Compilation and evaluation of validations of computer codes for spent nuclear fuel – a review

Project for the degree of Bachelor of Science
2019

Author: Staffan Fors

Supervisors: Peter Jansson, Uppsala University
Kristina Stenström, Lund University

Abstract:

This report describes a systematic review regarding validations of calculations by measurements of isotopic compositions, or decay heat, on spent nuclear fuel, SNF. 15 relevant or otherwise interesting studies, were found. Two reports were chosen among the found studies and analysed. One of the studies described validations made with experimental data from measurements of isotopic compositions on samples from SNF from three different BWR in Germany, Japan and USA. The other report treated validation of the ORIGEN code by calorimetric measurements of decay heat from SNF. The measurement were made at CLAB in Sweden.

The design requirements for decay heat from the SNF in the final repository is in the order of a few percentage. The results from calculations of isotopic compositions is significantly higher for key isotopes regarding decay heat, like ^{137}Cs,^{90}Sr and ^{241}Am. Calorimetric measurements gives, more accurate results but the precision is not high enough for design purposes. Another observation from both the review and from analysing selected reports is the need for much more experimental measurements on SNF. Both regarding isotopic composition and measurements based on other methods eg. calorimetric measurements of decay heat.

Table of contents

List of figures:

List of tables:

List of acronyms:

SNF	-	Spent Nuclear Fuel
BWR	-	Boiling Water Reactor
PWR	-	Pressurised Water Reactor
LWR	-	Light Water Reactor
SKB	-	Svensk Kärnbränslehantering
ORIGEN	-	Oak Ridge Isotope GENeration
SCALE	-	Standardized Computer Analyses for Licensing Evaluation
MOX	-	Mixed Oxide Fuel
JPDR	-	Japan Demonstration Reactor
PNL	-	Pacific Northwest Laboratory
EJCR	-	European Joint Research Center
JAERI	-	Japanese Atomic Energy Research Institute
ENDF	-	Evaluated Nuclear Data File
CLAB	-	Centrallager för använt kärnbränsle
SFCOMPO	-	Spent Fuel Isotopic Composition database

1.   Introduction

It is essential to have knowledge of the isotopic composition of irradiated nuclear fuel before it is placed into the final nuclear waste repository. The isotopic composition of irradiated nuclear fuel determines how much residual heat each fuel-bundle will generate in the waste repository. It is also important to be fully informed of the nuclear reactivity of spent nuclear fuel (SNF) in order to reduce the risk for a reactivity accident in any step of the waste management chain. This is because SNF still contains is so much fissile material that in an unfavourable geometric configuration can achieve criticality. The isotopic composition is also highly relevant when it comes to the aspect of the safe-guarding of fissile material against nuclear proliferation, i.e. how suitable it is as material for building nuclear weapons.

The isotopic composition of SNF is usually determined by calculations. Computer programs used for this purpose are e.g. ORIGEN Oak Ridge Isotope GENeration, developed by Oak Ridge National Laboratory, which is part of the SCALE package [1]**.** This makes it necessary to validate the calculations against radiochemical or other measurements. Several such studies have been performed eg Ref. [13]. Nevertheless, a more comprehensive compilation and review of evaluations of calculated values has not been found in the literature.

The main aim of this thesis is to make a compilation and review of studies of experimental validations of calculations of isotopic compositions of irradiated nuclear fuel. Secondary, it suggests which studies are relevant for drawing conclusions concerning comparisons between calculations and measurements. Furthermore, this thesis aims to evaluate the difference between calculations and measurements.

Scientific Method

The method chosen for this thesis is to make a systematic review of relevant studies of experimental validations of calculated isotope compositions of spent nuclear fuel. A description of how to make a review in a scientific research can be found in Ref. [1]. Reviews are valuable for summarizing previous work. The review in this thesis consists of a survey regarding which validation studies have been published. Thereafter, the found studies are graded after relevance. One or more reports are then selected according to specific criteria and are evaluated in respect to accuracy compared to measurements.

This report is organized in one introductory part, where the relevant physics is briefly explained. Secondly, the search result of the review is presented together with analyses of selected reports from the review. At last, an analysis of the review and selected reports are presented together with an outlook about possible areas to investigate in the future.

2 Nuclear Fission

2.1 History and background

The discovery of nuclear fission was a process in several steps. Investigations of artificially induced radioactivity by Irene and Fredric Joliot-Curie were the first step. Later Enrico Fermi found that bombardment of ^{238}U by neutrons had the effect that a new element was formed. For this work he was awarded the 1938 Nobel Prize in Physics.

In later work by Hahn, Meitner and Strassman, results indicated the formation of barium (element 56). This fact formed the idea that uranium was split into fragments. The idea of nuclear fission emerged when Otto Frisch - who worked at the Niels Bohr institute in Copenhagen - visited his aunt, Lise Meitner, in Sweden during Christmas 1939. Frisch informed Bohr of the discovery of fission and soon afterwards Meitner and Frisch submitted a paper to Nature [2], describing the phenomenon. The term fission was borrowed from cell biology.

Frisch was later the first scientist who measured the energy of the fission fragments. He bombarded thorium with neutrons of different energies. Thorium proved to act like uranium with the difference that it was only fissioned by fast neutrons. This led Bohr to investigate which uranium isotope was being fissioned by slow neutrons. At the time, only two uranium isotopes were known: ^{235}U (even Z/odd A) and ^{238}U (even Z/even A). Thorium has only one naturally occurring isotope ^{232}Th (even Z/even A). This led Bohr to the conclusion by logic that ^{235}U must be the nuclide which was fissioned by slow neutrons.

2.2 The mechanism of nuclear fission- The Bohr-Wheeler theory

In 1939 Bohr published a paper stating that of the known isotopes of uranium the lighter must be the one being fissioned and the heavier is only absorbing thermal neutrons followed by a decay [3]. This was experimentally verified via mass spectroscopy by Nier [4]. Bohr did more work with John Wheeler and as a result they presented an analysis and explanation of the experimental results [5]. This famous article presented a theoretical analysis and explanation of the phenomenon of nuclear fission. Two important discoveries were reported in the article:

1. There exist a natural limit, $Z^2/A=48$, beyond which a nucleus is unstable against disintegration by spontaneous fission.

2. For nuclei with $Z^2/A<48$ an activation energy must be supplied. This is called the fission barrier.

Bohr and Wheeler made a model of the nucleus as a liquid drop and the shape of the nucleus is described by a sum of Legendre polynomials. These polynomials are configured to conserve volume during the process of deformation. The total energy of the two contributions, the surface energy U_A, proportional of the surface area of the nucleus, and the electrostatic contribution U_C from the Coulomb potential, is described by the sum of these two contributions. The Coulomb term originates from the mutual repulsion of protons.

If a nucleus is deformed from its original spherical shape, U_A will increase because of the increase in surface area. At the same time U_C will decrease as the nuclear charge will be more spread out. So, if $(U_A + U_C)_{deformed} < (U_A + U_C)_{original}$ the nucleus will be unstable against more deformation and fission. The surface term comes from the fact that nucleons near the surface of the nucleus are less strongly bound compared to more centrally placed nucleons. The discovery of nuclear fission led to the Manhattan project which not only led to the development of fission bombs but also to techniques for enrichment and construction of nuclear reactors. The purpose was at first to enrich ^{235}U to obtain material for the first fission bomb which destroyed Hiroshima. Nuclear reactors were first developed with the purpose to a produce ^{239}Pu for implosion bombs. Trinity, the first nuclear explosion and the Nagasaki bomb was devices built by the implosion technique.

Just after Second World War, USA decided to spread the knowledge of nuclear power with the "Atoms for Peace" program. This started a rapid development of nuclear technology and the construction of reactors for none-military purpose all over the world.

3. Nuclear reactors and fuel

Nuclear fission has been used for the purpose of generating electricity for more than sixty years. The first civilian reactor built for this purpose was Calder Hall in Cumbria, England, which came into operation in 1956.

3.1 Reactor types

Several types of nuclear reactors and fuels have been developed. Today, the most predominant reactor construction is the Light Water Reactor, LWR, which uses ordinary water as both coolant and moderator. This simplifies the reactor construction and improves safety in relation to other types.

