Studying neutron-induced fission at IGISOL-4

From neutron source to yield measurements and model comparisons

ANDREA MATTERA
Fission yields represent the probability of producing a certain nuclide in a fission event, and are important observables for fission research. For applications, accurate knowledge of the yields is fundamental at all stages of the fuel cycle of nuclear reactors, e.g., for reactivity calculations, or to estimate (spent) fuel inventory. Fission yields also help in the basic understanding of the fission process, for nucleosynthesis models, and for radioactive ion beam production.

This thesis was developed in the framework of the AlFONS project, the objective of which was to measure neutron-induced fission yields of relevance for partitioning and transmutation of spent fuel. The work is performed at the IGISOL-4 facility in JYFL (University of Jyväskylä).

The first part of this thesis work is dedicated to the development and characterisation of a suitable $^9$Be(p$(30\text{MeV})$,nx) neutron source for IGISOL-4. The neutron energy spectrum and the neutron yield from a 5mm thick converter were studied with Monte Carlo simulations. Two characterisation campaigns that validated the MCNPX code were also performed. At the maximum current available from the cyclotron at JYFL, a total neutron yield between 2 and $5\times10^{12}$ neutrons/(sr s) can be obtained. This satisfies the design goal for studies of fission yields.

The neutron source was used in the measurement of fission yields from high-energy neutron-induced fission of $^{235}$U at IGISOL-4, discussed in the second part of this thesis. The fission products were online-separated with a dipole magnet. The isobars, with masses in the range $A = 128-133$, were identified using γ-spectroscopy. Data for the relative yields of tin and antimony, as well as isomeric yield ratios for five nuclides will be reported. The yields show trends not observed in the ENDF/B-VII.1 evaluation, and only in part confirmed by the GEF model.

The final part of this thesis concerns a study of the performance of different nuclear model codes, that aim at describing the states of the fission fragments right after scission. Reproduction of experimental data serves to benchmark the models and it indicates, to some extent, how reliably the results can be extrapolated to regions where no data exist.

A methodology to compare and test these models has been developed, which was implemented in the DE\(_{\text{EF}}\)FIN code. DE\(_{\text{EF}}\)FIN takes the excited fission fragments, defined by the model under test, and de-excites them in a standardised way using the nuclear model code TALYS. Eliminating any variability in the way the final observables are extracted helps focusing on each model's assumptions. DE\(_{\text{EF}}\)FIN was tested on five models, and interesting features in the prompt neutron multiplicity were found for some of them. This study will promote a better understanding of the ideas used in the development of fission models.

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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 wrote the paper and participated in the design and data-taking of the experimental campaign. I also carried out most of the analysis related to the Time-of-Flight setups (Liquid Scintillator and Thin-Film Breakdown Counters).


**My contribution:** I wrote the paper and developed the code to perform the comparison of fission codes.

III  A. Mattera et al. Comparing fission codes with the DEFIN code. Manuscript.

**My contribution:** I wrote the paper and developed the code to perform the comparison of fission codes.

IV  A. Mattera et al. Production of Sn and Sb isotopes in high-energy neutron induced fission of natU. Submitted to The European Physical Journal A.

**My contribution:** I wrote the paper, participated in the preparation of the experimental campaign and part of the data-taking, and performed the analysis.

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Papers not included in this Thesis

List of papers related to this thesis, but not included in the comprehensive summary.

A neutron source for IGISOL-4

Fission yields at JYFL

Developments at IGISOL-4

Others
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Preface

In every job that must be done,
there is an element of fun.
You find the fun and… *SNAP*
the job’s a game!

Mary Poppins

The initial motivation for this thesis work is the measurement of high-quality nuclear data. My PhD studies began in the framework of AlFONS (Accurate FissiOn data for Nuclear Safety), a project supported by SSM (Swedish Radiation Safety Authority) and SKB (Swedish Nuclear Fuel and Waste Management Company) that ran from July 2010 until the end of 2014.

