Nuclear safeguards evaluation and analysis techniques for application to nuclear fuel material in Generation IV nuclear energy systems

MATILDA ÅBERG LINDELL
A new generation of nuclear energy systems called Generation IV is under development to ensure that nuclear power will be a safe, reliable and sustainable energy source for the future. This thesis addresses the challenge of making future nuclear energy systems increasingly resistant to nuclear material diversion attempts.

Several tools have been developed for structured evaluation of a system's resistance to nuclear proliferation, in order to identify areas where nuclear energy systems are the most inherently vulnerable. In this thesis, the TOPS methodology has been applied to three different fuel cycles involving a fast reactor with fuel recycling and fuel fabrication capabilities. The recycling facility, where the fuel is dissolved and undergoes chemical separation, is identified as being particularly vulnerable. Nondestructive measurements for verification of fuel assemblies in the receipt area of the recycling facility are essential, since it is the last opportunity to verify intact fuel items. Moreover, iterative evaluation of proliferation resistance by using two different assessment methodologies – TOPS and PR&PP – as suggested in this thesis, may act as an aid in facility design and for proposing safeguards implementation.

Based on the identified need to measure irradiated fuel assemblies prior to dissolution in the recycling facility, new methods used for analyzing gamma-ray spectroscopy data using multivariate analysis methods have been investigated. Fuel parameters of modeled nuclear fuel have been determined without any reliance on operator-declared data. Nonlinear classifiers, e.g. support vector machines (SVM), have successfully been used for discrimination between uranium oxide fuels and mixed oxide fuels. Cooling time, burnup and initial fissile content have been determined using decision tree and SVM regression. The results are promising and indicate that the nuclear safeguards regime may benefit from using multivariate techniques for data analysis. It must be emphasized, however, that experimental verification of the multivariate analysis techniques is necessary.

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“You know what uranium is, right? It’s this thing called nuclear weapons and other things. Like, lots of things are done with uranium, including some bad things.”

– U.S. President Donald J. Trump [1]
List of papers

This thesis is based on the following papers, which are referred to in the text by their Roman numerals.


*My contribution*: I specified the facility under study, chose and modified the assessment methodology, and performed the analysis. I was the main author of the paper.


*My contribution*: I specified the facility under study, identified safeguards needs, and suggested measurement techniques. I was the main author of the paper.


*My contribution*: I specified the facility under study, chose and modified the combination of assessment methodologies, and provided illustrative examples of analyses. I was the main author of the paper.


*My contribution*: I generated the data, selected the analysis methods, and performed and evaluated the classifications. I was the main author of the paper.
M. Åberg Lindell, P. Andersson, S. Grape, A. Håkansson, M. Thulin

Determination of irradiated nuclear fuel characteristics by nonlinear multivariate regression of simulated gamma-ray emissions.

Submitted to Nuclear Instruments and Methods in Physics Research, A, 2017-12-10

My contribution: I generated the data, selected the analysis methods, and performed and evaluated the regression analyses. I was the main author of the paper.

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1. Introduction

Over the last twelve years, the world’s population has increased by approximately one billion inhabitants, numbering nearly 7.6 billion as of mid-2017. The population increase persists, and the number of inhabitants is projected to reach 8.6 billion by 2030, 9.8 billion in 2050, and 11.2 billion by 2100 [2]. For the Earth to support its population with a sustained or improved quality of life, we must increase the use of energy supplies that are clean, safe, and cost-effective [3].

At the 2015 United Nations Climate Change Conference in Paris, climate scientists suggested that only a combined strategy employing all major sustainable clean energy options – including renewables and nuclear power – would suffice to reduce carbon emissions enough to meet climate goals [4]. Nuclear energy is a mature source of baseload power with low greenhouse gas emissions, that could provide an increased share of the world’s electricity supply, today amounting to 11% [5, 6].

Some hesitations exist about the future of nuclear development, especially in the aftermath of the Fukushima Daiichi nuclear power plant accident [7]. However, several issues of concern, including operational safety, are addressed by emerging fuel cycles and reactor technologies known as Generation IV (Gen IV). Part of the Gen IV systems are fast reactors, that have a very efficient utilization of natural resources compared with conventional light water reactors (LWR) by deriving energy from the current used nuclear fuel. In this way, the need for uranium mining and enrichment, which are the most CO$_2$ intensive steps in the nuclear fuel cycle, can be drastically reduced. Furthermore, their possibility to operate with used fuel can reduce the amount of long-lived radionuclides in the waste products and the volume of waste requiring deep geological disposal [8].

Another concern with nuclear energy is the possibility to produce nuclear weapons from uranium or plutonium, which are elements occurring in the nuclear fuel cycle. It must therefore be assured to the international community, via the international control system known as nuclear safeguards, that all nuclear material in nuclear power programs remains in peaceful use. The nuclear weapons issue is underlined by current events in North Korea which, with its development of nuclear weapons and missile launches, has become a threat to global peace and security [9].

New reactor concepts and nuclear facilities should be at least as resistant to diversion of nuclear material, as were the previous ones. The emerging generation of nuclear power systems will give rise to new challenges to the international safeguards community due to new and increased flows of nuclear
material in the nuclear fuel cycle. Before a wide implementation of Gen IV nuclear power systems takes place, there lies an opportunity to strengthen their inherent proliferation resistance and formulate safeguards requirements. Papers I–III in this thesis address the challenge of making future nuclear energy systems increasingly resistant to nuclear material diversion attempts.

Abnormal conditions found in processes for nuclear fuel management, or parameters of the fuel itself, could be an indication of ongoing efforts to divert nuclear material for non-peaceful purposes. Organizations such as the International Atomic Energy Agency (IAEA) review nuclear facility operation records and other information provided by a state to verify that it acts in compliance with its commitments. Inspections of nuclear facilities and measurements of nuclear materials are also conducted such that safeguards inspectors can independently draw conclusions about the accuracy of the declared information. In addition to verifying operator declared parameters of spent nuclear fuel, the ability to experimentally determine such parameters with a minimum of intrusiveness is of great interest for safeguards purposes. In particular, in a case where declared information about spent nuclear fuel is lost or deficient, such ability is of great importance [10].

The development of new methodologies, equipment and analysis techniques that meet requirements of inspecting authorities and complement their already existing inventory of tools is of high relevance. Analysis of measurement data using multivariate analysis techniques may enhance capabilities to discern safeguards-relevant information from nondestructive measurements on irradiated nuclear fuel, due to their abilities to find and exploit correlations in large amounts of seemingly unstructured data. The aim of papers IV–V in this thesis was to theoretically investigate the feasibility of using nondestructive assay and multivariate analysis methods to, without reliance on operator declarations, determine characteristics of irradiated pressurized water reactor (PWR) fuel, with the ultimate goal to strengthen nuclear safeguards operations in future nuclear energy systems.

The outline of this thesis is the following: In chapter 2, a general overview of the international safeguards regime is provided. Chapter 3 provides some background information on Generation IV systems and nuclear fuel cycle options, including some possible challenges associated with safeguarding future nuclear energy systems. Chapter 4 introduces the concept of proliferation resistance and some of the methodologies used to evaluate it. In chapter 5, focus is shifted from evaluation of safeguards implementations and vulnerability to proliferation, to analysis of measurement data collected in the sensitive recycling part of a Gen IV fuel cycle. In chapter 6, multivariate analysis and some available methods are introduced, and in chapter 7, approaches for applying multivariate analysis to safeguards-relevant data are discussed. Finally, chapter 8 contains conclusions drawn from papers I–V, and an outlook on future research.
2. Nuclear safeguards

To combat the proliferation of nuclear weapons, the application of nuclear safeguards is central. Nuclear safeguards is briefly presented in this chapter, whereas the notion of proliferation resistance is covered in chapter 4.

2.1 Legal framework

A vast majority of the states in the world have pledged not to misuse their nuclear energy programs for nuclear weapons production. The IAEA, which is directly subordinated the Security Council of the United Nations, uses the framework known as nuclear safeguards to verify that a state lives up to its commitments according to international treaties. In 1968, the Non-Proliferation Treaty (NPT) [11] was introduced with the aim to:

1. prevent the spread of nuclear weapons and weapons technology,
2. foster the peaceful uses of nuclear energy, and
3. further the goal of disarmament.

The treaty forbids nuclear-weapon states to transfer nuclear weapons, directly or indirectly, to non-nuclear-weapon states, and to supply help in acquiring them. The latter part in turn undertake not to receive any nuclear explosives nor assistance in the manufacturing of nuclear weapons. Each non-nuclear-weapon state that signs the NPT undertakes also to accept nuclear safeguards under the IAEA safeguards system, with control and supervision of the signatory parties as a result. Nonetheless, the NPT is a voluntary agreement based on every state’s will to meet its obligations. Currently, 191 states adhere to the treaty, making it the most ratified arms control agreement in history. The only non-signatory states of NPT are India, Pakistan, Israel, and South Sudan. The Democratic People’s Republic of Korea was a signatory state of the NPT until 2003, when it announced its withdrawal from the treaty [12].

Additional voluntary agreements that give the IAEA extended access and inspection rights, such as the Additional Protocol, are widely implemented. There are also voluntary agreements for nuclear weapon states, enabling the application of safeguards, although not on a comprehensive scale [13].

The IAEA is not the only existing safeguards organization. Sweden, for instance, also reports to Euratom (under the European Commission) and the Swedish Radiation Safety Authority (under the Swedish government).
2.2 Materials accountancy and inspections

Accountancy of nuclear materials is the foundation for the IAEA’s control of signatory states’ compliance with the NPT agreement. All quantities of nuclear material present in the state’s nuclear fuel cycle must be established and book-kept on a regular basis. The IAEA performs inspections to verify that the records are correct, so that nuclear material is not diverted to weapons production.

A significant quantity (SQ), defined as “the approximate amount of nuclear material for which the possibility of manufacturing a nuclear explosive device cannot be excluded”, is a central concept in IAEA safeguards [13]. The mass of 1 SQ varies depending on the type of material, see table 2.1.

Table 2.1. Significant quantities of the most important weapons-usable nuclear materials [13].

<table>
<thead>
<tr>
<th>Material</th>
<th>Significant Quantity</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu</td>
<td>8 kg</td>
</tr>
<tr>
<td>High-enriched U ((^{235})U (\geq) 20%)</td>
<td>25 kg (^{235})U</td>
</tr>
<tr>
<td>Low-enriched U ((^{235})U &lt; 20%)</td>
<td>75 kg (^{235})U</td>
</tr>
</tbody>
</table>

The IAEA must be able to draw the conclusion that there has been no diversion of 1 SQ or more of nuclear material with a certain frequency that fulfills the IAEA’s timeliness detection goals, see table 2.2, which have been set up depending on the specific expected time spans required for conversion of various nuclear materials to weapons-usable materials. Nuclear material that can be used for the manufacture of weapons without transmutation or further enrichment, e.g. plutonium in fuel assemblies, is called direct use material. It is associated with relatively short timeliness goals, in the order of months. Indirect use material, e.g. low-enriched uranium, must be further processed before weapons-production is possible, and therefore has longer timeliness goals [13].

Table 2.2. Timeliness goals as stated by the IAEA [13].

<table>
<thead>
<tr>
<th>Material</th>
<th>Timeliness goal</th>
</tr>
</thead>
<tbody>
<tr>
<td>Unirradiated direct use material</td>
<td>1 month</td>
</tr>
<tr>
<td>Irradiated direct use material</td>
<td>3 months</td>
</tr>
<tr>
<td>Indirect use material</td>
<td>1 year</td>
</tr>
</tbody>
</table>

Different plutonium isotopes have different properties, whereof the most attractive, fissile, isotopes are \(^{239}\)Pu and \(^{241}\)Pu. High levels of neutron emission and decay heat make even-numbered isotopes such as \(^{238}\)Pu, \(^{240}\)Pu and \(^{242}\)Pu difficult to use for weapons production. It is worth noting that, according to IAEA’s conservative definitions, all plutonium isotopes are considered equal in
terms of significant quantities and timeliness detection goals. Thus, plutonium with isotopic compositions of poor quality does not generally reduce the need for safeguards, even though the material attractiveness may be lower compared to other plutonium material compositions.

Similarly, different processes and technologies used in the nuclear fuel cycle can be more or less sensitive, depending on their inherent susceptibilities to material diversion and technology misuse, and therefore require more or less intense attention from safeguards authorities.