LWRs are divided into Boiling Water Reactors (BWRs) and Pressurized Water Reactors (PWRs). The major difference between BWRs and PWRs is the way heat is being transported from the nuclear fuel in the reactor core to the turbines.

In a PWR, the water in the primary cooling circuit is heated at very high pressure without being allowed to boil. The heat of the water is then pumped through a heat exchanger which transforms the heat to the secondary cooling circuit where the water boils at a much lower pressure and drives the turbine, which, through electric generators, produces electricity.

A BWR has a much more simple construction. Heat from the nuclear fuel in the reactor core is directly transformed to the water surrounding it, causing the water to boils. The steam is then the lead through the turbines, which in the same manner as in a PWR produce electricity.

3.2 Nuclear fuel

The mechanical design of LWR nuclear fuel consists of uranium dioxide in a ceramic form which is pressed to pellets. The pellets are put in tubes of a Zr alloy, which are filled up with helium gas. The rods are then sealed and joined together with spacers to form a fuel bundle. A fuel bundle consists of 64-100 rods. A PWR fuel bundle consists of a lattice of 16x16, 17x17 or 18x18 fuel rods. Fuel bundles designed for use in a BWR are placed in a metal box made of the same alloy as the fuel tubes. The box is necessary for thermohydraulic reasons. Zr-alloy is used as material for fuel tubes and -boxes because Zr has a very low absorption cross-section for neutrons and it is nearly transparent to neutrons. New nuclear fuel consists of a mixture of ^{235}U and ^{238}U. Natural uranium consists of 0.7 % ^{235}U and 99.3 % ^{238}U. Nuclear fuel used in light water reactors is partly enriched in its content of ^{235}U up to approx. 5 %. The nuclear fuel is usually irradiated in the reactor for about 5-8 years. There is no precise limit of when a fuel bundle has reached the end of its life in the reactor. The remaining amount of ^{235}U is only one parameter to take into consideration, but it is usually approx. 1.5-2.0 %.

4 Nuclear Reactor Physics

4.1 Neutron Economy

To make a chain reaction self-sustainable in a reactor it is necessary to have enough neutrons in the reactor. When a thermal neutron is absorbed by a ^{235}U nucleus it may become unstable and fission occurs and on average 2.4 neutrons are emitted. Those neutrons are called prompt neutrons. There are fission products which may decay by emitting neutrons. Those neutrons are called delayed neutrons and make it possible to control the reactor.

When a neutron reacts with a fissile nucleus three processes may occur:

- Fission, i.e. the neutron cause fission the nucleus
- Absorption, i.e the neutron is absorbed by the nucleus
- Scattering, i.e. the neutron is scattered by the target nucleus with loss of kinetic energy

If there at any given moment are N_0 neutrons in the reactor and they are considered as the first generation, then in the n:th generation there will be N_n neutrons:

$$N_n = N_0 \cdot k^{n-1}$$

The multiplication factor k is defined as:

$$k = \frac{number\ of\ neutrons\ in\ one\ generation}{number\ of\ neutrons\ in\ the\ preceeding\ generation}$$

To calculate k during the life of a neutron, it must be followed from when it is born in fission to when it has either leaked out of the system or has been absorbed in the fuel or other materials in the reactor core.

4.2 The four factor formula for an infinite reactor

The so called four factor formula is essential for the understanding the function of a nuclear reactor.

An intuitive derivation of the four factor formula can be found in [7]. In an infinite reactor the neutron cycle will start when a thermal neutron is absorbed by a ^{238}U or ^{235}U nucleus. On average η fast neutrons with an average energy of 2 MeV, which is above the fast fission limit, is created. The fast neutrons will be captured in ^{238}U and cause fast fission and produce new fast neutrons. The total number of neutrons slowing down past the fission threshold of ^{238}U per thermal neutron absorbed in uranium would be larger than η.

This number will be denoted $\eta\varepsilon$, where ε is the fast fission factor. ε is defined as the ratio of the total number of neutrons slowing down past the fission threshold in ^{238}U per original fission neutron. When the energy of neutrons decreases below the fission threshold in ^{238}U no further fissions will occur in this isotope. Neutrons continue to slow down approaching thermal energies of the moderator nuclei. Since ^{238}U has a strong ability to absorb neutrons in non-fission capture, in the intermediate energy range due to strong resonances, which are called resonance absorption, there are a large probability that a number of neutrons are removed from the cycle/system without creating new neutrons. Figure 1 describes the fission cross section for fission of ^{235}U by neutrons in the low and intermediate energy region, including the giant resonance peaks.
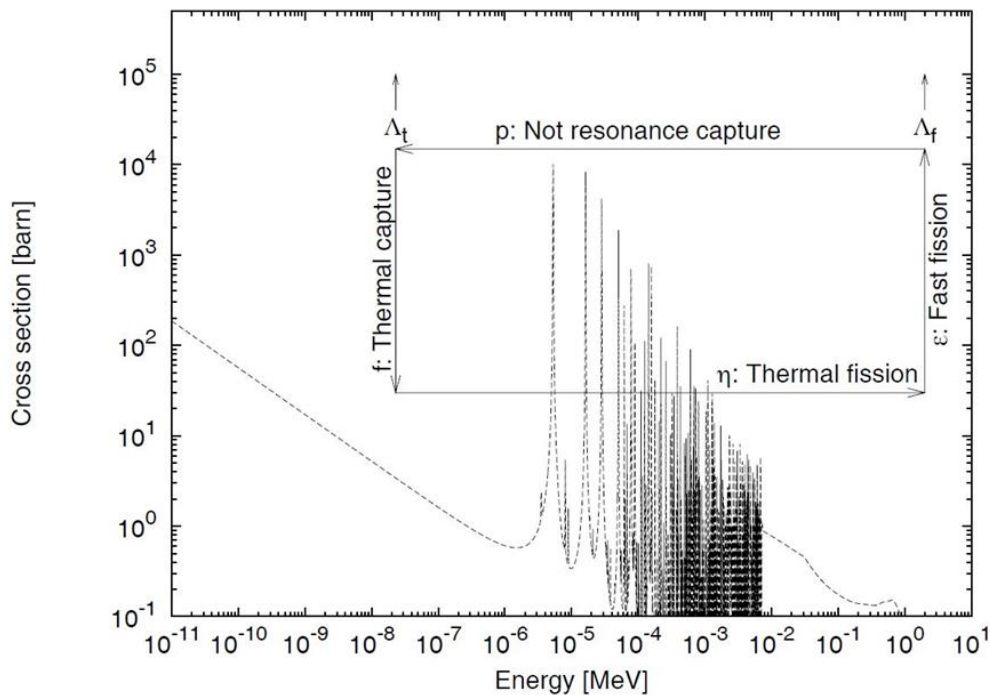
Figure 1. Cross-sections for absorptions of neutrons in ^{238}U from [8]

The probability that a neutron escape being captured by these ^{238}U resonance peaks is denoted p. Thus, the number of neutrons available for being moderated to thermal energies is $\eta\varepsilon p$.

The factor p is called the resonance escape probability and is defined as the ratio of thermal neutrons absorbed in the total system and the total number of neutrons absorbed at all energies below the fast fission threshold. Some of the $\eta\varepsilon p$ thermal neutrons are involved in nuclear reactions with the moderator and other materials. A fraction f that is absorbed in the fuel and creates further fissions is called the thermal utilization factor. f is defined as the ratio of the number of thermal neutrons captured by uranium nuclei to the total number of neutrons

absorbed in the system.

If the neutron cycle begins with a capture of one thermal neutron in the uranium then in the next neutron generation the number of neutrons captured would be: $\eta\varepsilon pf$.

This can also be denoted k_∞ and will be referred to as the "Infinite medium multiplication constant" or k-infinity of a reactor system of infinite size. This finally gives:

$$k_\infty = \eta \cdot \epsilon \cdot p \cdot f$$

This is the four factor formula for an infinite system.

4.3 Finite reactor system - the four factor formula demonstrated

For a finite reactor system, the four factor formula needs to be modified by taking into account the leakage of neutrons. Neutrons can leak out as fast neutrons, during moderation and by diffusion of thermal neutrons. If we denote the probabilities that neutrons will not leak out while being moderated and diffusing, thermal leakage with P_f and P_t, then for a finite system the formula becomes:

$$k = \eta \cdot \epsilon \cdot p \cdot f \cdot P_f \cdot P_t = k_\infty \cdot P_f \cdot P_t$$

With criticality condition, $k=1$ and $P_f P_t < 1$, which means that $\boldsymbol{k_\infty}$ must be larger than 1.