The objective of AlFONS was to measure neutron-induced independent fission yields of various actinides of relevance for partitioning and transmutation of spent fuel, as well as other quantities of interest for nuclear applications. The measurement was planned in collaboration with the University of Jyväskylä (Finland), at the IGISOL-4 facility.

In order to perform this measurement, the first goal of my PhD project has been to design, develop, characterise and test a suitable neutron source to deploy at IGISOL-4. This required more than three years of intense work by our group at Uppsala University, as well as by our collaborators in Finland.

The hope was to start measuring fission products as soon as possible, once the source was deployed. Unfortunately, as it sometimes happens in experimental physics, some of the things that could go wrong, went wrong. One of the key devices we need for the production of neutrons, the high-current MCC30/15 cyclotron, started suffering a long streak of technical glitches that gave (and still gives) the accelerator team in Jyväskylä a very hard time.

We were then forced to go for the backup plan: using the other cyclotron that serves the IGISOL-4 beam-line. This, despite some limitations on the maximum achievable current and, hence, on the fission rate, allowed us to systematically identify, for the first time at IGISOL-4, products from fission of $^{238}$U induced by high energy neutrons. The run, completed in December 2016, left us with plenty of data that, as of today, we have analysed only in part. The data include, besides fission yields, measurements to determine the efficiency of the extraction and transport of ions along the beam-line. Once fully studied,
these results will be an important step towards a better understanding of the IGISOL-4 facility and its use for the measurement of fission yields.

On a track that we advanced in parallel with the work I discussed above, we tried not to forget the more physical side of fission studies, that does not necessarily have to do with nuclear data for applications directly. We know, for example, that in order to interpret and understand data obtained using the white neutron source we developed for IGISOL-4, we need to include some degree of modelling in the discussion.

First of all, we want calculations to compare our data to. In addition, we would like to study how different neutron energies affect the results, and hopefully reach a point where we can study the energy dependence of fission yields.

From our point of view as users, the landscape of the models and codes used today for the simulation of fission-related quantities is not easy to navigate.

In order to answer some of these questions, we started a work aimed at comparing a selection of the available phenomenological codes. These models define the state of the fission fragments at scission. A few do so by using a phenomenological description of the compound nucleus from formation up to the scission point, while others start from experimentally measured quantities (typically kinetic energy and mass distributions). Assumptions specific to each model describe how the system evolves from there (e.g., how the energy is shared between the fragments or among the different degrees of freedom).

Finally, the fission fragments are de-excited, in order to extract the observables of interest, such as average neutron or gamma multiplicities, isomeric yield ratios, etc.

For this study, I developed the DE/FIN (De-Excitation of FIssion fRags) code, that aims at eliminating inconsistencies in the way the final observables are extracted. In its current form, DE/FIN takes the excited fission fragments and de-excites them in a reproducible and standardised way, using the physics models in the TALYS reaction code. We believe that eliminating any inconsistencies in the way the final observables are extracted can help focus on the fission models’ assumptions, and compare them fairly to each other. In this work, I mostly tested this methodology to extract prompt neutron distributions, $\bar{\nu}(A)$, but the same basic principle can be used to extract other quantities, e.g., isomeric yield ratios.

In chapter 1, I will present a theoretical background on fission, with a brief description of the evolution of the models, up to the latest phenomenological codes. In the same chapter I will also give an overview of current techniques.
and experiments for the measurement of fission yields, as well as a description of the IGISOL-4 facility.

Chapter 2 is entirely dedicated to the neutron source: its development, and its characterisation in two campaigns in Sweden and Finland aimed at validating the Monte Carlo models used to study its energy spectrum. Here I will also discuss what we do not know about the neutron source; the measurements that we should still perform, before we can provide accurate fission data. This chapter is largely based on Paper I.

The results from the n-IGISOL campaign in December 2016 will follow, in chapter 3. Here I will discuss what we learned from the failed attempts that preceded this successful measurement, as well as the analysis of a selection of the available data, that were used to obtain, for the first time at IGISOL-4, yields from neutron-induced fission. The results presented in this chapter are summarised in Paper IV.

