied on experimental fuel with short cooling time at the HBWR as part of the PIE performed on experimental fuel. In fact, measurements of the ratio of $^{133}\text{Xe}/^{85}\text{Kr}$ have already been performed on experimental fuel [36].

Since the novel applications proposed in this work require the fuel to be disassembled for measurement of short-lived fission gasses, it is not likely to be executed on fuel at commercial reactors, where there are constraints on disassembling fuel at short cooling times.

10.2. Gamma Tomography

10.2.1. Future Measurements and Analyses at the HBWR

The gamma tomography measurement system developed at the HBWR has been successfully demonstrated through experimental measurements on a fuel assembly made of rods with long cooling time. Measurement data from the fuel stack and the gas plenum region was reconstructed using the analytic FBP method, returning qualitative cross-sectional images of the fuel at each axial level. It is foreseen that the gamma tomography system will become an important part of the PIE capabilities at the HBWR since, without the need to disassemble the fuel, it has the capability to non-destructively characterize experimental fuel assemblies which may not otherwise be dismantled for characterization. Rod-wise characterization of fuel assemblies using gamma tomography will also save time and reduce dose to workers in comparison to the alternative of gamma scanning measurements of individual fuel rods.

The FBP method as applied to nuclear fuel assemblies in this work yields images which are useful mainly for qualitative assessment of the radioactive source distribution in the fuel. As such, the precision for reconstructing the quantitative activity distribution in the fuel has not yet been evaluated. In order to evaluate the precision of the instrument, the algebraic reconstruction method presented in this work should be applied to the measurement data,
similar to what was demonstrated for simulated data. However, some challenges that must be addressed in the analyses of the measured data are: (1) the strong migration of $^{137}$Cs to the outer surface of the pellets, which was observed in the measured projections, and (2) the dislocation of individual fuel rods from their nominal positions, which could be observed in the reconstructed images, and possibly also dislocations of the tie rods. The former challenge is illustrated in Figure 40 in terms of a simulated projection for a homogeneous distribution of $^{137}$Cs in each fuel rod as compared to a measured projection with the highly inhomogeneous distribution encountered in the actual fuel.

![Figure 40](image)

*Figure 40.* A drawing of the nine-rod fuel assembly configuration that was measured (left) shown together with simulated $^{137}$Cs intensities (middle) and the measured $^{137}$Cs intensities (right) at the illustrated angular position. Due to Cs migration, the measured intensities show a significantly different distribution as compared to the simulated ones. The internal source distribution in the fuel rods constitutes one of the challenges encountered when performing quantitative assessment of the rod-by-rod source contents.

In order to perform quantitative reconstructions, the internal source distribution in the rods may be taken into account by deploying an adequate pixel pattern, and the locations of fuel rods and the structural components may be obtained from analysis of reconstructed images. This type of analysis will be subject for further studies; however, the qualitative images have already proven useful for assessing fuel performance and material properties.

The gamma tomography system at the HBWR is currently placed in the bunker building, which is outside the reactor hall. Due to handling constraints at the facility, fuel may not normally be transported outside the reactor hall until it has been allowed to decay for several days, such that many of the short-lived isotopes decay prior to the fuel being available for measurement outside the reactor hall. The HBWR gamma tomography system has a modular construction such that it may be installed in the reactor hall. Placement of the gamma tomography system in the reactor hall, would enable
measurement of fuel with cooling times on the order of hours or days, such that many short-lived isotopes may be measured, potentially leading to new measurement applications for investigation of fuel performance.

10.2.2. Novel Applications of Tomography on Research Reactor Fuel

**Rod-wise Determination of the Fission Gas Release Fraction Based on Short-lived Isotopes \(^{133}\text{Xe} \text{ and }^{140}\text{Ba}^{140}\text{La}\)**

The gamma spectroscopy method described in section 4.3.3 and Paper IV, may be extended to implementation in gamma tomography measurements. Determining the rod-wise fission gas release fraction of short-lived isotopes is especially useful at the HBWR since it is a research facility where the fuel may be operated in unusual conditions, such as power transients, and loss-of-coolant and similar accidental conditions, and also since the operation cycles may be short such that the inventory of the short-lived isotopes corresponds to the entire experimental irradiation cycle. For this application, tomographic measurements will be useful to enable the assessment of instrumented fuel and allow for reinsertion of the fuel in the core after assessment.