Enrichment

The enrichment of ^{235}U decides how the multiplication constant k varies with burn up of the fuel. As the fuel gets more burnt out the thermal utilization factor, η, decreases as the amount of ^{235}U decrease.

Uranium fuel and moderator

It is most advantageous to arrange the uranium in rods with cladding of small diameter since the absorption in ^{238}U will be at a minimum at the same time as the moderation between the rods is large. By bringing the moderation to a maximum before the neutrons reach the next rod the resonance escape probability, p, is maximized, and better neutron economy is achieved.

Absorbing material

When new fuel is inserted into the reactor for use in a new operating cycle, the reactivity of the fuel is very high. For this reason, a neutron-absorbing material must be put in the core to keep the multiplication constant k at 1. This achieved by control rods and burnable absorbers mixed into the fuel. By having absorbers in the core, the total absorption is increased and the thermal utilization factor f is reduced.

4. Fission products and decay heat

Fission products are important in the scope of this thesis since the energy released by the decay of their nuclei generates heat by β- and γ-emissions. About 10-15 % of the total fission energy is released through these emissions.

The average energy of 200 MeV released by nuclear fission is roughly divided up as follows:
- Kinetic energy from fission fragments: 165 MeV
- Prompt neutrons: 5 MeV
- Prompt γ-rays   7 MeV
- Radioactive decay from fission products   25 MeV

The fragments produced by nuclear fission have an excess of neutrons and deviates heavily from the line of β-stability . The line of β-stability in an isotope chart is the line which describes the set of β-decay stable nuclides. Beta decay stable nuclides are the nuclides which cannot decay through β-emissions.

In order to reach stability, the fission product nuclei have to undergo β^--decay. Every fission product follows a mass chain in order to reach stability and during their journey along the mass chain, two effects can generally be observed. The decay energy decreases and the half-life increases for each step in the chain.

4.1 Fission yields

Mass yield distribution

The mass yield spectra of heavy fissile nuclei are characteristically asymmetric with peaks of distributions separated by approx. 35 – 50 units of mass number *A* (see Figure 2). As a function of the mass number of the fissioning nuclide, the heavier of the asymmetric peaks is nearly stable around 140 and the lighter peak moves in order to conserve total mass.
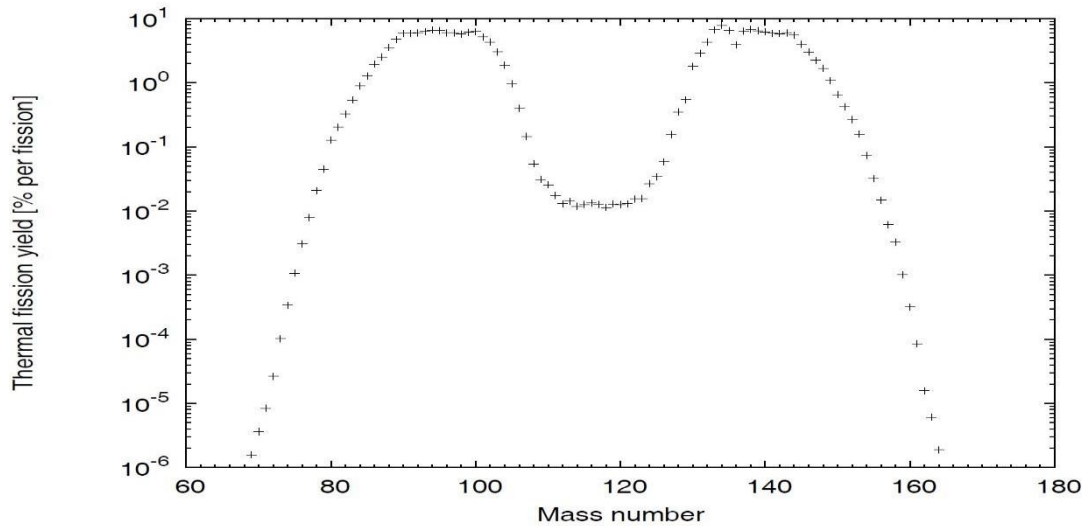
Figure 2. The distribution of fission fragment mass from thermal fission of ^{235}U from [8]

4.2 Decay heat

Decay heat originates mainly from two sources:

- Fission products which decay via β- and γ−emissions

- Actinides which are formed in the reactor during operation and decay via α−,β− and γ−emissions.

Concentrations of fission products as function of decay time are the key data required in aggregate decay heat calculations for designs and operations for SNF management and processing. The relative contributions from the most important isotopes to decay heat is found in Figure 3. Decay heat is important for the final repository for spent nuclear fuel (see chapter 5). The surface temperature must be kept below 100 °C to ensure the integrity of the buffer of bentonite clay in which the canister with fuel bundles is placed [8]. To optimize cost for the repository and safety it is important to have accurate knowledge about how much heat is generated in a fuel canister before encapsulation.

Examples of important fission product isotopes for the production of residual heat in the repository is ^{137}Cs: $(^{137m}Ba+^{137}Cs)$[1], ^{90}Sr: $(^{90}Sr +^{90}Y)$[1]. Among the actinides ^{241}Am is an example of nuclide which is important for generation of residual heat. See table 1. Maximum temperature in the bentonite clay surrounding the SNF canister will be reached after approx. 10 years [**9**]. A ranking of contributions to decay heat from fission products, actinides and activated light elements can be found be found in Table 1 [11].

1: means (mother + daughter nuclide) in the same decay-branch

15
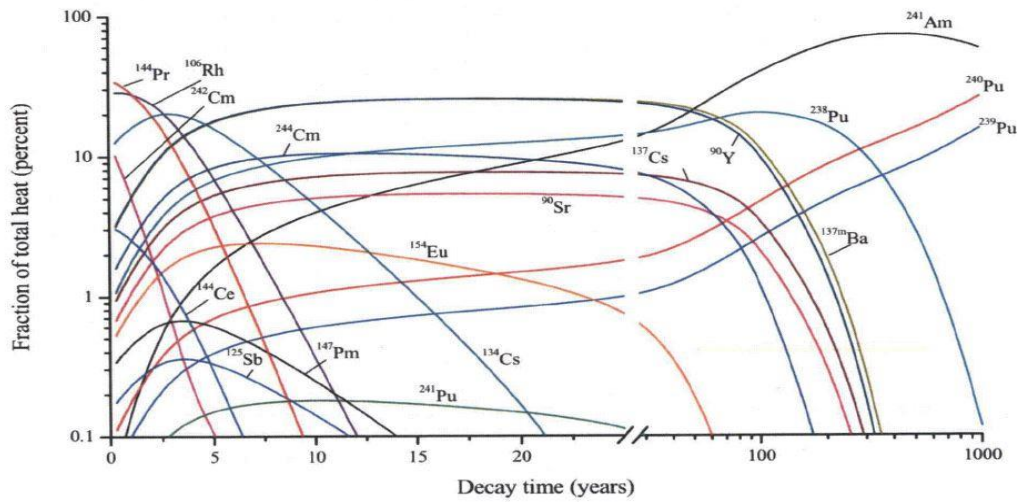
Figure 3. Relative isotopic contributions to the total decay heat for typical high burnup fuel from [10] with permission from I.C. Gauld

Table 1. Decay heat ranking of actinides and fission products and activated light element with greater than 1 % of total decay heat at 5 and 10 000 years from [11]

| Nuclide | 5 years | | 10 000 years | |
	20 GWd/t 3 .0WT% ^{235}U	50 GWd/t 4 .5WT% ^{235}U	20 GWd/t 3 .0WT% ^{235}U	50 GWd/t 4.5WT% ^{235}U
^{238}Pu	1(2)	2(7)	-	-
^{241}Am	2(2)	3(2)	-	-
^{240}Pu	-	-	2(33)	2(41)
^{239}Pu	-	-	1(65)	1(54)
^{244}Cm	-	1(7)	-	-
^{243}Am	-	-	-	3(3)
^{90}Y	1(23)	1(19)	-	-
^{137m}Ba	2(20)	2(18)	-	-
^{134}Cs	3(11)	3(17)	-	-
^{106}Rh	4(10)	4(7)	-	-
^{144}Pr	5(9)	7(4)	-	-
^{137}Cs	6(6)	5(5)	-	-
^{90}Sr	7(5)	6(4)	-	-
^{60}Co	8(5)	8(3)	-	-
^{154}Eu	9(1)	9(2)	-	-