As an aid in the accountancy structure, a nuclear facility may be divided into several different Material Balance Areas (MBAs). Within each MBA, records of the quantities of nuclear material are maintained and updated to account for inventory changes. Changes of the chemical or physical form of nuclear material is also recorded, as are transfers of material into and out of MBAs. The size of an MBA should be related to the accuracy with which the material balance can be established. Flows and inventories of material are measured at Key Measurement Points (KMP), located at least at the inputs, outputs and storages in an MBA [13].

Papers II and III of this thesis discuss, based on the findings in paper I, MBAs and KMPs that can be foreseen in a nuclear fuel recycling facility.

2.3 Techniques and equipment

A wide range of measuring techniques and equipment is available to the safeguards inspectors, to help them verify the completeness and correctness of a state’s accountancy reports. A comprehensive overview of the techniques and equipment is provided in the IAEA document “Safeguards Techniques and Equipment” [14].

The detection of missing items (gross defects) can be reached through counting of items, and measurements of their attributes using non-destructive analysis (NDA) techniques. More than 100 NDA systems are used by the IAEA, out of which the most widely used are based on the detection of radiation such as neutrons or gamma rays [14].

For detection of the absence of fractions of nuclear material from an item (partial defects), the use of weighing of items and NDA measurements such as neutron counting or gamma-ray spectrometry may be involved [14].

Very small material diversions (bias defects), which may be conducted over a protracted time, may be detected via sample taking and subsequent destructive analysis (DA) using techniques with high accuracy [14]. Elemental and isotopic analyses of many kinds are performed at qualified laboratories located in 20 IAEA member states [15].

Furthermore, containment and surveillance (C/S) techniques are applied to complement measurements, by monitoring access to the material and assuring that it follows predetermined routes, thus maintaining continuity of knowledge
of the whereabouts of the nuclear material. Seals and optical surveillance are the most commonly used C/S measures [14].

The techniques used by the IAEA should ideally be cost-efficient as well as non-intrusive to the regular operations of the nuclear facility. Unattended and remote monitoring techniques, where data from safeguards systems is transmitted off-site to the IAEA headquarters, permit reduced inspection efforts. The usage of remote monitoring increases continuously [14].

The ways in which safeguards measures can be combined in order to provide safeguards inspectors with sufficient information is to a certain degree depending on the characteristics of the facility at hand. For example, paper II in this thesis points towards difficulties in safeguarding an envisioned recycling facility and discusses some prerequisites for suitable instrumentation.

2.4 Safeguards by design

Safeguards by design is the process of including the consideration of safeguards implementation throughout all phases of a nuclear facility project, from the initial conceptual design to facility construction, into operations and, finally, decommissioning. Safeguards should be considered early in the design so that it can be better integrated with other design considerations, e.g. operations, safety and security. Cooperation between various stakeholders, such as designers, vendors, operators, and safeguards authorities, can facilitate a more cost effective and efficient implementation of safeguards, while minimizing the impact on nuclear facility operations. In fact, this cooperation is explicitly required under comprehensive safeguards agreements. The ‘by design’ concept does not introduce new requirements, but rather seeks to reduce the impact of existing safeguards requirements on the design and construction cost and schedule, for example by avoiding costly and time consuming retrofits or redesigns of new nuclear facilities to accommodate safeguards [16].
3. Generation IV nuclear energy systems

3.1 Reactor technologies and goals

Generation IV (Gen IV) nuclear energy systems is the denomination of a group of emerging nuclear power technologies. The system aspect of Gen IV is strong and comprises reactors, recycling of used nuclear fuel and final waste storage. There are currently a number of promising reactor concepts under development to address various issues of concern of the current fleet of reactors. An international collaboration known as the Generation IV International Forum (GIF) has selected six proposed nuclear energy systems to represent Gen IV, see table 3.1.

Table 3.1. The six selected Generation IV reactor systems and their main characteristics [3].

<table>
<thead>
<tr>
<th>Generation IV System</th>
<th>Acronym</th>
<th>Coolant</th>
<th>Neutrons</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas-Cooled Fast Reactor System</td>
<td>GFR</td>
<td>Helium</td>
<td>Fast</td>
</tr>
<tr>
<td>Lead-Cooled Fast Reactor System</td>
<td>LFR</td>
<td>Lead</td>
<td>Fast</td>
</tr>
<tr>
<td>Molten Salt Reactor System</td>
<td>MSR</td>
<td>Fluoride salts</td>
<td>Thermal/fast</td>
</tr>
<tr>
<td>Sodium-Cooled Fast Reactor System</td>
<td>SFR</td>
<td>Sodium</td>
<td>Fast</td>
</tr>
<tr>
<td>Supercritical-Water-Cooled Reactor System</td>
<td>SCWR</td>
<td>Water</td>
<td>Thermal/fast</td>
</tr>
<tr>
<td>Very-High-Temperature Reactor System</td>
<td>VHTR</td>
<td>Helium</td>
<td>Thermal</td>
</tr>
</tbody>
</table>

The Gen IV technology is issued with eight common goals, listed here in table 3.2. All goals should be fulfilled in order to ensure that nuclear power will be a safe, reliable and sustainable energy source for the future. The scope of the research presented in this thesis is connected to the proliferation resistance goal, which will be discussed further in chapter 4.

The research presented in this thesis has partly been performed in the framework of the Swedish R&D program GENIUS, funded by the Swedish Research Council, which addresses generic aspects of Gen IV. Papers I–V consider lead-cooled fast reactors and aqueous recycling, in order to align with the goals set by the GENIUS program.

3.2 Fast neutron reactors

Neutrons sustain the chain reaction in a nuclear power reactor by inducing fissions in heavy nuclei and at the same time emitting new neutrons. A fission
### Table 3.2. The eight goals set for Generation IV systems by GIF, with the goal concerning proliferation resistance and physical protection highlighted. From [3].

| Sustainability | 1. Generation IV nuclear energy systems will provide sustainable energy generation that meets clean air objectives and promotes long-term availability of systems and effective fuel utilization for worldwide energy production. |
| 2. Generation IV nuclear energy systems will minimize and manage their nuclear waste and notably reduce the long-term stewardship burden, thereby improving protection for the public health and the environment. |
| Economics  | 1. Generation IV nuclear energy systems will have a clear life-cycle cost advantage over other energy sources. |
| 2. Generation IV nuclear energy systems will have a level of financial risk comparable to other energy projects. |
| Safety and Reliability  | 1. Generation IV nuclear energy systems operations will excel in safety and reliability. |
| 2. Generation IV nuclear energy systems will have a very low likelihood and degree of reactor core damage. |
| 3. Generation IV nuclear energy systems will eliminate the need for offsite emergency response. |
| Proliferation Resistance and Physical Protection | 1. Generation IV nuclear energy systems will increase the assurance that they are a very unattractive and the least desirable route for diversion or theft of weapons-usable materials, and provide increased physical protection against acts of terrorism. |

Event typically releases around 200 MeV of energy, distributed among fission products, neutrons and other particles. Neutrons are born with a mean kinetic energy of 2 MeV per neutron. The probability that a neutron that is absorbed in a nuclide causes a fission can be expressed in terms of cross-sections as:

\[
\frac{\sigma_f}{(\sigma_f + \sigma_\gamma)},
\]

where \(\sigma_f\) denotes the fission cross section and \(\sigma_\gamma\) denotes the radiative capture cross section, in which the incident neutron is completely absorbed in the target.
Table 3.3. Heavy nuclei with half-lives and fission-to-absorption ratios, $\sigma_f / (\sigma_f + \sigma_\gamma)$ for PWRs and fast reactors (FR). Adapted from [18].

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life</th>
<th>PWR</th>
<th>FR</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{235}$U</td>
<td>$7.0 \times 10^8$ y</td>
<td>82%</td>
<td>77%</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>$4.5 \times 10^9$ y</td>
<td>11%</td>
<td>12%</td>
</tr>
<tr>
<td>$^{237}$Np</td>
<td>$2.1 \times 10^6$ y</td>
<td>1.6%</td>
<td>16%</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>$2.4 \times 10^4$ y</td>
<td>63%</td>
<td>77%</td>
</tr>
<tr>
<td>$^{241}$Am</td>
<td>430 y</td>
<td>1.0%</td>
<td>12%</td>
</tr>
<tr>
<td>$^{243}$Am</td>
<td>7400 y</td>
<td>2.3%</td>
<td>10%</td>
</tr>
<tr>
<td>$^{244}$Cm</td>
<td>18 y</td>
<td>5.9%</td>
<td>41%</td>
</tr>
<tr>
<td>$^{245}$Cm</td>
<td>8500 y</td>
<td>87%</td>
<td>85%</td>
</tr>
</tbody>
</table>

nuclide. The new, heavier, nucleus that is formed emits gamma-rays. The cross sections are heavily dependent on the energy of the neutron.

In today’s LWRs, neutrons are moderated, i.e. slowed down to thermal energies (0.025 eV) by the water surrounding the nuclear fuel, because absorbed thermal neutrons have a higher probability than fast neutrons of inducing new fissions in the fissile material $^{235}$U. A fast reactor, on the other hand, relies on fast neutrons to cause fission. They are often fueled with plutonium, and the coolant is often a liquid metal (usually sodium or lead) which does not act as a neutron moderator. Neutrons with high energies cause more neutrons to be released per fission, meaning that, if present, more $^{238}$U nuclei can be converted to $^{239}$Pu via neutron capture [17]. A fast reactor having a conversion ratio (the ratio of produced to consumed fissile nuclei) exceeding 1 is called a breeder reactor, due to its ability to ‘breed’ new fissile material. Furthermore, as shown in Table 3.3, several heavy nuclides have much higher fission-to-absorption ratios in the fast part of the neutron spectrum, which enables ‘burning’ of non-fissile heavy nuclei in fast reactors.

Fast neutron reactors have been under development for decades. Early fast reactors would not, however, qualify as Gen IV concepts, since the systemic perspective was lacking. Since the 1950s, about 20 fast neutron reactors (experimental, prototype, and commercial) have already been operating and, as of 2010, 400 reactor-years of operating experience have been accumulated. The overall operational experience is positive, and great strides have been made in fast reactor technology [19, 20, 21]. Nevertheless, there is less experience with fast reactors than with the LWRs used for most power plants today. In addition, the technology required to build fast reactors and their associated fuel cycle facilities is more complex, which can explain why they were not selected for the first industrial deployment of nuclear power plants. Further developments are
still necessary for fast reactor systems to meet today’s high level requirements on economy, safety and reliability [17].

3.3 Nuclear fuel cycles

Today’s nuclear fuel cycle options govern the fate of used nuclear fuel after it has been discharged from a LWR. 96% of the original uranium remains intact in irradiated fuel, albeit with a lower share of the fissile isotope $^{235}$U than in fresh fuel. Plutonium is built up during reactor operation, and constitutes about 1% of used LWR fuel. The remainder consists of fission products and heavy transuranic nuclides [22], which cannot be reused in fuel for LWRs. In Gen IV systems, however, some of the heavy nuclides may be recycled and ultimately fissioned in fuel for fast reactors, thus resulting in more efficient fuel utilization.

There are three main options for the back-end of a nuclear fuel cycle; the open, closed, and advanced closed fuel cycles.

The open, or once-through, fuel cycle, widely used in nuclear power production today, implies the following: Uranium is mined, enriched to 3–5% in $^{235}$U, and manufactured into fuel assemblies, which are loaded into a reactor. In LWRs, the fuel is irradiated in the reactor for about five years. After that, the fissile content has decreased due to fissioning of uranium and plutonium, and the fuel is treated as waste. However, as mentioned above, there is still a significant amount of extractable energy in terms of heavy nuclei in the discarded fuel assembly.

Some countries have chosen to adopt the conventional closed fuel cycle. This fuel cycle option saves up to 30% of the natural uranium otherwise required for fabrication of new fuel [22], by recovering uranium and plutonium for repeated use in LWRs [23]. Fuel recycling requires reprocessing, i.e. separation of the uranium and plutonium from the fission products. The chemical process commercially used for this purpose is called Purex. In a Purex plant, the nuclear fuel is dissolved in nitric acid and the desired elements are subsequently separated via solvent extraction. For more information on the chemical processing, see [24]. The new fuel, which encompasses the recycled material, is called mixed oxide fuel (MOX). MOX fuel is disposed of after one period of irradiation (about 5 years) since the isotopic quality of the plutonium in a MOX assembly degrades with increasing burnup to contain less fissile isotopes. Furthermore, an increased fraction of $^{240}$Pu affects operating safety margins in thermal reactors, thereby limiting the amount of MOX fuel that can safely be used in a LWR [25].