**Rod-wise Determination of rod-wise ratio of released \(^{133}\text{Xe} /^{85}\text{Kr}\)**

As mentioned above, the single-rod application of determining the \(^{133}\text{Xe} /^{85}\text{Kr}\) ratio which has been presented in this work may also be extended for use with gamma tomography. Again, tomographic measurements will be useful to enable the assessment of instrumented fuel and allow for reinsertion of the fuel in the core after assessment.

10.2.3. Commercial Application of Tomography

Gamma tomography has previously been experimentally demonstrated on commercial fuel for determining the rod-wise power distribution [19]; however, the experimental device in [19] was not designed for industrial use and easy transport between NPPs. The device in [19] utilized a design in which the detectors rotated around the fuel, meaning that the device was rather large. The device presented in this work utilizes a design in which the fuel rotates and the detector translates. In this design, the instrument may remain compact such that it is relatively lightweight and easily transportable.

The experience gained with the HBWR device and the mechanisms for positioning the fuel and detector/collimator are expected to be useful in designing and building an eventual gamma tomography system for use at commercial NPPs. In order to reduce measurement times, such a system could utilize several detectors, with each detector attached to a separate collimator slit, as opposed to the one-detector system developed for the HBWR.
As compared to conventional gamma scanning, such a system could also enable the assessment of short-cooled fuel at commercial facilities, since it would not require disassembling of the fuel for measurement.
Acknowledgements

First of all, I’d like to thank the organizations that funded this research: The Swedish Research Council (Vetenskapsrådet), Westinghouse Electric Company, and the Norwegian Institute for Energy Technology – OECD Halden Reactor Project.

It’s been about five years since I started this work, and during this time I’ve been simultaneously employed by Westinghouse in Västerås, Sweden, by Uppsala University in Uppsala, Sweden, and the OECD Halden Reactor Project in Halden, Norway. Over these past five years I’ve had offices in all three places and I’ve travelled back and forth between Västerås and Uppsala and Halden and Uppsala many, many times. I’ve had the privilege to work with many talented people along the way, who I will now try to acknowledge.

At Westinghouse, Dr. Christofer Willman and Dr. Lars Hallstadius were instrumental in initiating this project and obtaining funding from within Westinghouse and from the Swedish Research Council. Christofer and Lars have also both served as official members of the reference group for my Ph.D. studies. While I haven’t been located in Västerås since 2010 we have usually had the occasion to meet at least a couple of times per year at the reference group meetings in Uppsala.

Speaking of reference group meetings, I would also like to mention the other official members Professor Ane Håkansson, and my Ph.D. supervisor Associate Professor Staffan Jacobsson Svärd, as well as the unofficial members from Westinghouse, Juan Casal and Björn Andersson. I have looked forward to all of the reference group meetings. All of you, Lars, Christofer, Ane, Staffan, Juan, and Björn, always gave very helpful feedback and we had many very productive discussions, which I have very much enjoyed.

For these past five years I’ve shared an office at the Ångström Laboratory in Uppsala with a fellow Ph.D. student, Dr. Peter Andersson; however, I was only located in Uppsala for the first year of my Ph.D. studies. Peter has been the main beneficiary of this since he has effectively had his own private office for at least four years. You’re welcome Peter!

When I was about one year into my Ph.D. studies the OECD Halden Reactor Project joined the project. From Westinghouse’s side, Lars Hallstadius was instrumental in coordinating this cooperation and from Halden, Dr. Margaret McGrath, and Carlo Vitanza agreed to host the project in Halden. Ultimately this has been a great advantage for this work since it allowed easy
access to the Halden research facilities. Thank you Margaret and Carlo for making this happen!