GWd/t U: Giga Watt Days per tonnes U

16

5. Final repository for spent fuel

The method for final repository in Sweden

A major concern about the final nuclear waste repository is to keep its integrity intact over a very long period of time to prevent leakage of radioactivity out of the repository. The final repository for spent nuclear fuel, which is going to be used in Sweden, is designed with a method called KBS-3 (see Figure 4). It has its foundation on three barriers: copper canisters, bentonite clay and the granite rock in Sweden. Before the SNF is placed in the final repository it is kept in the central intermediate storage facility, CLAB, in Oskarshamn in Sweden. During the storage in CLAB the radioactivity is decreasing and the SNF becomes less dangerous and easier to handle. However, it still contains large amounts of radioactive substances with very long half-lives and needs to be kept safe for at least 100 000 years. SKB, Svensk Kärnbränslehantering, has plans to build a final repository in Forsmark, Sweden, for SNF. The method is built on encapsulation of the spent fuel in copper canisters which are put in a tunnel system in the granite rock at a depth of 500 m where the canisters are embedded in bentonite clay.
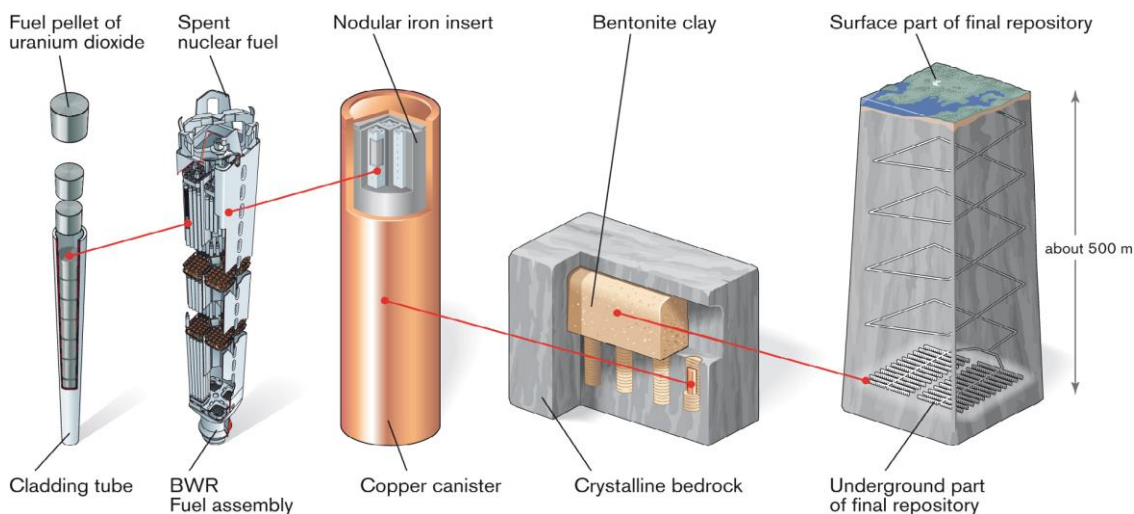
Figure 4: The final repository for spent nuclear fuel in Sweden according to KBS-3 method with permission from SKB

The first barrier - the copper canister

The copper canisters will have an inside of soft iron around the SNF bundles. The canisters will have a length of 5 m and a single canister will have a weight of approximately 25 ton. The outer part of the canister will be a 5 cm thick layer of copper. The canister is constructed to resist corrosion and the mechanical strain caused by movements in the rock.

The second barrier - the bentonite clay

The copper canister will be put into a drilled hole in shafts in the bedrock. The canisters will be embedded in bentonite clay in the tunnels in the repository. The clay protects the canister and works as a buffer if the canister is broken and radioactivity leaks out and against corrosion and minor movements in the granite rock. When all canisters in a tunnel are in place, the tunnels will be filled and sealed. The natural water in the rock will return into the material which will surround the canister including the bentonite clay. The clay buffer will the swell and fill up holes and cracks in the rock surrounding the hole in which the canister is placed. Should the bentonite clay dry-out, the ability to prevent leakage would be reduced.

The third barrier - the bedrock

The final barrier is the bedrock itself. The bedrock separates the SNF from the world above ground. It is a stable chemical environment and the depth itself protects the repository from activities on the ground. As mentioned above, there is water in the bedrock and any radioactive particles leaking from the SNF will, during the transport through the bedrock, be stuck on surfaces of cracks and micro pores in the bedrock. In this way the bedrock and the large depths will contribute to keep the SNF safe and distant from the environment at the surface.

6. Method

The method used in this thesis is to perform a review of relevant studies about validations of calculated values of isotopic compositions in spent nuclear fuel. A structured way of how to make reviews in the scientific field can be found in [2]. This handbook describes a method of how to make reviews in the field of medical science. Inspired by this handbook a simplified process has been used.

6.1 Process of the review

1. Research question
   a. Formulate the research question for the review in a structured way
   b. Establish inclusion- and exclusion criteria

2. Choice of literature
   a. Searching for published studies in any available sources, databases, the internet etc
   b. Performing a course search among abstracts
   c. Reading relevant articles and reports
   d. Deciding of a read article or report is inside or outside of the established criteria

3. Assessment of chosen studies
   a. Grading of relevance
   b. Picking out studies for detailed analysis

The process applied to this review**:**

Research question for this review

The research question for this review is to find published studies of validations of calculated isotopic compositions in spent nuclear fuel by measurements. In addition, there is a detailed question of finding validated studies covering isotopes specifically interesting for the generation of residual heat in the spent fuel canister.

 Inclusion and exclusion criteria
The inclusion criteria selected for the thesis:

- Studies should be written in English
- Studies should be accessible through databases or the internet
- A study should contain information about experimental validations of calculations made by nuclear simulation software on spent nuclear fuel. Both BWR and PWR should be included in the search.

Exclusion criteria is:

- Studies on non-LWR nuclear fuel should not be treated.

Assessment of found studies - Grading of relevance

Grading of relevance is made in four categories:

1. **Not relevant**
   Grading for found studies which are included but not relevant

2. **Useful**
   Grading for found studies which is not relevant but can be useful in another way

3. **Partly relevant**
   Grading for found studies which is partly relevant but do not completely answer the research question

4. **Relevant**
   Grading for found studies which contributes to completely or almost answer the research question

Detailed analysis

Requirements for selecting a study for detailed analysis depends of details in research question.

Delimitations

The search for studies has been done by: search on the internet by Google and Google scholar, assessment of reference lists, personal discussions with experienced scientist in the field.

7. Result - review

The found studies were listed with a grading of relevance according to interpretations of the search criteria during assessment of the study. If a report has been selected for detailed analysis it is also reported. All studies were found using Google and Google scholar between September 2018 and March 2019 and the compilation of studies does not claim to be comprehensive. There may very well exist more studies which have been missed or could have been found by other keywords. All literature found here can be found in the reference list with numbering:

List of found studies

1. Gauld, I C., Ilas, C. and Radulescu, G: Uncertainties in Predicted Isotopic Compositions for Uncertainties in Predicted Isotopic Compositions for High Burnup PWR Spent Nuclear Fuel NUREG/CR-7013, NUREG/CR-7012 ORNL/TM-2010/41 [10]
Grading of relevance: Relevant
Chosen for detailed analysis: No

Abstract: This study covers validation of calculated predictions of isotopic compositions from 6 PWR:s in USA, Japan, Switzerland, Germany and Spain. In total 51 samples were radio chemically analysed by 7 laboratories in Japan, USA, Russia, Belgium and Switzerland. Calculations were made with SCALE 5 code and cross sections were based on data from ENDF/B-V libraries.