With fast reactor technologies, a new fuel cycle option emerges; the advanced closed fuel cycle. One of the advantages of fast reactors is their ability to burn all heavy nuclides from used nuclear fuel, including the long-lived so-
Figure 3.1. Some advanced recycling techniques allow for repeated recycling of fuel for fast reactors, with all actinides extracted as a group.

called minor actinides\(^1\): americium, curium and neptunium. Thus, by launching fast reactor power production, one may adopt a closed fuel cycle, where actinides are separated and reused multiple times, see figure 3.1. This option requires less uranium mining and gives rise to smaller amounts of long-lived waste than the classical closed fuel cycle. Highly radioactive waste will still be produced, mainly consisting of relatively short-lived fission products. These must be put in a final repository, but for a shorter period of time than waste from the other fuel cycle options; approximately 1000 years instead of on the order of 100,000 years. In addition, the required repository space can be minimized. The resulting waste will also be less radiotoxic after the removal of actinides and it cannot be used for nuclear weapons production.

A multitude of advanced recycling techniques are under development, one of which is called Ganex\(^2\) (Group actinide extraction) [26]. In the Ganex process, uranium, plutonium and minor actinides are extracted together to become fuel in Gen IV fast neutron reactors. Since there is no intermediate step containing pure plutonium, which would be weapons useable, and since the radiation hazard associated with minor actinides may deter a diverter, Ganex has

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\(^1\)Actinides are radioactive chemical elements with atomic numbers from 89 to 103, including uranium and plutonium.

\(^2\)More precisely, Ganex is a group of similar techniques, with some variations in the involved molecules.
the potential to enhance proliferation resistance [27]. The Ganex process is a long-term goal which has, so far, proven to be efficient at the laboratory scale on kilograms of spent nuclear fuel, but is not yet mature enough for industrialization [28, 29].

In addition to the three fuel cycles mentioned above, there is also a possibility to combine various reactors in so-called symbiotic fuel cycles [7]. Such fuel cycles can be obtained e.g. through combinations of thermal reactors and fast reactors to accommodate transition periods, or through combinations of breeding and burning fast reactors.

In the papers presented in this thesis, different options for closed fuel cycles have been examined: Paper I discusses three different recycling options; Purex (for uranium and plutonium recycling only), Ganex (where all actinides are extracted as a group), and combination of Purex, Diamex and Sanex (Purex with subsequent separation of minor actinides and lanthanides).

Paper II describes two facility operation phases, where different recycling techniques would be implemented. It is assumed that one may need to use Purex initially, to obtain enough plutonium for starting a new fast reactor. Thereafter the reactor would be self-sufficient in plutonium production, and Ganex may instead be used. In Paper III, Purex is the assumed recycling process. Paper I accounts for a system with a fast reactor, recycling and fuel fabrication facilities, whereas Papers II and III only deal with the recycling part of the fuel cycle.

### 3.4 Safeguards in Generation IV systems

As an increasing number of countries launch nuclear power programs, larger quantities of nuclear material will be present in the system over all. Furthermore, the knowledge and experience of nuclear technology will spread. Thirty countries are currently using nuclear power, and about the same number are considering, planning or actively working to supplement their energy mix with nuclear power. Out of the 30 countries using nuclear power, 13 are currently constructing new power plants [30]. Accordingly, an increased need for safeguards inspections can be expected, in order to account for the possible expansion in nuclear power. Assuming the available safeguards resources to be fairly constant, the safeguards regime may also need to become more efficient and cost-effective in order to cope with the increased work load, e.g. by extending the use of short-notice random inspections and unattended monitoring.

Implementation of new groundbreaking Gen IV technologies in the nuclear power systems will inevitably require reassessment of the existing safeguards.

---

3Lanthanides are chemical elements with atomic numbers 57–71, often having large thermal neutron absorption cross sections. Lanthanides and actinides have similar chemical properties, which makes separation difficult.
requirements. Some of the safeguards-related benefits and challenges facing the emerging nuclear power systems are discussed below.

With a comprehensive employment of Gen IV and closed nuclear fuel cycles, transport and handling of nuclear material will increase, as will the need for recycling, which is considered to be a sensitive technology in nuclear safeguards. This is partly mitigated by the reduced or eliminated need for another sensitive technology: uranium enrichment.

Facilities for recycling and fuel fabrication are so-called *bulk handling facilities*. They are in general more demanding to safeguard than item handling facilities, where ID numbering of the material may be applied. Also some of the Gen IV reactor concepts with unusual fuel materials (pebbles and liquid salt) are bulk handling facilities, and operation of such a reactor demands a re-evaluation of safeguards measures in the reactor facility.

As a part of the start-up phase of fast reactors, large amounts of plutonium may likely be used initially for fueling the new reactors. This poses new challenges to the international safeguards regime. On the other hand, disarmament will be supported by the extended recycling capabilities, since plutonium from the existing stockpile of nuclear weapons can be used to manufacture new fuel for power-producing reactors [23].

Isotopic compositions and types of Gen IV fast reactor fuel generally differ from the traditionally used oxide fuels for LWRs. The same measuring equipment may not necessarily be applicable to the new types of fuel, higher burn-up levels etc. It must therefore be ensured that there is safeguards equipment suited for measurements of all new fuel types, at all fuel cycle stages. Safeguarding Gen IV related activities may accordingly require extended efforts in the development and evaluation of safeguards instrumentation.

Careful selection of the fuel composition and recycling techniques may further increase the proliferation resistance of the Gen IV nuclear fuel cycle. Making nuclear material less suitable for use in a nuclear weapon, or less prone to diversion for such use, can be achieved by:

- increasing the radiological intensity of the material itself, so that it cannot be handled without severely exposing the people handling it unless they use heavy and specialized shielding equipment, and
- ensuring that the isotopic composition of the fuel will not at any point in the fuel cycle be suitable for the production of an explosive nuclear device, without prior complex processing of the material.

Most of the Gen IV systems involve fast reactors relying on multiple reprocessing and recycling of fuel, which essentially address the above strategies [31].
4. Evaluation of proliferation resistance

A state in possession of nuclear facilities may be able to divert nuclear material such as uranium or plutonium, or misuse a facility such that nuclear materials are produced, for subsequent weapons production. However, proliferation attempts can be obstructed if the nuclear facilities are designed to inherently withstand them. According to the IAEA, proliferation resistance is defined as “that characteristic of a nuclear energy system that impedes the diversion or undeclared production of nuclear material, or misuse of technology, by States in order to acquire nuclear weapons or other nuclear explosive devices” [32]. Security threats posed by non-host-state actors, e.g. terrorist groups, are not included in this definition, and the means applied to impede such threats are commonly denoted as ‘physical protection’. While one should not be led to believe that there is such a thing as a “proliferation proof” facility, it is however possible to make diversion more difficult by increasing the time and cost needed for a state to complete a diversion attempt. For Gen IV systems to comply with the proliferation resistance goal set by the Generation IV International Forum, i.e. “Generation IV nuclear energy systems will increase the assurance that they are a very unattractive and the least desirable route for diversion or theft of weapons usable materials”, proliferation resistance needs to be addressed when designing the new systems.

As discussed in chapter 2.4, the dedicated design of safeguards for new facilities, starting from the early design stages, will help ensure that the subsequent implementation of safeguards in the facility will be as smooth, cost efficient and non-intrusive as possible. Discussions on proliferation resistance should take place between operators, safeguards authorities and other stakeholders, during all stages of planning a new facility design and throughout the facility’s lifetime. Proliferation resistance assessments could be used as a base for such discussions, and as a guide for refining an unfinished facility design. Authorities performing licensing procedures may as well benefit from the support of a proliferation resistance evaluation.

4.1 Assessment methodologies

To determine whether GIF’s proliferation resistance goal is fulfilled or not, and to compare the viability of different fuel cycle options, one may assess proliferation resistances of different nuclear energy systems. There are numerous different methodologies developed for this purpose. Proliferation resistance
assessments of nuclear energy systems provide a structured approach for assessing the value of both intrinsic and extrinsic barriers to proliferation, and a basis for improving and enhancing safeguards.

Proliferation resistance assessments should be carried out following a standardized approach that has international acceptance and that provides consistent results independent of the analysts carrying out the evaluation. The assessments should preferably be performed by a group of experts, but can also be performed and motivated by individual assessors. Biased judgment in the group of analysts is a possible weakness in the proliferation resistance assessments, and any evaluation that will be used to support decision-making should be peer reviewed to ensure its quality [33]. Rigorous uncertainty and sensitivity analyses are rarely executed, and it may even be difficult to do so in a meaningful way, depending on the chosen assessment methodology and the system under study. Assessment methodologies may combine various quantitative metrics to provide a single overall nuclear security measure, which can be used to compare different systems [34]. Regardless of the methodology being used, the valuable information about the proliferation resistance of a nuclear energy system should be easily displayed with different levels of detail, and must ultimately be made usable and understandable to decision makers [35, 33].

There are multiple tools available for evaluating proliferation resistance and diversion risks of a nuclear installation. Which one of them is the best to use may depend on e.g. which stakeholder performs the analysis and what the results will be used for. The two established methodologies used in papers I–III, TOPS and PR&PP, are briefly described below. TOPS may lend itself best to quick assessments and comparisons of facility alternatives, whereas PR&PP is a more extensive and demanding methodology. A combination of the two, as presented in Paper III, is an attempt to reap the benefits of both methodologies. No matter which methodology or combination of methodologies that is chosen, structured proliferation resistance assessments should be made before constructing a new nuclear facility, to compare different material flows, operation processes etc., and evaluate the resulting potential resistances to diversion.

**TOPS**

A task force established by the U.S. Department of Energy in 1999 developed the so-called TOPS methodology (Technological Opportunities To Increase The Proliferation Resistance Of Global Civilian Nuclear Power Systems) [36]. It has become the basis for other evaluation methods [37].

They identified a set of barriers impeding proliferation. An overview of the barriers, divided into three barrier categories, is provided in figure 4.1. Material and technical barriers are intrinsic, meaning that they concern inherent properties of a fuel cycle, its facilities and equipment. Material barriers represent inherent material qualities that describe to which extent the material is attractive to a proliferator, whereas the technical barriers relate to the intrinsic
technical features of facilities, equipment and processes, which obstruct prolif-erators’ access to materials and facilities. Institutional, or extrinsic, barriers, including safeguards and access control, form compensation for weaknesses in the former categories. This information is not required for an assessment of the inherent proliferation resistance of a general facility design, as is the case in paper I, and they are therefore omitted in the paper.

Figures 4.1. Graphical overview of the TOPS methodology illustrating how the total resistance (left) is estimated based on the barrier categories (center) and barriers (right).
Effectiveness of the barriers can be assessed for different stages of the examined nuclear facility or nuclear fuel cycle, in this thesis denoted ‘segments’. Higher, more effective, barriers require greater efforts to be overcome than lower, less effective, barriers. Classification of the effectiveness of the barriers can be semantic, ranging e.g. from ‘insignificant’ to ‘very high’, or numeric, with values ranging from e.g. 1 to 5. Barriers are not absolute, and do not always act independently, and the semantic alternative for classification may have an advantage in reflecting this. Numerical values may however be better suited for aggregation of values and comparison of results, as long as it is stressed by the analysts that the barrier strengths should not be seen as absolute values.

Various scenarios or proliferation threats can be evaluated using the TOPS methodology. A threat can e.g. include overt or covert diversion posed either by a State, or by a subnational group. For comprehensive comparison between the different nuclear facilities or nuclear fuel cycles, several threat contexts should be chosen for the TOPS analysis.

The TOPS methodology is described in more detail in papers I and III, together with examples of performed analyses showing its application in practice.

PR&PP
The PR&PP methodology (Proliferation Resistance and Physical Protection) is a methodology developed for Gen IV systems by the Generation IV International Forum. It is based, much like TOPS, on the definition of a specific proliferation threat. The analysts assess the response of the examined facility or nuclear fuel cycle to the evaluated threat, by identifying potential targets such as materials that can be diverted and processes that can be misused, and all possible diversion paths, i.e. sequences of events leading to weapons production. After systematically defining targets and diversion paths, measures are estimated for each one of the different pathways, using qualitative or quantitative descriptors (‘Very low’–‘Very high’ resistance, or 1–5). The proliferation resistance measures are listed in table 4.1. In the example analyses in paper III, costs were not estimated, meaning that the ‘proliferation cost’ and ‘detection resource efficiency’ measures were omitted.