I will finally discuss the ideas behind the DE\textsubscript{FIN} code in chapter 4. I will show the validation of the methodology and the results of the comparison between five phenomenological codes, along with some plans for the future developments of this study. The work on DE\textsubscript{FIN} has been reported in Paper II and Paper III.
1. Introduction

Let’s start at the very beginning  
A very good place to start . . .  
Maria, *The Sound of Music*

In this chapter, I will give a brief overview of the background behind my thesis work: what is fission? How can it be described? Why is it still important to measure fission-related nuclear data? What are fission yields? How can they be measured?

I will then introduce, in sect. 1.3, the IGISOL-JYFLTRAP facility, where fission yields from proton-induced fission have been measured in the past with the JYFLTRAP Penning trap, and which was proposed as the location for studies of neutron-induced fission yields.

1.1 Theoretical Background

In nuclear fission, a heavy nucleus splits up in, typically, two lighter nuclei. This process can happen spontaneously, or be induced by providing excitation energy to the heavy nucleus through, e.g., absorption of a neutron. Due to the higher binding energy per nucleon in the products, a large amount of energy is released in the reaction.

1.1.1 The Dawn of Fission

Fission was recognised by Hahn, Strassmann and Meitner, in 1938 [1]. Fermi had been irradiating heavy nuclei with neutrons for the previous four years [2], with the intention of artificially producing previously unseen transuranic elements. The products of these irradiations were radioactive nuclides with varying half-lives, initially identified as isomeric states of $^{231}\text{Ra}$ and $^{231}\text{Ac}$ produced in $^{238}\text{U}(n,2\alpha)$ reactions.

It was a dedicated series of experiments by Hahn and Strassman, specifically aimed at identifying the chemical species of the reaction products that proved, beyond doubt, that what were thought to be radium and actinium were, in fact, isotopes of the much lighter barium and lanthanum.
Figure 1.1. Charge yields of $^{238}\text{U}$ (red) and $^{232}\text{Th}$ (blue) from the ENDF/B-VII.1 evaluated nuclear data library [5]. The shaded rectangle corresponds to the low-yield elements not seen in the first experiments for the identification of FPs.

The $^{238}\text{U}$ nucleus, absorbing a neutron, could split in two much lighter nuclei, “the precise ratio of sizes depending on finer structural features and perhaps partly on chance” (Meitner & Frisch (1939) [3]).

From an approximate calculation, based on the nuclear radii and charges of the Fission Products (FPs), Meitner and Frisch also estimated, in 1939, the kinetic energy available for the reaction: about 200 MeV [3]. In the same work, they proposed an explanation based on the Liquid Drop Model (LDM): the excitation energy provided when a neutron is absorbed by a heavy nucleus is enough to cause an instability in the drop that eventually leads to its fission.

By 1940, at least 13 elements had been experimentally identified among the FPs, mostly in fission of uranium and thorium [4]. From Turner’s review of all the main developments up to 1940, we also read:

> It is noteworthy that no elements between $^{42}\text{Mo}$ and $^{51}\text{Sb}$ have yet been found. It seems likely that this represents a real absence or relatively low yield of such elements, rather than merely an unfinished stage of comprehensive chemical investigation. (Turner (1940) [4])

This was the first recognition of the double-humped nature of the Fission Yield (FY) distribution of uranium and thorium, as shown in fig. 1.1. Today we have a better understanding of the reasons behind this phenomenon, as I will explain in the next sections.
Figure 1.2. Schematic representation of the neutron-induced fission of $^{235}$U. A neutron is captured by the $^{235}$U nucleus 1, creating the excited Compound Nucleus $^{236}$U$^*$ 2. The compound nucleus deforms and elongates 3, until it scissions in two highly-excited FFs 4. The FFs repel each other due to Coulomb interaction, and de-excite by emission of prompt neutrons and $\gamma$s 5.

1.1.2 Fission Observables

A fission event (e.g., fission of $^{235}$U induced by a neutron, in fig. 1.2) is characterised by a plethora of physical quantities. Studying how these quantities vary and influence each other is one of the keys to understanding the fission process.