2. Radulescu,G., Gauld, I C.,llas G.: :An Approach for Validating Actinide and Fission Product Burnup Credit Criticality Safety Analyses—Isotopic Composition: (April 2012), TN 37831-6170 NUREG/CR-7108 ORNL/TM-2011/509. [12]
Grading of relevance: Relevant
Chosen for detailed analysis: No
Abstract: This report describes an approach for finding depletion code biases uncertainty in reactivity calculation based on comparison between measured and calculated isotopic concentrations. SCALE 6.1 computer code and ENDF/B-VII library data were used. In total 28 actinides and fission products were analysed.

3. Radulescu, G., Gauld I C., Ilas G.: SCALE 5.1 Predictions of PWR Spent Nuclear Fuel Isotopic Compositions (March 2010) ORNL/TM-2010/44 [13]
Grading of relevance: Relevant
Chosen for detailed analysis: No
Abstract: This calculation report documents validation comparisons between calculated and measured isotopic compositions from 9 PWRs in Germany, USA, Japan and Switzerland. The radiochemical analyses were performed at 8 different laboratories in Germany, Belgium, Russia, Japan, USA and Switzerland. Calculations were done using SCALE 5.1 and TRITON/NEWT depletion model

4. O. W. Hermann M. D. DeHart: Validation of SCALE (SAS2H) Isotopic Predictions
for BWR Spent FuelORNL/TM-13315 [14]
Grading of relevance: Relevant
Chosen for detailed analysis: Yes

Abstract: This report treats comparisons between calculations and experimental measurements from 30 samples taken on SNF from the three BWR:s Cooper Nuclear Power Plant (USA), Grundremmingen Nuclear Power Plant (Germany) and the Japan Power Demonstration Reactor(Japan). Radiochemical analysis was made at PNNL, European Joint Research Center: in Karlsruhe and Ispra (Italy) and Japanese Atomic Energy Research Institute (JAERI). Calculations were made by version 4.3 of SCALE system and fuel depletion was performed by ORIGEN-S. Cross sections were based on data from ENDF/B-V and ENDF/B VI libraries.

 5. Ilas, G., Gauld I C., Liljenfeldt, H.: Validation of ORIGEN for LWR used fuel decay heat
analysis with SCALE, Nuclear engineering and design **273**, 58-67, (2014) [15]
Grading of relevance:
Chosen for detailed analysis: No

Abstract: Article from Nuclear Engineering and Design treats validation of ORIGEN by fuel heat decay analysis made by calorimetric measurements on SNF from four Swedish BWR and PWR reactors. The measurements were performed in CLAB in Sweden and the calculation was computed on a SCALE 6.1.2 system and used ENDF/B-VII nuclear data.

6 Gauld I C, Mertyurek U, Smith HJ.:
Analysis of Experimental Data for High Burnup BWR Spent Fuel Isotopic Validation –
SVEA-96 and GE14 Assembly Design. NUREG/CR-7162, ORNL/TM-2013/18 [17]
Grading of relevance: Relevant
Chosen for detailed analysis: No
Abstract: This report treats experimental measurements made on two sorts of BWR-fuel with different kind of mechanical design, SVEA-96 and GE14. The measurements where part of the MALIBU and ENUSA programs. Samples were taken from three different reactors in Sweden, Spain and Switzerland and analysed at three different laboratories: Studsvik Nuclear in Sweden, SCK-CEN in Belgium and Paul Scherer Institute in Switzerland. Calculations were made with TRITON, 2D depletion sequence in the SCALE 6.1 system. The isotopic composition was calculated with ORIGEN. All calculations used nuclear data from ENDF/B-VII 238 group cross-section library. Calculations for more than 60 isotopes was compared to experimental values.

7. Gauld, I C, Pigni,M T., Ilas, G.: Validation and Testing of ENDF/B-VII Decay Data, Nuclear Data Sheets **120**, (January 2014) [18]

Grading of relevancy: Useful

Chosen for detailed analysis: No

Abstract: This article in Nuclear Data Sheets is interesting since it covers validation of ENDF/B-VII by using ORIGEN computer code and comparisons with both measurements of decay heat on SNF by calorimeter and radiochemical measurements of isotopic compositions. The validation revealed significant errors in decay scheme for both actinides and fission products.

8. Ilias, G.,Gauld, I C.: Analysis of Experimental Data for High-Burnup PWR Spent Fuel Isotopic Validation-Vandellos II Reactor NUREG/CR-7013 ORNLITM-2009/321 [19]

Grading of relevance: Relevant

Chosen for detailed analysis: No

Abstract: In this report radiochemical measurements were performed by Studsvik laboratory on 6 samples taken from SNF from Vandellos II PWR in Spain. The measurements were used for validation of Triton depletion sequence in SCALE code system and used cross section data from ENDF/B-V library.

9. DeHart, M D.,Hermann, O W.: An Extension of the Validation of SCALE (SAS2H) Isotopic Predictions for PWR Spent Fuel ORNL/TM-13317 [20]

Grading of relevance: Relevant

Chosen for detailed analysis: Yes

Abstract: This report describes validation of computer code by comparison with experimental measurements with fuel samples from two PWR:s I USA and Italy. The report focuses on development of assembly specific depletion models. The measurements were performed by two laboratories, Ispra in Italy and European Joint Research Center in Germany. The calculations were produced by using the SAS2H control module in version 4.2 of SCALE. Cross sections were calculated with data from ENDF/B-V and ENDF/B-VI libraries. In addition, an approach to determine biases and uncertainties between calculated and measured isotopic compositions was made.

10. Mertyurek,U, Gauld, I C.: Development of ORIGEN libraries for mixed oxide (MOX) fuel assembly designs Nuclear Engineering and design **297**, 220-230(2016) [21]

Grading of relevance: Partly Relevant

Chosen for detailed analysis: No

Abstract: This study is interesting since it treats validation of SCALE computer calculations by destructive radiochemical measurements on MOX fuel. MOX fuel consist of fissile isotopes of both plutonium and uranium in the form of plutonium and uranium oxide. The SNF came from both BWR and PWR. Calculations were performed by ORIGEN code which is part of the SCALE system and nuclear data were from the ENDF/B-VII library.

11. Hermann, O W, Bowman, S M. Brady, M C., Parks, C V.: Validation of the Scale System for PWR Spent Fuel Isotopic Composition Analyses ORNL/TM-12667 (1995) **[22]**
Grading of relevancy: Partly relevant
Chosen for detailed analysis: No
Abstract: In this report 19 samples from 3 PWR reactors in Germany and USA were analysed and the isotopic compositions were measured and used for validation of the SAS2H control module in SCALE 4.2. Nuclear data was from ENDF/B-IV and ENDF/B-V

12. Ilias, G.,Gauld, I C., Radulescu, G.: Validation of new depletion capabilities and ENDF/B-VII data libraries in SCALE, Annals of nuclear energy **46** 43-55 (2012) **[23]**
Grading of relevance: Useful
Chosen for detailed analysis: No
Abstract: In this article from Annals of Nuclear Energy describes validating code performance after implementation of SCALE 6.1 and new ENDF/B-VII data. The validation has been made from existing experimental measurements from 9 different reactors in Italy, USA, Germany, Japan and Switzerland. The results has been compared to earlier validations which has used ENDF/B-V.

 13. DeHart, M. D.: Stochastic Method for Estimating the Effect of Isotopic Uncertainties in Spent Nuclear Fuel, ORNL/TM-2001/83 **[24]**
Grading of relevance: Partly relevant
Chosen for detailed analysis: No
Abstract: This study shows a new approach for estimating the uncertainty in predictions of neutron multiplication factor, k_{eff}, for SNF. The calculation of k_{eff} is an important step when SNF burnup calculated. When computing k_{eff} , isotopic compositions are relevant. For this reason, the report is interesting when estimating uncertainties in calculation of isotopic compositions.

14. Ilias, G., Liljenfeldt,: Decay heat uncertainty for BWR used fuel due to modelling and nuclear data uncertainties: Nuclear Engineering and Design Volume **319**, 1 August 2017, Pages 176-184**[25]**
Grading of relevance: Partly relevant
Chosen for detailed analysis: No
Abstract: This study is interesting because the validation of predicting isotopic compositions in SNF by ORIGEN and TRITON in the SCALE 6.2.1 system is done by comparing calculations with experimental data from the SFCOMPO database. An assessment of the effect of new developments on code performance was also made. Nuclear data from ENDF/B-VII library is used for the evaluation.