The level of detail of a PR&PP analysis may vary depending on the level of progress of the system design, and continuous assessments during the design phase of a new facility is encouraged. More information on the methodology can be found in ref. [33] as well as in Paper III of this thesis.

4.2 Practical application of assessment methodologies
In Paper I, the TOPS methodology was chosen for the proliferation resistance assessment because it is one of the most well-established existing methodolo-
Table 4.1. The proliferation measures used in the PR&PP methodology. From [33].

<table>
<thead>
<tr>
<th>Proliferation Measure</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Technical Difficulty</td>
<td>The inherent difficulty, arising from the need for technical sophistication and materials handling capabilities, required to overcome the multiple barriers to proliferation.</td>
</tr>
<tr>
<td>Cost</td>
<td>The economic and staffing investment required to overcome the multiple technical barriers to proliferation, including the use of existing or new facilities.</td>
</tr>
<tr>
<td>Time</td>
<td>The minimum time required to overcome the multiple barriers to proliferation (i.e., the total time planned by the Host State for the project).</td>
</tr>
<tr>
<td>Fissile Material Type</td>
<td>A categorization of material based on the degree to which its characteristics affect its utility for use in nuclear explosives.</td>
</tr>
<tr>
<td>Detection Probability</td>
<td>The cumulative probability of detecting a proliferation segment or pathway.</td>
</tr>
<tr>
<td>Resource Efficiency</td>
<td>The efficiency in the use of staffing, equipment, and funding to apply international safeguards to the nuclear energy system.</td>
</tr>
</tbody>
</table>

The hypothetical facility examined in this work is a 100 MWe lead-cooled fast reactor, with associated reprocessing and fuel fabrication capabilities included on the reactor site in a separate building. Three different fuel recycling options, based on different reprocessing techniques, were considered:

- Purex: for uranium and plutonium recycling only,
- Purex together with Diamex and Sanex: Purex with subsequent separation of minor actinides and lanthanides which enables actinide recycling,
- Ganex: all actinides are extracted as a group.

Each of the three facilities was divided into segments, roughly representing the different forms of nuclear material occurring in each examined fuel cycle stage, see table 4.2. The fuel used in the reactor is either MOX or MOX with minor actinides incorporated in the fuel, depending on the choice of reprocessing technique.

The analysis in the paper covers proliferation threats where a technically advanced state performs covert diversion, of either abrupt or protracted kind. In order to be compatible with the threat investigated and the assumed facility design information, some of the barriers defined by the methodology were not
Table 4.2. Division of the hypothetical facility into segments, which represent different forms of nuclear material.

<table>
<thead>
<tr>
<th>Reactor</th>
<th>Reprocessing</th>
<th>Fuel fabrication</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fresh fuel kept in storage</td>
<td>Used fuel in storage</td>
<td>Dissolved material from the reprocessing step</td>
</tr>
<tr>
<td>Fuel inside the core</td>
<td>Used fuel dissolved in nitric acid</td>
<td>Pellets</td>
</tr>
<tr>
<td>Used fuel in storage</td>
<td>Different streams containing the separated materials</td>
<td>Fresh fuel assemblies</td>
</tr>
</tbody>
</table>

Table 4.3. Scale of measurement in pairwise comparison. Adapted from [38]. If barrier $i$ has a number assigned to it compared with barrier $j$, then $j$ has the reciprocal value when compared with $i$.

<table>
<thead>
<tr>
<th>Intensity of importance</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Equal importance</td>
</tr>
<tr>
<td>3</td>
<td>Moderate importance of one over another</td>
</tr>
<tr>
<td>5</td>
<td>Essential or strong importance of one over another</td>
</tr>
<tr>
<td>7</td>
<td>Very strong importance of one over another</td>
</tr>
<tr>
<td>9</td>
<td>Extreme importance of one over another</td>
</tr>
<tr>
<td>2, 4, 6, 8</td>
<td>Intermediate values</td>
</tr>
</tbody>
</table>

The material barriers and technical barriers were not considered equally significant to the proliferation resistance of the fuel cycle, and they were therefore weighted in proportion to their estimated importances using the Analytic Hierarchy Process (AHP) [38]. In short, the barriers within each category were compared pairwise regarding significance to the proliferation resistance. Each pair of barriers was assigned a value, according to table 4.3.

The barrier strengths were thereafter assessed for each facility segment, representing different forms of nuclear fuel, such as irradiated fuel assemblies and fresh fuel pellets. The metrics used were numerical values ranging from 1 (representing ‘Very low resistance’), to 5 (‘Very high resistance’). The results were aggregated such that the most vulnerable segment of each facility (reactor, reprocessing facility and fuel fabrication facility), considering material and technical barriers separately, was singled out to represent the weakest link of that facility. Estimated resistance values for the weakest links in the reprocessing facility can be seen in figure 4.2.
One of the conclusions drawn in the study is that group actinide extraction is preferred to classical reprocessing due to less attractive material compositions in the system. Results from paper I more specifically show that, in the reactor, fresh fuel is more vulnerable than used fuel regardless of the chosen fuel cycle, due to its chemical and radiological properties and easier access. Material wise, Purex offers the lowest proliferation resistance, whereas from a technical perspective all three fuel cycles are equally resistant. In the reprocessing plant, the pure plutonium streams are the weakest links of the Purex and Purex-Diamex/Sanex reprocessing, regarding both the material and technical barrier categories, since the isotopic and chemical barriers of pure plutonium offer low proliferation resistance. In the Ganex case, the pure uranium stream and used fuel with minor actinides included in it, are instead the most vulnerable segments regarding material and technical barriers, respectively. Regarding technical barriers, used fuel elements are more vulnerable than all dissolved and separated materials, since they are considered to be more accessible. From both the material and technical perspectives, Ganex offers the highest proliferation resistance. In the fuel factory, powders and fuel pellets are more vulnerable than fuel elements, since they are easier to conceal and transport, and more difficult to account for. The Purex fuel cycle is the most vulnerable material wise. The lowest technical resistance values in the fuel fabrication segment collection are the same, regardless of the chosen reprocessing technique.
Differences between the fuel cycle options discussed in paper I, regarding weakest links of the TOPS technical barrier group are, in this study, seen only in the reprocessing plant. In order to increase the technical proliferation resistance values also in the reactor and fuel fabrication facilities of Gen IV systems, the facilities should be made difficult to modify and access, and safeguards equipment should be developed to improve precision and reliability. In this way, not only the new material compositions will improve the proliferation resistance of future nuclear power systems, but also new technical solutions will contribute.

Efforts to make Ganex and other group actinide separation techniques commercial are important, considering the expected increase in recycling activities that follow along an implementation of Gen IV nuclear energy systems. In order to cope with this change, it is important to keep developing safeguards instrumentation and methods, to fit the new types of materials and the increased material flows that may arise. Appropriate safeguards instrumentation should be in place and readily available for the implementation of Gen IV systems. An identification of the needs for safeguards measurements in a facility, as performed to some extent in paper II, is a good starting point for the development of such instrumentation.

Paper III proposes a combination of the TOPS and PR&PP methodologies for iterative assessment of proliferation resistance during the design phase of a recycling facility and throughout its lifetime, as depicted in figure 4.3. Here, a facility design is evaluated using TOPS, which may, as in paper I, point to the parts of the facility most vulnerable to diversion. The obtained results may be used by the analysts as an indication of the areas where efforts to identify diversion pathways are the most crucial. TOPS also appears straightforward to use even when the facility design is primitive. Next, a PR&PP analysis gives proliferation resistance estimates of several identified diversion pathways. PR&PP was chosen to be part of the iterative procedure in paper III because it is extensive, well established and believed to complement TOPS well with its slightly different approach.

The outcome of the two analyses may lead to alterations, refinements and variations of the facility design, which will then become subject to a new round of analyses. Uncertainties in the analyses will decrease as the number of iterations increases and the facility design and safeguards approach become well defined to a greater extent. In paper III, a partial analysis was performed, where a limited number of diversion paths were identified and studied. This should be seen as an exemplification of what the first iteration step of an analysis might look like when performed using the combined methodology.

It should be noted that the proliferation resistance estimates obtained in this paper are based on one sole analyst’s judgment, and merely presented for illustrative purposes. In an actual design process, an evaluation group should be involved in the analyses. A guide to preparing and performing a PR&PP analysis with a group of experts is provided in [33].
Figure 4.3. The facility design process suggested in paper III, including the two proliferation resistance assessment methodologies and their outcomes. There is no given ending to the iterative process, which reflects the fact that assessments may be performed throughout the entire lifetime of the facility.
5. Considerations on nondestructive measurements on irradiated fuel assemblies in a recycling facility

The recycling of nuclear fuel involves dissolution of nuclear fuel assemblies, meaning that the fuels can no longer be counted as separate items. In today’s commercial reprocessing facilities, one single batch of dissolved fuel usually contains several assemblies. As a consequence, when the contents of the fuels are mixed, the identities of the separate fuel assemblies are erased and can no longer be recovered. Thus, in order to verify the individual fuel identities, and that the operators’ records are correct, the fuel needs to undergo measurements prior to the dissolution process, i.e. in a fuel reception or storage area. Verification of the conceptually simple ‘in-equals-out’ material balance is dependent on measurements of all material streams going into and out of the facility. The outputs of a recycling facility that are in bulk form can be sampled and measured with destructive methods, giving rather accurate estimates of the fissile content. To minimize the material unaccounted for, it is optimal if the estimated uncertainties are as small in the reception area, as in the output streams. Therefore, it is advisable to accurately measure the incoming fuel assemblies. Well defined fuels also benefit safety, since risks of criticality due to underestimated fissile content, can be decreased. Furthermore, more detailed information may improve the operators’ control over facility processes.

In paper II of this thesis, it is suggested that neutron and gamma based NDA measurements should be carried out on irradiated fuel assemblies in the head-end area, and that efforts should be made to improve the related measurement accuracies. Also advanced measurement techniques such as gamma tomography could be employed to detect partial defects at this stage. The last opportunity to carefully measure the fuel as an item should not be missed. For this reason, it was decided that part of the work in this thesis should focus on nondestructive measurements on irradiated fuel assemblies, performed at a proposed KMP in a recycling facility’s head-end storage area.

Measurement instruments used by safeguards inspectors are continuously developed and refined. Although some measurement concepts, such as high-resolution gamma-ray spectroscopy, provide ample amounts of data from active nuclides in the fuel, the analysis of this data has been performed by using rather simple, albeit effective, methods. However, there is a potential to extract more information out of a measurement, by using data relating to fission
products that would traditionally be overlooked. If the full potential of a measurement is to be realized, new ways of analyzing the data need to be introduced. Multivariate analysis techniques are strong candidates in this context, due to their abilities to find and exploit correlations in large amounts of seemingly unstructured data. Papers IV and V both discuss the feasibility of using multivariate analysis methods to analyze NDA measurement data acquired at the reception area of a recycling facility. The topic of multivariate analysis of measurement data for safeguards purposes is explored further in chapters 6 and 7.

Although this work relates to closed Gen IV fuel cycles, proposed measurements and analyses can be applied to irradiated fuel assemblies in today’s LWR based open fuel cycle.

5.1 Radiation measurements as a means of fuel verification

Due to the strong neutron and gamma radiation emanating from irradiated fuel assemblies, nondestructive measurements of radiation are well suited for verification of used fuel, and have been used extensively for safeguards purposes during many years. The gamma-rays from fission products and the neutrons from built-up heavy actinides drown the signals from uranium and plutonium isotopes in the fuel, and the nuclear materials of main interest for safeguards authorities, uranium and plutonium, cannot be measured directly by using non-destructive methods. Instead, the basis for fuel characterization by neutron and gamma-ray measurements is the correlations between amounts of isotopes present in the fuel, and the sought-after fuel parameters. The buildup and decay of each isotope is governed by specific physical properties such as half-life, neutron capture cross section and fission yield, in a way that provides information about the irradiation history and characteristics of the fuel. For instance, the production of the strong gamma-ray emitter $^{137}$Cs is linear with respect to the burnup, whereas production of $^{134}$Cs and $^{154}$Eu can be roughly approximated as quadratic functions of the burnup [39]. Each gamma-emitting isotope has its own individual set of gamma-ray energies, that acts as a signature of the isotope’s presence in the fuel. Table 5.1 contains half-lives and some prominent gamma-ray energies for nine measurable gamma-ray emitting fission products.