In our example, a neutron is first captured by the actinide (1 in fig. 1.2) to form a Compound Nucleus (CN), $^{236}$U$^*$. The CN is characterised by an excitation energy $E^*_\text{CN}$ (2), that in this simple picture is always enough for it to undergo fission. $E^*_\text{CN}$ depends on the neutron separation energy $S_n$ of the CN, and on the energy of the incoming neutron $E_n$. If $E_n$ is sufficiently high, the CN will be able to emit a neutron and still have enough energy to fission: this is called second-chance fission. By increasing $E_n$, the energy will be sufficient to emit more than one neutron before fissioning, leading to third-chance fission, fourth-chance fission, etc.

The excited CN deforms and elongates (3), until it splits. Later in this chapter, I will give an overview of some of the models that explain how the nucleus can reach the scission point (i.e., the point where the neck between the two fragments snaps).
Once the CN splits, two new nuclei are formed: the Fission Fragments (FFs) (4). FFs generally preserve both the number of nucleons and the A/Z of the parent nucleus. This leads to the fact that — being much lighter than the CN — the FFs will be far off the valley of stability of the chart of nuclides, on the neutron-rich side (see inset of fig. 1.2).

Being two positively charged nuclei in close proximity, the FFs are subjected to a strong repulsive Coulomb force. The sum of the kinetic energy of the FF pair defines the Total Kinetic Energy, $TKE$.

The remaining energy\(^1\) appears as excitation energy of the FFs ($TXE$), due to their highly deformed shapes at scission. The FFs release this excitation energy by emitting particles: prompt neutrons first, followed by prompt $\gamma$s (5). The average number of neutrons emitted in a fission event is called $\bar{\nu}$ (nu-bar), and its value can also be expressed as a function of the mass number of the fragment, $\bar{\nu}(A)$.

The emission of prompt radiation is generally enough to de-excite the nuclei, that are now conventionally called Fission Products (FPs). It is often not sufficient, however, for them to reach stability. This usually happens through $\beta^-$-decay, that allows the FP to move to less neutron-rich nuclei, towards the valley of stability. If the end-state of a $\beta^-$-decay is above the daughter’s neutron separation energy $S_n$, this can lead to the emission of a neutron (in this case referred to as a delayed neutron, to distinguish it from prompt radiation).

Many of these quantities have been studied experimentally throughout the years, in order to better understand fission. Among the many experiments that were undertaken, since 1939, was the measurement of FPs in terms of their kinetic energy and production yield.

Later, experiments also focused on the determination or direct measurement of the prompt radiation (both neutrons and photons). Now, multi-parametric experiments, that try to reconstruct as many of the observables as possible for each fission event, are the last frontier of experimental fission research.

**Fission Yields**

Fission yields, the amount of a certain nuclide produced after fission, are important quantities that were measured very early-on, when large radiochemistry laboratories were set up in the framework of the Manhattan project. Anderson, Fermi and Grosse were among the first to measure what were then referred to as branching ratios of $^{235}$U [6].

Due to the complexity of the fission process and since the amount of a certain isotopic species will change with time, it is useful to define different types of FYs (adapted from ref. [7]):

- **Independent fission yields**: number of atoms of a specific nuclide produced directly in the fission process (i.e., not via radioactive decay of precursors).

\(^1\)i.e., the difference between the mass and excitation energy of the CN ($M_{CN} + E_C^{CN}$), and the masses and kinetic energies of the two FFs ($M_{FF1} + M_{FF2} + TKE$).
• **Cumulative fission yields**: total number of atoms of a specific nuclide produced (directly and via decay of precursors).

• **Total chain yields**: defined as the sum of cumulative yield(s) of the last (stable or long-lived) chain member(s).

• **Mass number yields**: defined as the sum of all independent yields of a particular mass chain and are in this way distinguished from chain yields. Some of the most modern methods to measure fission yields provide sets of truly independent yields which - at summation - will produce mass number yields rather than chain yields.