15. Francis, W. F., Weber, C.F., Pigni, M. T., Gauld, I.C.: Reactor fuel Isotopic and Code Validation for Nuclear Applications: ORNL/TM-2014/464 **[26]**

Grading of relevance: Useful

Chosen for detailed analysis: No

Abstract: In this study databases with experimentally measured isotopic concentrations of SNF with well-known characteristics are used to validate accuracy of depletion code. The most important database for this purpose is SFCOMPO. All calculations were made with ORIGEN depletion code from ORNL. The study is an extensive compilation of data regarding depletion modelling and can be used with the purpose to perform more in depth studies of calculation modelling and accuracy.

8. Results and discussion - selected reports

A method accepted by the nuclear engineering community, for use in depletion code validations, is founded on comparing computed isotope concentrations with measured isotope concentrations from destructive radiochemical analyses of fuel sample [**12**].

Uncertainties

Calculations of fission product concentrations have uncertainties due to large uncertainties in fission product cross section data [**10**]. There exist several cross-section libraries for neutron energies to be used in reactor physics calculations. The major difference between them is the number of neutron energy groups available in the library.

Calculated uncertainties can be arranged in three groups according to [**10**]:

1. Uncertainty in the actual system parameters being modelled (e.g. power history, initial fuel compositions, unknown reactor conditions, value of the fuel discharge burnup etc)

2. Uncertainty due to the approximations been made when the model was setup (geometry approximations, simplification of power history etc.)

3. Uncertainty coupled to calculation procedures and data used in the calculations.

Usually, measurement uncertainties for radiochemical analysis are dependent on the measurement instruments and radiochemical procedures, and can be from <1% for the important uranium and plutonium nuclides when using modern radiochemical analysis methods, to more than 5% when using less accurate methods [**12**].

8.1 Selection of reports

Two reports were selected for analysis regarding uncertainties in code results. A comparison for three isotopes important for the generation of residual heat in SNF in the final repository, have been chosen: ^{137}Cs, ^{90}Sr and ^{241}Am.

To be selected a study should fulfill at least one of the following criteria:

* ^{137}Cs, ^{90}Sr and ^{241}Am, should be treated in the same report

*   Decay heat calculation via isotopic calculations validated to direct measurements of decay heat.

It has been particularly hard to find reports presenting validation data for ^{90}Sr. A not completely satisfying fact is the difference in origin of the SNF regarding in which reactor type it has been irradiated. The difference in thermohydraulic operating conditions for PWRs and BWRs makes it hard to compare because of the more simple reactor physics model in PWRs. Selected reports are:

- O. W. Hermann M. D. DeHart: Validation of SCALE (SAS2H) Isotopic Predictions for BWR Spent Fuel ORNL/TM-13315, [14]

- Ilias, G.,Gauld, I C., Liljenfeldt, H.: Validation of Origen for LWR used fuel decay heat analysis with SCALE, Nuclear Engineering and design 273, 58-67(2014) [15]

8.2 Selected report – Validation of SCALE (SAS2H) Isotopic Predictions for BWR Spent Fuel

In [**14**] the ORIGEN-S code (part of the SA2H code package) has - from known parameters regarding the physical history in the reactor - been used for calculating isotopic compositions for a large number of fission products and actinides.

Preconditions in the report

30 fuel samples from six separate fuel bundles from three different BWRs were analyzed: Cooper Nuclear Power Plant (USA), Grundremmingen Nuclear Power Plant (Germany) and the Japan Power Demonstration Reactor (JPDR). Radiochemical analysis of six Cooper fuel samples were conducted by Material Characteristics Center (MCC) at Pacific Northwest laboratory (PNL). The eight Grundremmingen fuel samples were analyzed by European Joint Research Center (JRC). 16 fuel samples were analyzed by the Japanese Atomic Energy Research Institute (JAERI). The calculated isotopic compositions were performed using the SAS2H control module in the SCALE code system version 4.3. This control module produces burnup dependent cross sections using a neutronics model in SCALE and the fuel depletion analysis of the ORIGEN-S code. The cross section library used in these analyses contains 44 energy groups and was based on Evaluated Nuclear Data Files (ENDF). ENDF/B – V was used for all calculations except for 3 nuclides: ^{16}O, ^{154}Eu and ^{155}Eu. For those nuclides cross section data were used from ENDF/B-VI.

Results from comparisons between calculations and measurements

In this report three isotopes, ^{137}Cs, ^{90}Sr and ^{241}Am, were selected for evaluation. The nuclides were chosen because of their significance for residual heat generation in the waste repository in a medium time perspective, 1-10 years.

The result from comparisons between measurements and calculations can be seen in table 2 and specifically for ^{137}Cs, ^{90}Sr and ^{241}Am for all sites in table 3. Figure 4 shows percentage difference between measured and calculated results for all fuel samples.

Table 2: Summary of percentage differences, $\delta=$ (calculated/measured$-1)\cdot100\%$, between measured and computed isotopic composition in [**14**] as average and spreads for all validated isotopic compositions. All calculated data computed using the ENDF 44 group library

Nr:	Nuclide	No of cases	Average δ (%)	Max δ (%)	Min δ (%)	Standard deviation σ (%)
1	^{79}Se	6	23.6	28.1	20.8	2.6
2	^{90}Sr	6	8.9	10.8	5.9	1.9
3	^{99}Tc	6	12.1	14.6	7.0	3.1
4	^{106}Ru	12	47.8	61.9	37.6	8.4
5	^{126}Sn	6	205.2	218.4	195.4	8.2
6	^{134}Cs	20	0.9	18.5	-23.4	11.4
7	^{135}Cs	6	5.1	10.4	-6.5	6.2
8	^{137}Cs	30	1.5	17.3	-13.1	5.9
9	^{144}Ce	12	95.5	114.7	78.8	10.9
10	^{143}Nd	16	0.4	1.8	-0.6	0.6
11	^{144}Nd	16	-0.4	0.5	-1.7	0.7
12	^{145}Nd	24	0.4	1.6	-0.4	0.5
13	^{146}Nd	16	0.2	1.4	-0.3	0.5
14	^{148}Nd	20	-0.5	1.5	-4.4	1.3
15	^{150}Nd	6	-0.3	1.5	-1.6	0.9
16	^{154}Eu	22	-7.8	6.9	-32.3	11.0
17	^{155}Eu	30	-42.6	-39.3	-44.4	1.8
18	^{234}U	30	-0.2	4.4	-6.1	2.6
19	^{235}U	30	-2.0	4.0	-11.7	3.3
20	^{236}U	18	-1.2	4.5	-7.8	2.7
21	^{238}U	30	-0.1	0.2	-1.5	0.4
22	^{237}Np	18	-1.1	17.3	-11.2	8.7
23	^{238}Pu	30	-7.0	45.3	-27.3	16.3
24	^{239}Pu	30	-2.1	8.8	-17.3	6.0
25	^{240}Pu	30	-0.9	6.9	-11.1	4.8
26	^{241}Pu	30	-4.5	17.1	-20.1	9.3
27	^{242}Pu	30	0.5	42.9	-17.5	12.6
28	^{241}Am	22	4.1	28.4	-11.1	11.5
29	^{242m}Am	12	2.3	88.1	-37.5	34.3
30	^{242}Cm	16	13.5	46.3	-9.0	17.5
31	^{242}Cm	6	13.5	41.4	1.0	19.4
32	^{244}Cm	24	-19.4	16.3	-57.5	15.5

Table 3: Summary of percentage differences between measured and computed isotopic compositions for each site Ref. [3].

Nuclide	Site	No of samples	Average δ (%)	Standard deviation σ (%)
^{241}Am:	Cooper:	6	16.3	8.4
	JDPR:	16	-0.6	8.8
^{90}Sr:	Cooper	6	8.9	1.9
^{137}Cs:	Cooper	6	2.3	4.1
	JDPR	16	2.3	4.3
	Grundrem.	8	-0.5	9.3

There are several possible sources for the large discrepancies between measured and calculated results. First the physical model is much more complex in a BWR relative a PWR. For example, heterogeneous materials in the core, more complex neutron spectrum and more variable axial power profile. Secondly the total number of measurements made for the purpose of validation are far too few.