5.1.1 Analysis of isotope ratios for determination of cooling time and burnup

The methods used for analyzing the acquired measurement data, which enables drawing conclusions about the nuclear material content in the fuel, typically
Table 5.1. Half-lives and gamma-ray energies of nine radioactive fission products.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life</th>
<th>Prominent gamma energy [keV]</th>
<th>Branching ratio [%]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{141}\text{Ce}$</td>
<td>32.5 d</td>
<td>145</td>
<td>48.4</td>
</tr>
<tr>
<td>$^{95}\text{Nb}$</td>
<td>35.0 d</td>
<td>765</td>
<td>99.8</td>
</tr>
<tr>
<td>$^{91}\text{Y}$</td>
<td>58.5 d</td>
<td>1205</td>
<td>0.26</td>
</tr>
<tr>
<td>$^{95}\text{Zr}$</td>
<td>64.0 d</td>
<td>724, 757</td>
<td>44.3, 54.4</td>
</tr>
<tr>
<td>$^{144}\text{Ce}$</td>
<td>285 d</td>
<td>2186</td>
<td>0.69</td>
</tr>
<tr>
<td>$^{106}\text{Ru}$</td>
<td>372 d</td>
<td>621, 1050, 1562, 1766, 1797, 1988, 2112</td>
<td>9.93, 1.56, 0.163, 0.034, 0.028, 0.0026, 0.035</td>
</tr>
<tr>
<td>$^{134}\text{Cs}$</td>
<td>2.07 y</td>
<td>569, 605, 796, 802, 1167, 1365</td>
<td>15.37, 97.62, 85.46, 8.69, 1.793, 0.028, 0.0026, 0.035</td>
</tr>
<tr>
<td>$^{154}\text{Eu}$</td>
<td>8.60 y</td>
<td>723, 756, 873, 996, 1004, 1246, 1274, 1494, 1596</td>
<td>20.06, 4.52, 12.08, 10.48, 18.01, 34.8, 0.698, 1.797</td>
</tr>
<tr>
<td>$^{137}\text{Cs}$</td>
<td>30.1 y</td>
<td>662</td>
<td>85.10</td>
</tr>
</tbody>
</table>

*a* Through the decay of the short-lived daughter $^{144}\text{Pr}$

*b* Through the decay of the short-lived daughter $^{106}\text{Rh}$

relies on comparison of concentrations of two isotopes at a time. The isotopic ratios can be determined from gamma-ray activity ratios, usually $^{134}\text{Cs}/^{137}\text{Cs}$ and $^{154}\text{Eu}/^{137}\text{Cs}$ [40]. One may foresee cases in the future when information on a fuel assembly is partly or entirely missing. As described in [39], high-resolution gamma spectroscopy data can be used to experimentally obtain the burnup, $\beta$, and cooling time, CT, of a fuel assembly, even if no declared data is available, see equation 5.1.

$$i_x \cdot e^{\lambda_x CT} = C_x \beta \kappa_x \tag{5.1}$$

Here, $i_x$ represents the measured intensity of the isotope $x$, $\lambda_x$ is the decay constant of $x$, and $e^{\lambda_x CT}$ represents the correction factor for cooling time of the measured isotope. $\kappa_x$ represents the burnup dependence of isotope $x$, which is set to 1 when using $^{137}\text{Cs}$, and set to 2 for $^{134}\text{Cs}$ and $^{154}\text{Eu}$, due to their previously mentioned linear and quadratic dependencies on burnup. $C_x$ is a calibration constant.

From equation 5.1, expressions for cooling time and burnup can be derived (equations 5.2 and 5.3, respectively), based on a combination of two isotopes [39]. In equations 5.2 and 5.3, $^{137}\text{Cs}$ and $^{134}\text{Cs}$ have been used as examples.

$$CT = \frac{1}{\lambda_{134} - 2\lambda_{137}} \ln \left( \left( \frac{i_{137}}{C_{137}} \right)^2 \cdot \frac{C_{134}}{i_{134}} \right) \tag{5.2}$$

$$\beta = \left( \frac{i_{134}}{C_{134}} \cdot \frac{C_{137}}{i_{137}} \right)^{\frac{\lambda_{134}}{\lambda_{137}}} \exp \left( \frac{\lambda_{137} - \lambda_{134}}{2\lambda_{137} - \lambda_{134}} \right) \tag{5.3}$$
In a case where declared information about used nuclear fuel is lost or deficient, the ability to determine parameters of irradiated nuclear fuel is of great importance. While some concepts such as high-resolution gamma-ray spectroscopy provide data that may be suited for this purpose, the analysis of this data has thus far been performed by using rather basic methods. In paper V, it is suggested that the strong and complex correlations between isotope concentrations and various fuel parameters may call for an advanced analysis methodology in order to extract the potentially large amounts of information from the measurements. A methodology for determining burnup, cooling time and initial fissile content of PWR fuels, using passive gamma-ray spectroscopy and multivariate analysis methods is presented in the paper. The findings from paper V are discussed in chapter 7 of this thesis.

5.1.2 Analysis of isotope ratios for UOX–MOX discrimination

Used MOX fuel differs from used uranium oxide (UOX) fuel at the same burnup level, by generally having a larger fissile inventory, a slower decrease of decay heat generation, and an increased content of minor actinides. In a recycling facility, the characterization of fuel as MOX or UOX is essential to ensure that criticality safety requirements are met [41]. From a nuclear safeguards perspective, used MOX fuel contains degraded plutonium that has slightly less value to a proliferator than the plutonium from discharged UOX fuel. However, its attractiveness is still considered to be high [42].

Different fissile isotopes have their own characteristic fission product yields which affect the fuel composition and, in turn, the gamma emission signature of the fuel. Here, fission yields for the fissile isotopes $^{235}$U, which is the only fissile isotope in fresh UOX fuel, and $^{239}$Pu, which dominates in fresh MOX fuel, are of interest. The production of $^{134}$Cs is similar for both isotopes, whereas $^{154}$Eu is more abundantly produced in fissions of $^{239}$Pu than of $^{235}$U. The ratio $^{134}$Cs/$^{154}$Eu can therefore be used to determine whether a fuel assembly is of UOX or MOX type. As shown in figure 5.1, the ratio is typically around twice as high for UOX fuel as for MOX fuel [39].

A different approach for discriminating between UOX and MOX fuel assemblies is to measure the intensity of emitted neutrons. Used MOX fuel differs from used UOX fuel at the same burnup level, by generally having higher levels of neutron radiation. Neutron counts under a certain level indicate that the fuel is of UOX type, whereas neutron counts above a different, higher, level are indicators of MOX fuel. For intermediate neutron emission values, one cannot be certain whether the assembly is a MOX assembly after one cycle of irradiation or a UOX assembly after at least three cycles. In such cases, gamma spectroscopy can be used as a complement [43].

1The term ‘initial fissile content’ will be used throughout this thesis, as opposed to ‘enrichment’, since the plutonium in MOX fuel does not undergo an enrichment process.
Methodologies able to distinguish MOX fuel from UOX fuel are necessary for handling irradiated nuclear fuel. In particular, it can be anticipated that very high standards will be requisite in recycling facilities of a Gen IV system [44]. With the ultimate goal to further strengthen the safety and safeguards aspects in a recycling facility, a discrimination methodology for classification of UOX and MOX fuel, based on passive gamma-ray spectroscopy data and multivariate analysis methods, is presented in paper IV and in chapter 7 of this thesis.

5.2 The measurement situation
Although the scope of papers IV and V was to theoretically investigate the feasibility of using multivariate analysis on high-resolution gamma-ray measurement data, it may be relevant here to somewhat mention the practical implementation of the methodology. It is by no means a trivial task to measure low-energy gammas and high-energy gammas simultaneously for a fuel assembly with short cooling time. The three main reasons for this are:

1. A large detector volume is required to be able to record high-energy gammas. This implies that the counting rate easily saturates the detector due to a high efficiency for low-energy gammas.

\(^2\)The figure label ‘LEU’ denotes low-enriched uranium, which is the same thing as UOX.
2. Intense radiation levels may cause the dead-time of the detector system to reach unacceptable levels.

3. The large Compton background from high-energy gammas will to various degrees obscure low-energy events. To remedy these obstacles, a number of measures need to be employed, which imply reduction of Compton events and counting rate. The number of Compton events can be reduced by using a large detector and using active shielding of the detector. A conceivable solution to handle the situation could be to combine a large detector, taking care of the high-energy part of the spectrum, with a small planar one, covering the low-energy part. With a detector system designed specifically to mitigate these challenges, it would likely be possible to achieve measurement data of sufficient quality for multivariate analysis. In such a detector system, however, other practical features (e.g. portability and ease of use) would most likely be compromised.

One feasible detector setup, albeit not optimized for the measurements required for the multivariate analysis in this thesis, involves easily accessible and geometrically well-defined, stationary equipment for underwater gamma-spectroscopy on site. Built in installations, such as the one depicted in figure 5.2, exist in Swedish BWR reactor facilities as well as in the interim storage, CLAB. Here, a fuel assembly is placed in a movable fixture which is mounted on the pool wall. The fuel can be moved vertically, as well as being rotated around its own axis. On the opposite side of the pool wall, a high-purity germanium detector and a data acquisition system are located. A collimator is built into the pool wall, letting emitted radiation enter the detector in a well defined direction. The height of the collimator slit can be varied as to adjust the counting rate in the detector [39].

A stationary experimental setup facilitates measuring of relative rather than absolute activities, as proposed in papers IV and V. Assuming that identical detector systems and measurement geometries are used for all measurements, a self-calibration procedure could be envisioned. Measured relative intensities of multiple peaks from e.g. $^{154}$Eu can be compared with their respective tabulated emission probabilities, and the energy-dependent efficiency of the detector system can thus be obtained using only the original gamma-ray spectrum [40].
Figure 5.2. Mechanical installation used for performing gamma-spectroscopy measurements on used nuclear fuel in a storage pool. The figure shows the pool and pool wall from the side. The fuel assembly can be moved up and down as well as rotated around its vertical axis [39].
6. Multivariate analysis – concepts and methods

Multivariate analysis encompasses various statistical techniques for analyzing data that has a complex dependence on more than one variable. Via exploration of correlations between the variables, the analysis methods are able to extract information, even from massive amounts of convoluted data, which allows for an improved interpretation and understanding of the data. Among the many fields in which MVA techniques are routinely used, chemometric process control, genomics, and social sciences, can be mentioned. There are also several recent studies pertaining to nuclear fuel characterization.

Abnormal conditions found in processes for nuclear fuel management, or parameters of the fuel itself, could be an indication of ongoing efforts to divert nuclear material for non-peaceful purposes. Thus, applying multivariate analysis techniques, for detection and identification of off-normal traits in process streams as well as in nuclear fuel compositions, may strengthen nuclear safeguards operations. Multivariate methods have previously been used to analyze entire gamma spectra obtained from simulated measurements of dissolved nuclear fuel, with variations of parameters such as burnup and cooling time, as a means to verify process conditions in reprocessing facilities [45, 46, 47]. A multivariate technique applied to fission product content has been demonstrated to determine the fresh fuel type and reactor type from which simulated nuclear fuels originated [48]. Moreover, pattern recognition and machine learning techniques have previously proven very useful in the field of nuclear forensics. Such procedures have been applied to synthetic analytical chemistry data, where measurements of isotopic and elemental compositions of used nuclear fuel were simulated, with the purpose to determine the reactor type from which the fuel originated [49]. Similarly, the capabilities of applying multivariate analysis techniques to simulated uranium and plutonium isotopic concentrations have been investigated, with the aim to identify unknown nuclear material in the environment. The results suggest that plutonium isotopes can be used to determine both the source reactor type and the initial fuel composition [50].

6.1 Regression and classification

Regression analysis is a set of statistical techniques for estimating the relationships among continuous variables, i.e. how the value of a dependent variable,
Figure 6.1. Linear regression allows us to model relationships between a predictor variable \((x)\), and a response variable \((y)\). Here, the straight line can be used to make a prediction of the value \(\hat{y}\), that corresponds to a new observation, \(\hat{x}\).

or response variable, changes when one or more independent variables, or predictors, are varied. Regression techniques are widely used for prediction and forecasting. Perhaps the simplest example of the regression procedure is fitting a straight line to a set of data points representing \(y\) as a function of \(x\), see figure 6.1. Using the fitted line, one can predict a response value to a new observation, \(\hat{x}\).