Some experimental methods allow the measurement of yields prior to prompt neutron emission. These yield distributions are designated as fragment yields. Measurements of yields posterior to prompt neutron emission are instead referred to as product yields. (IAEA-TECDOC-1168 (2000) [7])

In a fission event (e.g., thermal fission of $^{235}$U), more than 140 nuclei can be produced directly after fission with a probability (independent fragment yield) larger than 0.1% [8].

Trying to explain how the shape of the FY distribution changes with the mass of the CN and its $E_{CN}^*$ was, and still is, one of the challenges that fission models have to face. Here follows a short description of these efforts, from the early LDM, to the latest phenomenological models used today to simulate fission observables.

### 1.1.3 The Liquid Drop Model

The first modelling of fission came almost immediately after the process was recognised and was based on a classical, macroscopic picture of the atomic nucleus, that had been described a few years earlier as a charged liquid drop in the works of Weizsäcker [9] and Bethe [10]. Just like a drop of liquid subjected to vibrations can divide into two droplets, so a nucleus could fission in two smaller fragments.

In the LDM, the binding energy of a nucleus (eqn. 1.1) is obtained as the sum of different terms: a volume term proportional to the nuclear mass $A$, a surface energy term proportional to the surface area (i.e., $A^{2/3}$), and a Coulomb energy term proportional to the inverse of the radius ($R^{-1} \approx A^{-1/3}$). Terms that account for effects due to the individual nucleons, $E_p$, are finally added [11]:

\[
E_{\text{LDM}} = E_v + E_s + E_C + E_p \\
= a_v A - a_s A^{2/3} - a_C \frac{Z(Z - 1)}{A^{1/3}} - a_{\text{sym}} \frac{(A - 2Z)^2}{A} + \delta
\] (1.1)

$E_v$: the volume term, is strongly attractive, due to the strong force among nucleons. The dependence on $A$ (instead of the combinatorial $A(A - 1) \approx A^2$) suggests that nucleons only see the attraction of the immediate neighbours, *i.e.*, the strong force has short range.
**$E_s$:** the surface term is slightly repulsive. It takes into account the fact that the nucleons on the surface of the drop are surrounded by less nucleons, thus reducing the nucleus binding energy. It scales as the nuclear surface, $R^2 \approx A^{2/3}$.

**$E_C$:** the Coulomb repulsion tends to push protons away, reducing the binding energy of the nucleus. Unlike for strong force, the long range of Coulomb interaction causes all the protons to repel each other, making this term proportional to $Z(Z-1)/R \approx Z(Z-1)/A^{1/3}$.

**$E_p$:** in eqn. 1.1, this term includes two contributions that are due to behaviour attributable to individual nucleons. The asymmetry effect, makes it energetically unfavourable to have nuclei with heavily asymmetric distributions of protons vs neutrons (expressed by the $(A-2Z)$ factor, that is zero for $Z = A/2$); the pairing term $\delta$, accounts instead for the tendency of nucleons of the same species to couple in pairs in a more bound configuration.

In a nucleus deforming from its spherical shape, the negative contribution of $E_s$ will increase, due to a larger surface, and compete with that of $E_C$ that decreases, due to a larger average distance between the nucleons; A parameter $x$ that expresses stability against the deformation $\beta$ can then be defined: the fissibility parameter [12,13]. Referring to the energies of the spherical nucleus ($\beta = 0$), its value can be expressed as:

$$x = E_C(0)/2E_s(0),$$

(1.2)

proportional to $Z^2/A$. For $x < 1$, nuclei are stable against deformation.

The LDM predicts a local minimum for all stable nuclei at $\beta = 0$, i.e., a spherical shaped Ground State (GS). The fission barrier appears as a single saddle of $E_{LDM}$ along a deformation path that minimises the energy (dashed line in fig. 1.3). For actinides ($x > 0.4$), the LDM predicts a typically symmetric mass split of the FFs [12, 14].

The LDM is very successful in predicting the observed behaviour of the nuclear binding energy, and the absolute scale of the fission barrier as a function of deformation. It is however neither able to describe the observed double-humped structure of FY distributions, nor the deformed GS of actinides.