For the total average differences, the following observation can be made:

- ^{137}Cs: δ=1.5% σ=5.9%. The results have fairly large σ. If the results for each site are studied, consistent results for Cooper and JDPR can be observed but for Grundremmingen the standard deviation is significantly higher.
- ^{90}Sr: δ=8.9%, σ=1.9%. Larger average but smaller standard deviation but too few measurements (N=6) were made for drawing more far-reaching conclusions.
- ^{241}Am: δ=4.1%, σ=11.5%. Less good results if results from each reactor is taken into account. Probable causes are different measurement techniques, different reactor and fuel models.
- The large uncertainty indicates the need for more measurements and comparisons especially if uncertainty on a level of 1-2 % is required.

The following observations can be made if detailed results are taken into account:

- The number of measurements being made in this study is low. This should be an exclusion criteria in the review.
- Significant differences between results from different reactors.
- The calculated values should be presented in the report. This is usually not the case in any report used in the review.
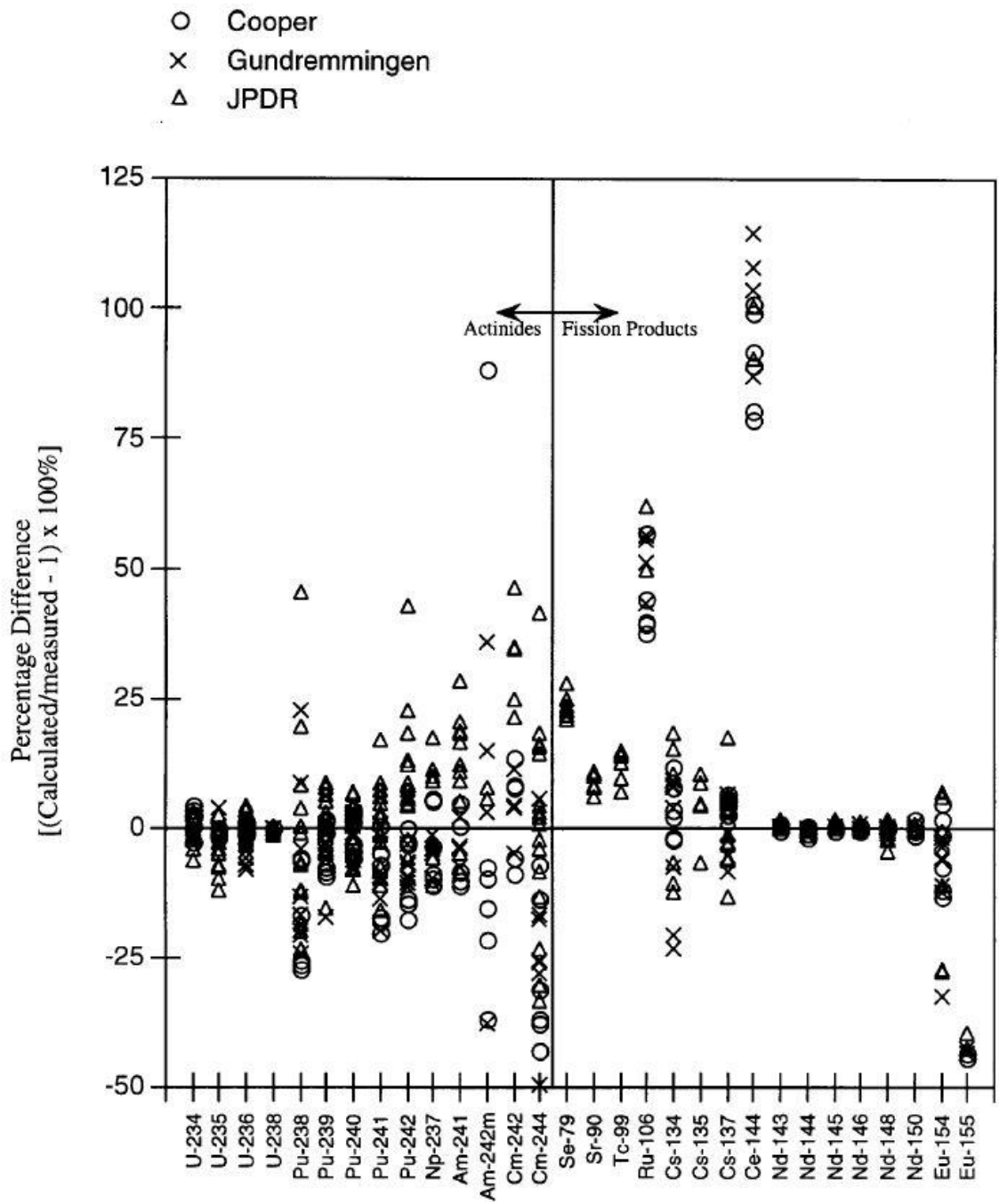
Figure 5. Percentage difference between measured and calculated results for all fuel samples, from [**14**], reproduced with permission from ORNL.

8.3 Selected report – Validation of Origen for LWR used fuel decay heat analysis with SCALE

This article from Nuclear Engineering and Design is particularly interesting since direct measurements of decay heat has been made and compared with decay heat calculations using ORIGEN. SCALE 5.1 with decay module ORIGEN and with ENDF/B-V data had been used for the calculations of isotopic compositions and decay heat. The decay heat measurements used for validations in this study were made at CLAB in Sweden 2003 and 2010. The experimental uncertainties in the study are, at 95 % confidence level depending of the assembly decay heat, reported as:

PWR assembly: $\pm$ 9.2 W (3.7 %) at 250 W
                     $\pm$18.8 W (2.1%) at 900 W
BWR assembly: $\pm$4.2 W (8.4 %) at 50 W
                     $\pm$6.2 W (1.8%) at 350 W

Table 4 shows a summary of the measurements and table 5 shows the results from comparisons between calculated and measured decay heat. All information in tables are from [**15**].

In this study SCALE 6.1.2 with decay module ORIGEN was validated against direct heat calorimetric measurements in CLAB. The data which was used in the calculation was in turn validated against measurements in CLAB 2003 using SCALE 5.1 and ENDF/B-V data [16].

The results reported give an average of calculated to experimental decay heat ratio of 1.002 (σ=0.012) for PWR and 0.997 (σ=0.024) for BWR. The mean value for the difference between calculated and measured values was 0.6 W (σ=4.9) for PWR and -0.3 W (σ=3.4) for BWR. The accuracy needed for decay heat matters is in the order of a few percent relative uncertainty) [**27**], implying that further investigations are needed in order to understand (and mitigate) the differences between calculations and measurements.

Table 4: Summary of measurements in study [**15**]

Reactor type	Reactor	Mechanical Design(pin arrangement)	Enrichment (wt % ^{235}U)	Burnup (GWd/MTU)	No of ass. Measured	No of Measurements
PWR	Ringhals 2	15×15	3.095–3.252	50.96	18	33
PWR	Ringhals 3	17×17	2.100–3.404	47.31	16	38
BWR	Ringhals 1	8×8	2.258–2,911	44.86	17	45
BWR	Oskarshamn2	8×8	2,201	24.47	5	5

Table 5: Summary of results: calculation–experiment comparison

Data set	No.of meas	C/E		R (W)	
		Mean	σ	Mean	σ
PWR	71	1.002	0.012	0.57	4.91
BWR	50	0.997	0.024	−0.25	3.36
PWR+BWR	121	1.000	0.017	0.23	4.34

C/E: Calculated to experimental decay heat ratio
R(W): Decay heat residual (difference between calculated and experimental decay heat)

The study has tested potential bias in reactor specific physical calculation model, see table 6. No significant bias was found in this test.

Table 6: Results by reactor calculation-experiment comparison for test of biases

Reactor type	Reactor	No of measurements	C/E		R	
			Mean	σ	Mean(w)	σ(w)
PWR	Ringhals 2	33	0.998	0.012	-0.96	5.11
PWR	Ringhals 3	38	1.005	0.011	1.89	4.36
BWR	Ringhals 1	45	0.999	0.024	-0.02	3.43
BWR	Oskarshamn2	5	0.975	0.020	-2.35	1.66

9. Conclusions

In this project a review has been performed with the purpose to find studies covering validations of calculations by experimental measurements. There exist a significant amount of data based on measurements of actinides but results for fission products are much more limited and the shortage of representative data limits the ability to directly validate computer code calculations and to quantify the uncertainties in calculated nuclide compositions.