Whereas regression techniques deal with continuous response values, classification techniques analyze the predictor variables to assign a discrete category, or class, to a new observation. In the example shown in figure 6.2, the two-dimensional space is divided into two regions by a linear decision boundary, or classifier. All data points falling on one side of the boundary are estimated to belong to 'Class 1', and the points on the other side are estimated to belong to 'Class 2'. A new observation with two arbitrary predictor values \(\hat{x}_1\) and \(\hat{x}_2\) will be categorized as one of the two classes.

The examples in figures 6.1 and 6.2 may seem trivial, but the addition of just a few more variables and, hence, a few more dimensions, can quickly make a problem difficult to survey. Here, having a multivariate analysis tool which can extract and highlight information about the most important correlations between the variables, regardless of the number of measured isotopes, may be profoundly useful.

Both regression and classification are examples of supervised machine learning, which can be utilized when there exists a training set of observations with
known predictor and response values. In a training procedure, a supervised learning algorithm analyzes correlations in the training data and produces a generalized function that can be used for mapping new examples. A trained model can be applied to predictor data representing new observations, resulting in an estimate of the corresponding response values. Figure 6.3 illustrates the process with a simple flowsheet.

\[ \hat{Y} \] for new data \( \hat{X} \) is estimated using a trained model. Figure 6.3. A model for the relationship between the variables \( X \) and \( Y \), trained with supervised machine learning algorithms, can be applied to new observations, \( \hat{X} \), resulting in an estimate of the corresponding response value, \( \hat{Y} \).
6.2 The use of different data sets for training, optimization and evaluation of a multivariate method

The regression analysis process in paper V can be divided into the steps presented in figure 6.4. By performing an optimization step, hyperparameters associated to the regression algorithm are adjusted in order to improve the performance of the regression model. This step can be carried out using training data or, as in this work, a separate optimization data set. In the training procedure, the regression algorithm analyzes correlations between isotopic activities and fuel parameters of the fuels in the training data set, and produces a generalized function that can be used for mapping new examples. Next, the trained model is applied to the evaluation data set, resulting in a prediction of the fuel parameters. By comparing the predicted fuel parameters to the known true values in the evaluation data set, an assessment of the regression model can be attained.

Figure 6.4. The regression analysis in this work comprised four steps.

6.3 Multivariate analysis techniques

In papers IV and V, several different regression and classification techniques are examined. Sections 6.3.1–6.3.3 briefly describe three of the most valuable techniques used in the papers. Principal component analysis (PCA) can be used for visualization of data, and may facilitate its interpretation. Decision trees provide simple yet powerful ways to perform regression or classification analyses. They are based on asking a series of questions about each observation in the training data, forming a tree from all possible series of answers. Finally, support vector machines (SVM), which can also be used for both regression and classification purposes, are found to be among the most successful techniques when it comes to predicting nuclear fuel parameters and fuel type. More information about all the employed techniques can be found in papers IV and V and, for more thorough descriptions, see e.g. [51, 52, 53].

6.3.1 Principal component analysis

Principal component analysis is a multivariate statistical tool that is frequently used for dimensionality reduction purposes, and can be used to facilitate visual
examination of a data set. The idea behind PCA is to project a multidimensional data set of independent variables onto a new set of orthonormal vectors, or principal components (PCs), that form a new coordinate system. The PCs are linear combinations of the original variables. They are identified in order of significance; the first PC being the most significant one in explaining the variance within the data set, and the second one being the second most important, while being orthogonal to the first component, etc. It is common that most of the variation can be explained by the first few PCs. By using only the first few PCs, the dimensionality of the transformed data can be reduced without losing much information.

6.3.2 Decision trees
Regression trees partition the feature space into a set of hyper-rectangles by asking a series of questions about the features of each observation in the training data. When a prediction is made for a new observation, the same set of questions is asked, and the new observation is placed in one of the rectangles according to the answers. The predicted value is the average value of the observations from the training data belonging to the same rectangle. Regression trees have been praised for their simplicity and ease of interpretation and for their ability to handle complex data.

Figure 6.5 illustrates a regression tree made for prediction of cooling time, that has been trained with data from the regression analysis in paper V. When predicting the cooling time of a new unknown fuel, one starts at the root node, to the left in the figure 6.5, and begins following a path to the right, along the tree branches. Each node in the tree structure represents a question, and the branches represent possible answers. The path will end in a so-called ‘leaf node’, which holds the value of the predicted cooling time. The tree in the figure is heavily pruned, meaning that fewer questions are asked, and the number of possible outcomes of the regression is reduced. In this case, the original tree had 375 nodes.

6.3.3 Support vector machines
The linear decision boundary in figure 6.2 is not the only possible line that can separate the two classes. In fact, there are infinitely many ways to fit a line in the gap between the observations from the two classes. SVM classifiers achieve good separation between classes by choosing the line or hyperplane that has the largest distance to the nearest training-data point of any class. In the training data, if there is a large margin between the hyperplane and the observations closest to it, the margin will also, ideally, be large in the test data. The larger the distance between a test object and the hyperplane, the more certain the classification is.
In the case of nonlinear input data, an SVM can project the data onto a high-
or infinite-dimensional space, in which linear patterns can be found. The SVM
consists of two parts, that:

1. project input data onto the high-dimensional feature space, and
2. use a learning algorithm to find hyperplanes in this feature space, which
can be used for classification or regression.

The model is finally transformed back to the original coordinate system, where
it is typically nonlinear. This allows for great flexibility in the modeling, and
SVMs are generally considered to perform very well when applied to real-
world problems. Figure 6.6 shows the contour of a hyperplane, obtained using
SVM, that separates two classes in a three-dimensional space.
Figure 6.5. This regression tree was trained using UOX fuels with cooling times of 0–1 y. Each node in the tree structure represents a question, e.g. “is the relative activity of $^{141}$Ce in the fuel greater than $3.25 \cdot 10^{-4}$”, and each branch represents a possible answer. By answering questions about a fuel with unknown cooling time, a path from left to right in the figure is created. The path ends in a value which represents the predicted cooling time of the fuel.
Figure 6.6. Two classes separated by a hyperplane which is clearly nonlinear in the 3D space.
7. Studies of the use of multivariate analysis tools for nuclear safeguards purposes

As discussed in chapter 5, discrimination between UOX and MOX fuel assemblies and determination of the fuel parameters cooling time, burnup and initial fissile content, is important prior to dissolution in a recycling facility. In two studies, described in papers IV and V, irradiated PWR fuels of UOX and MOX type were simulated, and the total gamma activities of the isotopes listed in table 5.1 were collected. In the subsequent data analyses, aimed to either discriminate between UOX and MOX fuels or determinate the fuel parameters, each isotope activity acted as a predictor variable. In paper IV, the classification study, there was one response variable with two possible values: UOX and MOX. The regression study had tree response variables: cooling time, burnup and initial fissile content.

Three data sets of simulated gamma-ray intensities were generated for specific purposes: a training set was used to train the multivariate model to predict the fuel parameters, an optimization set was used to fine-tune the multivariate model itself and an evaluation set was generated for assessing the overall performance of the methodology. In chapter 7.1, the generation of these data sets is described.

Gamma-ray emissions were chosen as input to the analysis algorithms, since gamma-ray spectroscopy is a widely used technique within the field of safeguards. Multivariate methods can handle an arbitrary number of variables, entire gamma spectra or even other types of detector signals. In papers IV and V a few gamma-ray energy peaks have been selected to demonstrate the concept. The number of energy peaks is larger than what is typically used in nuclear safeguards evaluations today, and still small enough to show and explain the relevance and importance of each selected energy peak.

In table 5.1, nine gamma-emitting isotopes, frequently mentioned in the literature relating to fuel characterization, are listed together with half-lives, gamma-ray energies of some prominent gamma peaks and their emission probabilities. They cover a wide range of half-lives, such that at least a few of them are detectable up to a cooling time of 40 years. Furthermore, they exhibit prominent gamma energies that can be identified using passive high-resolution gamma-ray spectroscopy.
7.1 Simulations of nuclear fuel

Simulations of PWR fuel with different combinations of fuel parameters were performed using the Monte Carlo-code Serpent [54]. The simulation procedure is described in more detail in papers IV and V. In brief, an infinite lattice of PWR fuel rods was modeled, with the fuel parameters initial fissile content, burnup, and cooling time, ranging between the values specified in table 7.1.

Table 7.1. Ranges of fuel parameters for modeled PWR fuels in papers IV and V.

<table>
<thead>
<tr>
<th>Fuel parameter</th>
<th>Paper IV</th>
<th>Paper V</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial fissile content</td>
<td>2–5%</td>
<td>2–5%</td>
</tr>
<tr>
<td>Burnup</td>
<td>20–60 MWd/kgHM</td>
<td>1–60 MWd/kgHM</td>
</tr>
<tr>
<td>Cooling time</td>
<td>0–40 years</td>
<td>0–20 years</td>
</tr>
</tbody>
</table>

The total number of modeled fuels in each separate data set can be seen in table 7.2. Each simulated fuel was given a unique set of parameter values. In paper IV, the cooling times were generated with the probability density function $1/CT$, such that each of the four sets of different cooling times would have roughly the same number of observations, see figure 7.1. In paper V, on the other hand, a flat distribution was used.

Table 7.2. Approximate numbers of modeled PWR fuels in the different data sets used in papers IV and V.

<table>
<thead>
<tr>
<th>Data set</th>
<th>Paper IV</th>
<th>Paper V</th>
</tr>
</thead>
<tbody>
<tr>
<td>Training</td>
<td>14,000</td>
<td>55,000</td>
</tr>
<tr>
<td>Optimization</td>
<td>N/A</td>
<td>14,000</td>
</tr>
<tr>
<td>Evaluation</td>
<td>1,400</td>
<td>14,000</td>
</tr>
</tbody>
</table>

Since high burnup levels are not achievable for fuels with low initial fissile contents, the empirical relationship between initial enrichment ($\varepsilon$, in percent), and maximal reachable burnup ($BU$, in MWd/kgHM) shown in equation 7.1 was applied to the data. Data points not satisfying this constraint were not included in generated data sets.

$$\varepsilon = l \cdot BU^k$$  \hspace{1cm} (7.1)

with $l = 0.31$ and $k = 0.65$ [55].

The concentrations of each of the nine selected isotopes in table 7.3 were generated by simulating irradiation and decay of the fuel. The concentrations were later used to calculate emitted isotope-specific total gamma activities, using equation 7.2, and the data was rescaled such that the sum of source activities for the chosen set of isotopes equaled 1.

$$A = \frac{N \cdot \ln 2}{t_{1/2}}$$  \hspace{1cm} (7.2)
Figure 7.1. In paper IV, cooling times were generated with the probability density function $1/CT$, to ensure that the four groups based on different cooling times would have roughly the same number of observations.

Table 7.3. List of the selected isotopes and their correspondence to the cooling time (CT) groups used in papers IV and V. Group 4 was used in paper IV only.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life</th>
<th>Group 1</th>
<th>Group 2</th>
<th>Group 3</th>
<th>Group 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{141}$Ce</td>
<td>32.5 d</td>
<td>✓</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{95}$Nb</td>
<td>35.0 d</td>
<td>✓</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{91}$Y</td>
<td>58.5 d</td>
<td>✓</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{95}$Zr</td>
<td>64.0 d</td>
<td>✓</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{144}$Ce</td>
<td>285 d</td>
<td>✓</td>
<td>✓</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{106}$Ru</td>
<td>372 d</td>
<td>✓</td>
<td>✓</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>2.07 y</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>$^{154}$Eu</td>
<td>8.60 y</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>30.1 y</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
</tr>
</tbody>
</table>

Table 7.4 shows an example of preprocessed data used for the multivariate analysis in paper IV, and figure 7.2 shows the relative activities of the nine selected isotopes as a function of the cooling time of a fuel, ranging from 0 to 20 years. Having the longest half-life, $^{137}$Cs clearly dominates the total activity at long cooling times.
Table 7.4. Example of preprocessed data. Relative gamma source activities [a.u.] in four modeled fuel items from paper IV.