I should probably acknowledge just about everyone in Halden. The design, construction and experimental demonstration of the gamma tomography instrument has been a huge effort with involvement from probably dozens of people. I have worked closely with Knut Eitrheim from the very beginning of my time here in Halden. Knut has answered about 1 million questions (sorry Knut!), and we have had lots of fun looking at gamma spectra and trying new measurements. Cato Edvardsen and Henrik Lersbryggen have spent countless hours designing the mechanical components of the gamma tomography system. The workshops deserve credit for precision manufacturing of the various the components, and the crew at the reactor also deserves a huge amount of credit for doing all of the necessary work to install and use the system in the Bunker building. Morten Limi has done the programming work for the software which operates the equipment, which I am very impressed with. The measurement system worked on the first try with no problems - you have all obviously done a great job! I am very impressed with the work that everyone has done.

In addition to all of the people that were involved in my project I would also like to acknowledge all of my other colleagues in the office that have made working here in Halden so extremely enjoyable.

Now to Staffan… Staffan, I have immensely enjoyed these past five years as your Ph.D. student. Although I have been far away from Uppsala most of the time, I feel like we have worked very well together. I have learned a lot from you and I sincerely hope that we can continue to work together in the future. I have really enjoyed it.

Finally, I would like to thank my wife, Heather. You have been very patient and supportive during these past five years. I know my odd working hours and sometimes being distracted were not always easy - Thank you for understanding and being there to keep me going!

This thesis is due in 30 minutes! I’d better stop writing…
Idag finns det 434 kärnkraftsreaktorer i världen, som tillsammans står för cirka 11% av världens elproduktion. Det finns också 266 forskningsreaktorer där huvudsyftet är inte att producera el utan att genomföra forskning på t.ex. de material som används i kommersiella kärnkraftverk. Kärnbränslet utgör källan till den energi som frigörs genom kä rnklyvning in en reaktor, och det är även bränslet som innehåller de radioaktiva ämnen som produceras till följd av klyvningen. På grund av dessa funktioner, medför bränslet självt en stor mängd begränsningar i hur ett kärnkraftverk drivs med hänsyn till effektuttag, vattenkemi, och så vidare. Det är således av stort intresse att förstå bränslets egenskaper och beteenden för att göra kärnkraftsdriften så säker och effektiv som möjlig och därigenom för att begränsa mängden kärnavfall som produceras.

**Kärnbränsle**

Kärnbränsle består av kutsar gjorda av urandioxid som staplas i små metalliska rör, som även kallas för bränslestavar, vilka monteras i bränsleelement. I ett kärnkraftverk är bränslestavarna ungefär 1 cm i diameter och 4 m i längd. Ett bränsleelement består typiskt av mellan 100 och 300 stavar, och en reaktorhärd innehåller ungefär 200-800 patroner – beroende på bränsle- och reaktortyp.

Temperaturfördelningen i härden, bestrålningen och den kemiska miljön förorsakar många ändringar i bränslets egenskaper. Kutsarnas och rörens fysikaliska mått och kemiska innehåll ändras under bestrålningen, och till följd av dessa ändringar försämras bränslets egenskaper under tiden i reaktorn. Ju längre bränslet bestrålas, desto sämre blir beteendet under normaldrift samt i händelse av eventuella olycksförlopp. Alltför höga temperaturer kan också orsaka snabba försämringar av kutsarnas och rörens prestanda genom ökad hastighet på kemiska reaktioner samt ökat tryck inuti stavarna.

När kärnbränslet bestrålas i en härdf upptar många nya ämnen i materialet. Dessa ämnen är klyvningsprodukter, aktiveringsprodukter och så kallade transuraner, som alla ofta är radioaktiva. När dessa radioaktiva ämnen sönderfaller sänder många av dem ut gammastrålning med energier som är karakteristiska för ämnet i fråga. Genom att analysera gammastrålningsfältet kring kärnbränslet kan man analysera vilka ämnen som finns i bränslet samt mängden av dessa ämnen. Med kunskap om hur dessa ämnen byggs upp under bestrålningen kan man dra slutsatser om hur bränslet har bestrålats och
om hur det har betett sig under bestrålningen. Denna avhandling handlar om mätning av gammastrålning med två metoder: *gammaspektroskopi* och *gammatomografi*, vilka beskrivs nedan.