Review

In total 15 relevant reports were found, but it might very well be possible to find more relevant studies if other ways of searching or other keywords had been used. It has been found that [1] can be useful in the basic education of physics as a foundation for methods for search of materials for reviews. The working process in how to perform a review in Ref. [1] can be applied in the field of physics. The observation from the analysis of Ref. [14], Validation of SCALE (SAS2H) Isotopic Predictions for BWR Spent Fuel, suggests that the number of measurements in the study should be a criterion when performing a review. Calculated values are not presented in any report used in the review, only differences to measured values, which delimits the values of the validation when estimating the quality.

The evaluation of selected reports Validation of SCALE (SAS2H) Isotopic Predictions for BWR Spent Fuel [14]

A general observation is that there are particularly large discrepancies between calculated and measured isotopic compositions for a number of nuclides. Those uncertainties are of such magnitude that it is questionable to draw conclusions from calculations of these nuclides. Examples of such nuclides are ^{144}Ce and ^{106}Ru and ^{244}Cm, see Figure 4 and table 3. On the other hand, there are a number of isotopes with small discrepancies eg.: ^{238}U, ^{242}Pu and ^{146}Nd. If calculation of isotopic compositions is going to be used for design purposes regarding residual heat, an uncertainty in the order of 1-2% is required [28] and with this in mind, isotopes relevant for decay heat the discrepancies is larger, which delimits the use of the validation.

Validation of Origen for LWR used fuel decay heat analysis with SCALE [15]
The results show, on average, good agreement between experimental and calculated data albeit with absolute uncertainties larger than the average difference. Calorimetric measurements are less elaborate to perform than radiochemical measurements and are an interesting alternative way of validating calculated values for residual heat.

10. Outlook

A method similar to [2] could be developed for use in the basic education in Physics. In Ref. [11] there are several proposals for future research areas but this work was made in 2001 and it is unclear how obsolete it is. An interesting approach would be to do a study about how relevant these proposals are today and suggest new issues to investigate. Another observation from the studies that have been found, is how few scientists and laboratories there are that do research in this important field. Because every country which have or have had a nuclear program, has an interest in research about safe handling of spent nuclear fuel. In 2019 an EU-funded program, EURAD, will most probably be initiated with the goal to gather more experimental data ref [29].

Many studies use the same measurements of isotopic compositions for validation purpose. For this reason, there should be made more measurements of isotopic compositions on spent nuclear fuel but there might exist measurements of isotopic compositions which are not available in open sources. For the measurements that have been made there exist more data about actinides than fission products. The absence of a physical standard for how to calibrate measurement techniques and instruments for SNF isotopic measurements is not satisfying because comparing measurements are difficult without a standard. This should be a goal for future work in the field.

This project has only made a first treatment of this subject so there might be a potential for a study more in depth in e.g. a Master thesis.

References

1. SCALE: https //www.ornl.gov/scale
2. Statens Beredning för medicinsk och social utvärdering: Utvärdering av metoder i hälso- och sjukvården och insatser i socialtjänsten en handbok.
3. Meitner, L.,Frisch,O.: Disintegration of uranium by neutrons: a new type of nuclear reaction Nature **143**, 239-240(1939)
4. Bohr, N .: Resonance in uranium and thorium disintegrations and the phenomenon of nuclear fission. Phys. Rev. **55**,418-419(1939)
5. Nier, A O., Booth, E T.,Dunning, J R.,Grosse, AV.: Nuclear fission of separated uranium isotopes. Phys. Rev **57**, 546(1940)
6. Bohr, N, Wheeler, J A.: The mechanism of nuclear fission. Phys Rev. **56**, 426-450(1939)
7. Garg,S,Ahmed,F.Kothari L S.: Physics of nuclear reactors(1987)
8. Jansson P. Studies of Nuclear fuel by means of Nuclear Spectroscopic Methods, Uppsala (2002)
9. Ikonen, K., Raiko , H.: Thermal Dimensioning of Olkiluoto Repository for Spent Fuel, working report 2012-56
10. Gauld, I C., Ilas, C. and Radulescu, G: Uncertainties in Predicted Isotopic Compositions for Uncertainties in Predicted Isotopic Compositions for High Burnup PWR Spent Nuclear Fuel NUREG/CR-7013, NUREG/CR-7012 ORNL/TM-2010/41
11. Gauld, CV.,Parks, C V.: Review of Technical Issues Related to Predicting Isotopic Compositions and Source Terms for High Burnup LWR Fuel, NUREG/CR-6701, ORNL/TM-2000/277
12. Radulescu,G., Gauld, I C.,llas G.: :An Approach for Validating Actinide and Fission Product Burnup Credit Criticality Safety Analyses—Isotopic Composition : (April 2012) ,TN 37831-6170 NUREG/CR-7108 ORNL/TM-2011/509
13. Radulescu, G.,Gauld I C., Ilas G.: SCALE 5.1 Predictions of PWR Spent Nuclear Fuel Isotopic Compositions (March 2010) ORNL/TM-2010/44
14. O. W. Hermann M. D. DeHart :Validation of SCALE (SAS2H) Isotopic Predictions for BWR Spent Fuel ORNL/TM-13315
15. Ilias, G.,Gauld, I C., Liljenfeldt, H.: Validation of Origen for LWR used fuel decay heat analysis with SCALE, Nuclear Engineering and design 273, 58-67(2014)
16. Ilias, G.,Gauld, I C.:SCALE analysis of CLAB decay heat measurements for LWR spent fuel, Annals of Nuclear Eenergy **35** , 37-48, (2008)
17. Gauld I C, Mertyurek U, Smith HJ.: Analysis of Experimental Data for High Burnup BWR Spent Fuel Isotopic Validation – SVEA-96 and GE14 Assembly Design. NUREG/CR-7162,ORNL/TM-2013/18
18. Gauld, I C, Pigni,M T., Ilas, G.: Validation and Testing of ENDF/B-VII Decay Data, Nuclear Data Sheets **120**, (January 2014)

19. Ilias, G.,Gauld, I C.: Analysis of Experimental Data for High-Burnup PWR Spent Fuel Isotopic Validation-Vandellos II Reactor NUREG/CR-7013 ORNLITM-2009/321

20. DeHart, M D.,Hermann, O W.: An Extension of the Validation of SCALE (SAS2H) Isotopic Predictions for PWR Spent Fuel ORNL/TM-13317

21. Mertyurek,U, Gauld, I C.: Development of ORIGEN libraries for mixed oxide (MOX) fuel assembly designs Nuclear Engineering and design **297**, 220-230(2016)

22. Hermann, O W, Bowman, S M. Brady, M C., Parks, C V.: Validation of the Scale System for PWR Spent Fuel Isotopic Composition Analyses ORNL/TM-12667 (1995)

23. Ilias, G.,Gauld, I C., Radulescu, G.: Validation of new depletion capabilities and ENDF/B-VII data libraries in SCALE, Annals of nuclear energy **46** 43-55 (2012)

24. DeHart, M D .:Stochastic Method for Estimating the Effect of Isotopic Uncertainties in Spent Nuclear Fuel, ORNL/TM-2001/83

25. Ilias, G., Liljenfeldt,: Decay heat uncertainty for BWR used fuel due to modeling and nuclear data uncertainties: Nuclear Engineering and Design Volume 319, 1 August 2017, Pages 176-184

26. Francis, W. F., Weber, C.F., Pigni, M. T., Gauld, I.C.: Reactor fuel Isotopic and Code Validation for Nuclear Applications: ORNL/TM-2014/464

27. http://urn.kb.se/resolve?urn=urn:nbn:se:uu:diva-340081 (slide 28)

28. "Exchange Forum 8": (https://igdtp.eu/event/igd-tp-exchange-forum-8/):

29. https://igdtp.eu/document/igd-tp-exchange-forum-8-day1-0940/

Acknowledgement

I would like to thank my supervisors in Lund and Uppsala, Prof. Kristina Stenström and Dr Peter Jansson for all their help, encouragement and patience. Without their support this thesis would never been completed..

I would also like to thank: Ian Gauld at ORNL for encouragement and permission to use figures from his reports.

I finally thank Prof. Ane Håkansson at Uppsala University for the idea for this project.