<table>
<thead>
<tr>
<th></th>
<th>$^{106}$Ru</th>
<th>$^{134}$Cs</th>
<th>$^{137}$Cs</th>
<th>$^{154}$Eu</th>
<th>$^{144}$Ce</th>
<th>Class</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.479</td>
<td>0.0474</td>
<td>0.00454</td>
<td>0.00118</td>
<td>0.468</td>
<td>MOX</td>
</tr>
<tr>
<td>2</td>
<td>0.496</td>
<td>0.0580</td>
<td>0.00647</td>
<td>0.00164</td>
<td>0.438</td>
<td>MOX</td>
</tr>
<tr>
<td>3</td>
<td>0.290</td>
<td>0.0587</td>
<td>0.00388</td>
<td>0.000710</td>
<td>0.646</td>
<td>UOX</td>
</tr>
<tr>
<td>4</td>
<td>0.304</td>
<td>0.0713</td>
<td>0.00539</td>
<td>0.000963</td>
<td>0.619</td>
<td>UOX</td>
</tr>
</tbody>
</table>

Figure 7.2. Relative activities of the selected isotopes in a fuel, as a function of cooling time. This particular fuel is of UOX type, and has an initial fissile content of 4.5% and a burnup level of 60 MWd/kgHM.

7.2 Visualization and interpretation of data

Before attempting to make predictions based on newly produced data sets, it is good practice to explore the data visually. By doing so, one may learn something about the data, and possibly detect anomalies and mistakes in the data acquisition.

As an example of the visualization, figure 7.3 shows short-cooled (0–1 y) MOX and UOX fuels. The clusters that represent the two fuel types are neatly ordered and behave similarly, which indicates that the data is well suited for analysis with dimensionality reducing multivariate analysis methods. Furthermore, the two groups are well separated in space, indicating that a classification process is likely to succeed.
Figure 7.3. Data points from short-cooled fuel (0–1 years) represented in three dimensions spanned by the three most important principal components obtained via PCA. MOX and UOX fuels form two separate clusters.

The training data produced for paper V was analyzed using PCA in order to reduce the isotope activity data to three dimensions. Figures 7.4 and 7.5 show training data based on MOX fuels and UOX fuels, respectively, with cooling times 1–10 y. Each figure shows the same dataset four times, but with the data points colored differently depending on four fuel parameters: cooling time, burnup, initial fissile content, and plutonium mass fraction. The color spectrum is a sliding scale where blue represents low values, and red represents high values.

It is easy to notice that there are correlations between the predictors and the fuel parameters. In figure 7.4, it appears as if PC1 roughly captures the cooling time, PC2 captures the burnup, and PC3 captures the initial fissile content and the plutonium mass fraction. It makes sense that the initial fissile content and the plutonium mass fraction behave similarly since, before the irradiation begins, plutonium makes up most of the fissile material in MOX fuel. Structures appear in figure 7.5 as well, but here the plutonium mass fraction and the burnup display great similarities, which reflects the fact that plutonium is produced as a function of burnup in UOX fuel.
Figure 7.4. MOX fuel data projected onto a three-dimensional space, and colored according to four different fuel parameters. In each of the four plots, blue represents low values, and red represents high values.
Figure 7.5. UOX fuel data projected onto a three-dimensional space, and colored according to four different fuel parameters. In each of the four plots, blue represents low values, and red represents high values.
7.3 Discrimination between UOX and MOX fuels

Paper IV investigates a novel discrimination methodology for classification of UOX and MOX fuel, based on passive gamma-ray spectroscopy data and multivariate analysis methods. In the study, the simulated gamma-ray activities were used as input to train the seven different linear and nonlinear multivariate classification techniques listed below.

- Classification tree
- k nearest neighbors (kNN)
- Linear discriminant analysis (LDA)
- Naïve Bayes
- Partial least squares discriminant analysis (PLS-DA)
- SVM (with a Gaussian kernel)
- SVM (with a linear kernel)

The trained classifiers were implemented and evaluated with respect to their capabilities to correctly predict the classes of unknown fuel items. Table 7.5 presents the misclassification rates for the evaluated classifiers, in a case where 3% noise was added to the training and evaluation data sets. The best results were acquired when using nonlinear classification techniques, such as the kNN method and the Gaussian kernel SVM. For fuel with cooling times up to 20 years, when it is considered that gamma-rays from the isotope $^{134}\text{Cs}$ can still be efficiently measured, success rates of 100% were obtained. A sensitivity analysis indicated that these methods were also robust to:

1. artificial noise added to the input data as to represent experimental uncertainties,
2. bias error of the simulated reactor power,
3. bias error of the size of the training set, and to
4. bias error of the number of cycles of irradiation.

Receiver operating characteristic (ROC) curves is one means to illustrate the performances of classification methods. As an example, figure 7.6 shows

<table>
<thead>
<tr>
<th>Classification technique</th>
<th>Group 1 C: 0–1 y</th>
<th>Group 2 C: 1–10 y</th>
<th>Group 3 C: 10–20 y</th>
<th>Group 4 C: 20–40 y</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tree</td>
<td>2.21</td>
<td>0.152</td>
<td>0</td>
<td>33.9</td>
</tr>
<tr>
<td>kNN</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>31.5</td>
</tr>
<tr>
<td>LDA</td>
<td>0.736</td>
<td>0</td>
<td>4.12</td>
<td>28.8</td>
</tr>
<tr>
<td>Naïve Bayes</td>
<td>40.8</td>
<td>1.51</td>
<td>1.64</td>
<td>28.4</td>
</tr>
<tr>
<td>PLS-DA</td>
<td>0.735</td>
<td>0</td>
<td>3.70</td>
<td>28.4</td>
</tr>
<tr>
<td>SVM (Gaussian)</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>27.6</td>
</tr>
<tr>
<td>SVM (linear)</td>
<td>0</td>
<td>0</td>
<td>1.24</td>
<td>27.6</td>
</tr>
</tbody>
</table>
ROC curves for the case where the examined fuels have cooling times of 20–40 years and a noise level of 3%. The true positive rate is also known as the probability of detection, whereas the false-positive rate is also known as the probability of false alarm. Here, MOX fuel is selected as the true positive value. The top-left corner of the graph represents a perfect classification, with 100% of the examined MOX fuels correctly classified as MOX, and no UOX fuels mistakenly classified as MOX. The opposite corner represents a 100% misclassification rate, whereas a random guess would fall on the diagonal between the bottom-left and the top-right corners. In figure 7.6, the LDA, Naïve Bayes, and PLS-DA methods are on average the best-performing method as they lie the closest to the top-left corner, whereas the kNN method lies close to the diagonal and thus represents the least successful classifier. A ROC plot for the case with 10% noise and CT=10–20 years is shown in figure 7.7. Despite the high level of noise, all seven methods perform relatively well. The vast difference in classification performance can be explained by the loss of $^{134}$Cs, which can not easily be measured in the 20–40 y cooling time interval. With only two isotopes left, $^{137}$Cs and $^{154}$Eu, there is not enough information in the data to discriminate between UOX and MOX fuels.

Figure 7.6. ROC curves illustrating the poor performances of all seven classification methods for data with CT = 20–40 y and 3% added noise.
7.4 Determination of cooling time, burnup and initial fissile content

In paper V, the determination of cooling time, burnup and initial fissile content, based on passive gamma-ray spectroscopy data and multivariate analysis methods, was investigated. First of all, cooling times were estimated using decision trees and only the three isotopes of group 3 in table 7.3. This analysis can be performed with no prior information about a fuel, regardless of its cooling time. The results show that, for 95% of the UOX fuels, a prediction of the cooling time falls less than 6 months from the true cooling time, and for MOX fuels, the corresponding value is around 7 months. It thus appears conceivable to divide the data into subsets according to the cooling time intervals in table 7.3.

Next, all three fuel parameters were individually determined. The regression models were trained for each combination of a fuel type (UOX or MOX) and a group based on cooling time (see table 7.3). The regression analyses followed the process presented in chapter 6.2 and figure 6.4, and the methods used for determining the different fuel parameters were:
Cooling time: Regression decision tree
Burnup: SVM regression
Initial fissile content: SVM regression

By applying the models to corresponding evaluation data, the fuel parameters for each evaluation fuel were determined. Performance of the regression was examined by evaluation of the root mean square error (RMSE) and the 95% prediction intervals (95% P.I. – the estimate of an interval in which 95% of future observations will fall), that resulted from each analysis. The prediction errors of the regressions are listed in Table 7.6.

**Table 7.6. Performance of the regression of cooling time, burnup and initial fissile content in terms of root-mean-square errors (RMSE) and 95% prediction intervals (P.I.).**

<table>
<thead>
<tr>
<th></th>
<th>Cooling time [days]</th>
<th>Burnup [MWd/kgHM]</th>
<th>Initial fissile content [percentage points]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>RMSE</td>
<td>95% P.I.</td>
<td>RMSE</td>
</tr>
<tr>
<td><strong>Group 1</strong></td>
<td><strong>UOX</strong></td>
<td></td>
<td><strong>MOX</strong></td>
</tr>
<tr>
<td><strong>CT: 0-1 y</strong></td>
<td>14 ± 26</td>
<td>0.77 ± 1.6</td>
<td>0.56 ± 1.2</td>
</tr>
<tr>
<td><strong>Group 2</strong></td>
<td><strong>UOX</strong></td>
<td></td>
<td><strong>MOX</strong></td>
</tr>
<tr>
<td><strong>CT: 1-10 y</strong></td>
<td>34 ± 69</td>
<td>1.4 ± 2.9</td>
<td>0.27 ± 0.50</td>
</tr>
<tr>
<td><strong>Group 3</strong></td>
<td><strong>UOX</strong></td>
<td></td>
<td><strong>MOX</strong></td>
</tr>
<tr>
<td><strong>CT: 10-20 y</strong></td>
<td>175 ± 221</td>
<td>2.5 ± 5.1</td>
<td>0.78 ± 1.4</td>
</tr>
<tr>
<td></td>
<td><strong>MOX</strong></td>
<td></td>
<td><strong>MOX</strong></td>
</tr>
<tr>
<td></td>
<td>114 ± 227</td>
<td>2.9 ± 6.7</td>
<td>0.73 ± 1.4</td>
</tr>
</tbody>
</table>

As seen in the table, the cooling time and the burnup can be determined with an accuracy that is low relative to the variances of the respective parameters. However, the accuracy of the determined initial fissile content in terms of the 95% prediction interval reaches up to 1.4 percentage points for both UOX and MOX fuels in group 3 (long cooling times). This is relatively large as compared to the variance of this parameter in the evaluation data, where it was varied between 2 and 5 percent. From the bottom-right plots in figure 7.8, showing the predicted vs. true cooling time, burnup and initial fissile content values for UOX fuels, and in figure 7.9, showing the corresponding residuals, it is evident that the regression models used in this study are not able to successfully capture the true initial fissile content.

A sensitivity analysis was performed, although not included in the paper, in order to evaluate the sensitivity of the regression methods to artificial noise, much like the sensitivity analysis in chapter 7.3. Besides the nominal 3% noise level, analyses were carried out with 5% and 10% noise added to the data. The results are shown in figure 7.10. In general, the addition of more noise to the input data does not seem to imply great deterioration of the accuracy of the results, and one may conclude that the regression models are fairly robust.
Figure 7.8. Predicted vs. true cooling time (CT), burnup (BU) and initial fissile content (IFC) values (top-down) for UOX fuels with cooling times 0–1 y, 1–10 y, and 10–20 y (left-right). The regression models were less successful in predicting initial fissile content, than the other two fuel parameters.
Figure 7.9. Cooling time (CT), burnup (BU) and initial fissile content (IFC) residuals (top-down) for UOX fuels with cooling times 0–1 y, 1–10 y, and 10–20 y (left-right). A good regression model results in residuals that are free from visible patterns such as the linear behavior seen in the bottom-right plot.

Figure 7.10. Prediction errors increase as more noise is added to the training and evaluation data sets.
7.5 Determination of plutonium content

As indicated in section 7.4, it is possible to use multivariate analysis methods to perform regression of the fuel parameters in terms of burnup, cooling time and, albeit with a lesser degree of accuracy, also the initial fissile content. Inspired by the relative success in determining these parameters, an attempt was made to, using SVM regression, also determine the total plutonium content in irradiated UOX fuels as part of paper V. Determination of this quantity is of special interest because the IAEA regards all plutonium isotopes as relevant from a safeguards point of view.

Results were encouraging, with RMSE and the 95% P.I. of the predicted mass fractions not exceeding 0.06 and 0.12 percentage points, respectively, see table 7.7 and figure 7.11. These promising results are likely facilitated by the nonexistent plutonium content in fresh UOX fuel. The buildup of plutonium essentially follows the burnup, as the fertile isotope $^{238}$U breeds $^{239}$Pu.

Similarly, but not included in the paper, the plutonium content in MOX fuels has been estimated, see figure 7.12. Here, the absolute prediction errors are 25 to 30 times larger than for UOX fuel. It is not obvious why the results are so different, but one possible reason might be that, aside from burnup, the initial fissile content plays an important role in the prediction of the plutonium mass.