**Gammaspektroskopi**

Metoden gammaspektroskopi består av mätning av gammastrålningen som emitteras från ett bestämt område i ett kärnbränsleelement och analys av den uppmätta energifördelningen, också kallat *energispektrum*. Topparna i ett spektrum motsvarar de ämnen som sänder ut gammastrålningen av denna energi. Typiska mätningar som görs med gammaspektroskopi är bestämning av effektfördelningen i bränslet, hur mycket energi som har tagits ut totalt över bränslet livslängd (också kallat *utbränning*), samt bestämning av hur stor del av de gasförmiga klyvningsprodukterna som har lämnat bränsle Kut-sarna och samlats i rören (vilket också kallas *fissionsgasavgivning* och mäts i procent).

I denna avhandling har gammaspektroskopiska mätningar på kortlivade klyvningsprodukter undersökts genom teoretiska och experimentella studier. Tre gasförmiga produkter har identifierats och mätts i en stav som bestrålats i en forskningsreaktor i Halden i Norge. Då utförbarheten bevisades föreslogs en metod för bestämning av andelen fissionsgasavgivning genom gammaspektroskopiska mätningar av klyvningsprodukterna $^{133}$Xe i rören och $^{140}$Ba i bränsle kut-sarna. Ytterligare en metod föreslogs baserat på samtidiga mätning av produkterna $^{133}$Xe och $^{85}$Kr, som har som syfte att undersöka den ur sprungliga platsen i kutsen varifrån fissionsgasser frigörs. Dessa två föreslagna metoderna har även demonstrerats genom experimentella mätningar.

Gammaspektroskopiska mätningar har också gjorts på de två långlivade klyvningsprodukterna $^{137}$Cs och $^{85}$Kr i bränslekutsarna respektive i rören i bränslestavar som körts på hög effekt i Haldenreaktorn och som hade kylts i 21 år vid mätningstillfället. Två slutsatser kunde dras; dels hade $^{137}$Cs förflyttat sig inuti kut-sarna under bestrålningen och dels hade en stor andel av de gasförmiga klyvningsprodukterna lämnat bränslet. Både dessa observationer hör samman med höga temperaturer och var i enlighet med den höga effekten detta bränsle hade utsatts för.

**Gammatomografi**

Gammatomografi är en metod som baseras på gammaspektroskopi och to- mografiska rekonsstruktionsmetoder. När gammatomografi tillämpas på kärnbränsleelement kan man bestämma den stavvisa aktivitetsfördelningen utan att behöva plocka isär bränslet för att mäta enstaka stavar. Därigenom kan man spara mycket tid och man undviker eventuella skador som kan uppstå när bränslet demonteras. Gammatomografimätningar består av två steg:

(1) Gammaspektroskopimätningar av strålningsfältet runt ett kärn- bränsleelement i ett stort antal punkter kring bränslet.
(2) Återskapande av aktivitetsfördelningen inuti bränsleelementet genom matematiska beräkningar, så kallade tomografiska rekonstruktioner av uppmätta data. I denna avhandling beskrivs och används några olika rekonstruktionsmetoder.

Ett instrument för genomförande av gammatomografiska mätningar på bränsle från forskningsreaktorn i Halden har konstruerats, monterats och använts för experimentella mätningar vid Haldenreaktorn.

Simulerade tomografiska mätningar
Datatorsimuleringar har genomförts som modellera tomografiska mätningar av Haldenbränsle för att undersöka nya tillämpningar av gammatomografimetoden samt för att undersöka den förväntade prestandan av utrustningen som byggdes.

En ny tillämpning var att lokalisera var i ett bränsleelement en stav som läcker ut gasformiga klyvningsprodukter befinner sig genom att rekonstruera aktiviteten i rören och hitta den stav som har förlorat sitt förväntade gasinnhåll. De mätningarna som simulerades använde gammaenergier från tre gasformiga klyvningsprodukter: \(^{133}\text{Xe}\), \(^{135}\text{Xe}\), och \(^{85}\text{Kr}\). Figur 1 visar de rekonstruerade aktivitetsfördelningarna i dessa tre fall. Simuleringsstudien visar att denna tillämpning är genomförbar med de två senare källorna, men man måste ta hänsyn till dessas egenskaper för att kunna utföra mätningen på ett praktiskt sätt.