Table 7.7. Performance of the SVM regression of plutonium content in terms of root-mean-square errors (RMSE) and 95% prediction intervals (P.I.).

<table>
<thead>
<tr>
<th>Pu mass fraction [percentage points]</th>
<th>RMSE</th>
<th>95% P.I.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Group 1</td>
<td>UOX</td>
<td>0.032</td>
</tr>
<tr>
<td>CT: 0–1 y</td>
<td>MOX</td>
<td>0.87</td>
</tr>
<tr>
<td>Group 2</td>
<td>UOX</td>
<td>0.031</td>
</tr>
<tr>
<td>CT: 0–1 y</td>
<td>MOX</td>
<td>0.90</td>
</tr>
<tr>
<td>Group 3</td>
<td>UOX</td>
<td>0.061</td>
</tr>
<tr>
<td>CT: 10–20 y</td>
<td>MOX</td>
<td>1.6</td>
</tr>
</tbody>
</table>

7.6 A case study concerning misdeclared fuel assemblies

The relative abundances of the isotopes $^{134}$Cs, $^{137}$Cs and $^{154}$Eu have, as described in chapter 5, historically been utilized for calculations of the burnup levels of irradiated fuel, thereby facilitating verification of operators’ declarations. This type of verification is today extensively used. In this section, it is investigated if there are verification cases where the traditionally used tech-
Figure 7.11. Evaluation of the plutonium content determination in UOX fuels with cooling times of 0–1 y, 1–10 y, and 10–20 y. The regression residuals of the plutonium concentration are relatively small in all three cases.

Figure 7.12. Evaluation of the plutonium content determination in MOX fuels with cooling times of 0–1 y, 1–10 y, and 10–20 y. The prediction errors are much larger than for UOX fuel, likely since the pattern for build-up and decay of the plutonium isotopes is more intricate.

In this previously unpublished study, using the UOX evaluation data set from section 7.4 and 7.5, the ratios $^{134}\text{Cs}/^{137}\text{Cs}$ and $^{154}\text{Eu}/^{137}\text{Cs}$ have been calculated and subsequently plotted versus burnup and cooling time as shown in figure 7.13 (left) and 7.14 (left), respectively. A LOWESS (locally weighted scatterplot smoothing [56]) procedure was used to fit a surface to the data, to visualize the continuous change in the ratios. The corresponding two-dimensional contour plot, shown in figure 7.13 (right) and 7.14 (right) illustrates that some fuels with different combinations of burnup and cooling time give rise to the...
same isotopic ratios. For instance, if a fuel is determined to have a $^{154}\text{Eu}/^{137}\text{Cs}$ ratio of 0.2, the fuel can have any combination of burnup and cooling time that corresponds to the contour line for the ratio 0.2. When the two contour plots, corresponding to the two ratios, are superimposed as shown in figure 7.15, one can note that there are indeed lines that intersect twice, representing two burnup–cooling time combinations with identical $^{134}\text{Cs}/^{137}\text{Cs}$ and $^{154}\text{Eu}/^{137}\text{Cs}$ ratios. Hence, in a scenario where measurements of the two isotope ratios provide the only verification of operator data, fuels with those burnup–cooling time combinations could be interchanged without discovery.

Figure 7.13. Lowess surface (left) and Lowess contour plot (right) illustrating how the $^{134}\text{Cs}/^{137}\text{Cs}$ ratio depends on cooling time and burnup.

Figure 7.14. Lowess surface (left) and Lowess contour plot (right) illustrating how the $^{154}\text{Eu}/^{137}\text{Cs}$ ratio depends on cooling time and burnup.
100 pairs of fuels were found from studying an arbitrary number of contour intersections as illustrated in figure 7.15. The identified fuels were modeled in Serpent, using the burnup and cooling time combinations shown in figure 7.15, and an initial fissile content estimated using regression trees. Data from paper V was used to train new SVM models for the UOX fuels, using only the relative activities of $^{134}$Cs, $^{137}$Cs and $^{154}$Eu as predictor variables. Burnup levels and cooling times were predicted and compared with the true values, see figure 7.16.

![Figure 7.15](image)

*Figure 7.15.* Points that have the same colors and are connected with dotted lines represent fuels that share the same two isotope ratios, and therefore cannot be separated using traditional analysis methods.

Because of uncertainties in the estimation of initial fuel contents required for simulations, and because only three isotopes were used for the analysis, the prediction errors were slightly larger than those presented in table 7.6. However, the predictions were accurate enough to distinguish between the two fuels in each pair. Hence, the method presented here may be able to detect an anomaly in the case of a fuel interchange. However, nearly all fuels in the identified fuel pairs had burnup levels of 35 MWd/kgHM or more, which means that the plutonium would be of rather poor quality. A state that aims to produce weapons-grade plutonium by irradiating uranium fuel would most likely
Figure 7.16. The plot shows burnup and cooling time prediction errors, with lines marking RMSE and 95% P.I. The predictions were, for almost all observations, accurate enough to distinguish between the two fuels in each pair.

decide to remove and reprocess the fuel at a lower burnup. In such a case, the two isotope ratios $^{134}\text{Cs}/^{137}\text{Cs}$ and $^{154}\text{Eu}/^{137}\text{Cs}$ are sufficient for verification.
8. Conclusions and outlook

Gen IV nuclear energy systems are aimed to be the least desirable route for diversification, and efforts should be made to include as much inherent proliferation resistance as reasonably achievable into new facility and system designs. Iterative evaluation of proliferation resistance by using two different assessment methodologies – TOPS and PR&PP – as proposed in this thesis, is suggested to be used as a guide towards a refined facility design, which in turn can be subject to analysis in the following iteration. As the number of iterations increases, the facility design will become better defined, and the proliferation resistance assessments increasingly detailed. Hence, it may act as an aid in the technical development of safeguards instrumentation in the areas where nuclear facilities and fuel cycle systems are the most inherently vulnerable.

No system is considered to be more resistant to proliferation, than its weakest link. In this work, the TOPS methodology has been applied to three different types of fuel cycles involving a lead-cooled fast reactor and its associated recycling and fuel fabrication facilities. Not surprisingly, recycling facilities were identified as sensitive from a safeguards perspective, since they are bulk handling facilities and potentially hold pure plutonium streams. Fuel reprocessing facilities have been further examined, and key measurement points for safeguards verification measurements have been suggested. Nondestructive measurements for verification in the head-end area of the recycling facility are identified as being essential, since it is the last stage in which it is possible to verify intact fuel items. The results of the evaluations could have been more detailed, and associated with less uncertainties, if the facility designs were developed further, and if several experts in relevant subject areas were consulted to bring their points of view to the assessments.

Based on the identified need to measure irradiated fuel assemblies prior to dissolution in the recycling facility, analysis of nondestructive measurements for verification of nuclear fuel has been studied in more depth. More specifically, new methods used for analyzing gamma-ray spectroscopy data using multivariate analysis methods have been investigated. Fuel parameters of modeled nuclear fuel were determined with no reliance on operator-declared data. The parameters cooling time, burnup and initial fissile content were determined using decision tree and SVM regression, whereas the fuel type (UOX or MOX) was successfully determined using nonlinear classifiers, e.g. Gaussian kernel SVM. The results are promising and indicate that the nuclear safeguards regime may benefit from including multivariate techniques. It must be
emphasized, however, that experimental verification of the multivariate analysis techniques is necessary, and such verification using experimental data are anticipated in the near future.

According to the investigations presented in this thesis, passive measurements of gamma-rays may suffice for verification of fuel assemblies cooled for up to 20 years. Combinations of gamma-ray spectroscopy with other types of nondestructive measurements can extend this cooling time span and help extract more information about the irradiated fuels, and future studies incorporating measurements of neutron radiation are foreseen.
Acknowledgments

"It was the best of times, it was the worst of times"

– Charles Dickens, A Tale of Two Cities

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• everyone I failed to mention.

Now it’s time for me to move on to my next adventure, and it’s going to be kjempeflott!
Summary in Swedish

Klimatförändringar och dess effekter är bland de svåraste utmaningarna som världen ställts inför. För att bekämpa klimatpåverkan orsakad av växthusgaser krävs stora insatser för att fasa ut fossila bränslen och de utsläpp som de genererar. Driften av kärnkraftverk är i det närmaste koldioxidneutral, och ger upphov till endast små mängder utsläpp ur ett livscykelperspektiv. Kärnkraftens roll som energiproducent kan därför förväntas bli viktigare allt eftersom beroendet av fossila bränslen minskas till förmån för tekniker som inte genererar lika stora mängder växthusgaser.

Samtidigt står kärnkraften inför egna utmaningar inom flera områden såsom säkerhet vid drift och uranbrytning, slutförvaring av bränsle, och icke-spridning av kärnämnen som kan användas för vapentillverkning. En ny generation av kärnkraftssystem kallad Generation IV är under utveckling för att angripa bland annat dessa frågor. Generation IV-systemen skall utgöra ett säkert och uthålligt alternativ för energiförsörjning, och innefattar nya kärnbränslecyclor som delvis baseras på återvinning av använt kärnbränsle från dagens reaktorer. Vidare ska skyddet mot stöld och spridning av kärnämnen (t.ex. uran och plutonium) stärkas in de nya systemen.

Kärnämneskontroll, eller safeguards, syftar till att säkerställa att kärnteknisk verksamhet bedrivs i enlighet med nationella och internationella krav, så att kärnämnen och kärntekniska anläggningar inte används för framställning av kärnvapen. Länder som inte har kärnvapen och är anslutna till Internationella atomenergiorganets (IAEA) icke-spridningsavtal, måste bokföra och rapportera sina innehav av kärnämnen och ställa sin kärntekniska verksamhet till förfrågande för IAEA:s inspektioner. Vid inspektion av en anläggning kontrolleras dess bokföring, och det verifieras, bland annat genom strålningssättningar, att materialet finns där det är sagt och att det används på rätt sätt.

Detta arbete behandlar kärnämneskontroll och icke-spridning i framtida Generation IV-system. Artiklarna I–III som tillhör denna avhandling berör mer specifikt det motstånd mot kärnämnesspridning som finns inneboende i olika delar av ett kärnkraftssystem med bränsleåtervinning. Ett sådant motstånd kan exempelvis bestå i svårigheter att fysiskt komma åt fissionerat material, eller egen-skaper hos materialet som gör att det inte lämpar sig för vapenproduktion. Flera verktyg har utvecklats för att på ett strukturerat sätt kunna utvärdera ett systems motstånd mot spridning av kärnämnen. Användning av två sådana verktyg har i detta arbete demonsterrats på tänkta Generation IV-system, med syfte att bland annat identifiera svaga punkter där extra kontrollinsatser av IAEA kan vara lämpliga. Ett sådant område är anläggningen för återvinning av
kärnbränsle, där bränslet löses upp och genomgår kemisk separation, och där materialströmmar av rent plutonium kan förekomma. I synnerhet mottagningsdelen av en återviningsanläggning, där bränslet tas emot i intakt form, identifieras i detta arbete som en lämplig punkt för mätningar av gammastrålning från bränslet. Det beror på att verifikation av materialet försvaras efter att bränslet lösts upp.

Artiklarna IV och V berör de mätningar på gammastrålning från använt kärnbränsle som kan tänkas ske vid mottagning av bränsle på en återviningsanläggning. Olika energinivåer hos de uppmätta gammastrålarna fungerar som fingeravtryck för olika isotoper, och kan därmed ge information om vilka isotoper, och i hur stora mängder, som finns i bränslet. Det kan i sin tur göra det möjligt att bekräfta de uppgifter om bränslet som lämnats av reaktoroperatören. I denna avhandling undersöks ett nytt sätt att, med hjälp av multivariata statistiska metoder, analysera data från gammamätningar. Sådana analysmetoder gör det möjligt att ta fram betydelsefull information från stora mängder av data som till synes saknar struktur. Genomförd analys av simulerad gammastrålning i artikel IV och V tyder på att metoderna kan vara värdefulla för bestämning av egenskaper hos bränslet, såsom utbränning, kyltid och anrikning, vilket på sikt kan gynna kärnämnescollagen och dess inspektörer. Nästa steg i undersökningarna av multivariat analys för kärnämnescollagen bör vara att testa metoderna på experimentella data från mätningar på verkligt kärnbränsle.
References


[45] Christopher R. Orton, Carlos G. Fraga, Richard N. Christensen, and Jon M. Schwantes. Proof of concept simulations of the multi-isotope process monitor:


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