Figur 1, Reconstruerade bilder av ett bränsleelement med en stav som läcker ut gasformiga fissionsprodukter. Tre olika gaser användes som källor, till vänster: \(^{133}\text{Xe}\); i mitten: \(^{135}\text{Xe}\); till höger: \(^{85}\text{Kr}\). Den förstnämnda befanns vara olämplig på grund av sin oförmåga att nå ut från bränsleelementets centrala delar, medan de två senare bedöms kunna användas för att identifiera den läckande staven.

Simuleringsstudier gjordes också för att undersöka den förväntade prestandan av den gammatomografiska utrustningen som har byggts vid Haldenreaktorn. Simuleringsarna motsvarade mätning av klyvningsprodukten \(^{137}\text{Cs}\) i bränslekutsarna samt den gasformiga produkten \(^{85}\text{Kr}\) i rören i ett Haldenbränslelement. Bilderna som rekonstruerades redovisas i Figur 2, där fördelningen av \(^{137}\text{Cs}\) i kutsarna återfinns till vänster och fördelningen av \(^{85}\text{Kr}\) i
rören återfinns till höger. På ett kvalitativt sätt motsvarar de rekonstruerade bilderna de simulerade fördelningarna.

Figur 2, Rekonstruerade bilder av simulerade mätdata från ett bränsleelement från forskningsreaktorn i Halden. Bilden till vänster motsvarar fördelningen av klyvningsprodukten $^{137}$Cs i bränslekutsarna och bilden till höger motsvarar fördelningen av den gasformiga produkten $^{85}$Kr i rören.

Särskilda rekonstruktioner genomfördes också för att med hög noggrannhet bestämma aktivitetsinnehållet i varje stav. Både den rekonstruerade fördelningen av $^{137}$Cs i bränslekutsarna och den rekonstruerade fördelningen av $^{85}$Kr i rören matchade den simulerade aktivitetsfördelningen med mycket hög noggrannhet.

Experimentella tomografiska mätningar:
Gammatomografiska mätningar genomfördes med det instrument som byggdes vid Haldenreaktorn på ett Halden bränsleelement. Två mätningar gjordes: en mätning på bränslekutsarna och en mätning på rören. Mätdata från fyra gammaenergier som sänds ut från två fissionsprodukter och två aktiveringprodukter rekonstruerades. Figur 3 redovisar de återskapade bilderna av fördelningen av klyvningsprodukten $^{137}$Cs i kutspelarområdet (till vänster) och fördelningen av den gasformiga produkten $^{85}$Kr i rören (till höger). Fördelningen av $^{137}$Cs visar att den har förflyttat sig från kutsarnas centrala delar till dess ytor, vilket är förväntat med hänsyn till de höga temperaturer som detta bränsle hade utsatts för.
Figur 3, Rekonstruerade bilder av fördelningen av klyvningsprodukten $^{137}$Cs i bränslekutsarna (vänster) och fördelningen av den gasformiga produkten $^{85}$Kr i rören (höger). Bilderna har tagits fram utifrån mätdata som samlats in med den tomografska mätutrustning som har byggts inom ramen för detta doktorandprojekt.

Figur 4 redovisar återskapade bilder av fördelningarna av aktiveringsprodukten $^{60}$Co (till vänster) och $^{178m}$Hf (till höger), både uppmätta i rören just ovanför bränslekutsarna. Fördelningen av $^{60}$Co visar de metallfjädrar som finns i detta område i varje stav och fördelningen av $^{178m}$Hf visar de fyra bärande metallstavar som håller samman bränsleelementet.

Sammanfattningsvis har de mätningar som genomförts visat att den tomografska mätutrustning som har byggts vid forskningsreaktorn i Halden i kombination med de tomografska analysmetoder som har tagits fram kan ge ny och värdefull information om det bränsle som bestrålas i denna reaktor, vilket på sikt kan bidra till säkrare och mer effektiv drift av kommersiella kärnkraftverk.
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Editor: The Dean of the Faculty of Science and Technology

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