Microscopic single-particle effects must be included to reproduce these observations. A way to do that is the one proposed by Strutinsky.

### 1.1.4 Correction for Shell-Effects

In order to allow for an asymmetric mass split, microscopic effects due to nuclear shells must be included in the picture.

A successful description was introduced by Strutinsky [15], with the *macroscopic-microscopic* approach. This method combines the accurate determination of the energy scale coming from the LDM, with the fluctuations due to single-particle effects. The energy of the nucleus is calculated as:
\[ E = U - \bar{U} + E_{LDM} + \delta P = E_{LDM} + \delta U + \delta P, \]  

(1.3)

where \( U = \sum_\nu n_\nu \varepsilon_\nu \) is the discrete single-particle energy; \( \bar{U} = \int_{-\infty}^{\varepsilon_{\text{max}}} \varepsilon \tilde{g}(\varepsilon) d\varepsilon \) is the single-particle energy in which the density of states \( \tilde{g}(\varepsilon) \) is smeared to wash-out shell-effects; \( E_{LDM} \) is the energy calculated with the LDM, as in eqn. 1.1 and \( \delta P \) is a correction for the pairing effects [16].

For actinide nuclei, the shell correction \( \delta U \) will be strongly negative at the deformation that corresponds to the maximum of the LDM potential [17]. This correction — depending on the mass of the CN — changes the shape of the potential energy landscape and can lead to a double-humped fission barrier (solid line in fig. 1.3). Also, the inclusion of shell effects moves the minimum of the potential energy corresponding to the GS from a spherical shape, to one with a finite deformation.

While the description introduced with the shell correction is able to reproduce the main features of the FY mass distributions, this model is still not adequate for a general description accurate enough to be used, e.g., for application purposes. The models describe one fission mode, identified by the path of minimum potential through the barrier, that is too simplistic. In order to explain the features of the FY(A) distribution as well as of TKE(A), a more refined model is required.

A considerable improvement was introduced by Brosa, with the Multi-Modal Random Neck Rupture (MM-RNR) model.

1.1.5 The Multi-Modal Random-Neck Rupture Model
The MM-RNR model proposed by Brosa [18], combines two main ideas: (1) multi-channel fission states that, by riding the potential energy landscapes using different deformation paths, multiple nuclear shapes can be reached at scission; (2) once the shape of the nucleus is defined based on the fission mode, the break-up happens in a random location along a flat neck between the two nascent fragments.

The model assumes four main fission modes (depicted in fig. 1.4), each with different characteristics of the nascent FFs mass asymmetry and neck length:

- **Standard (S1/S2/S3)** mode, of which three flavours exist, with increasing asymmetry. This mode is slightly asymmetric, with a neck of a standard length. It is the dominating mode (≈95%) in near-barrier fission of heavy nuclei of intermediate mass (230 < A < 255).

- **Super-Long (SL)** mode. A symmetric mode with a longer than average neck. It dominates in near-barrier fission at lower masses (A < 230). It contributes to a mass distribution centred at \( A = A_{CN}/2 \), with a large variance (the longer neck allows for more possible configurations of masses).
Figure 1.3. Schematic representation of the potential energy from the LDM as a function of the nucleus deformation, with (blue) and without (red) shell-effect corrections. See the text for further explanation.

The longer distance between the nascent fragments leads to a lower total kinetic energy ($TKE$).

- **Super-Short (SS)** mode. Another symmetric mode, but with a neck shorter than average. This mode dominates in near-barrier fission at higher masses ($A > 255$). The variance of the distribution is smaller, while the $TKE$ is higher.

- **Super-Asymmetric (SA)** mode. As the name suggests, it accounts for the FPs at the edges of the mass distribution.

This description is able to reproduce not only the mass yield distributions, but also the systematics of $TKE$ and the prompt neutron emission $\bar{\nu}(A)$, and it has been extensively used to predict and model fission quantities.

The $\bar{\nu}(A)$ distributions expected for all modes is also shown, in red, in fig. 1.4. The excitation of the fragments, and hence neutron emission, is related to their deformation. Modes with an asymmetric pre-scission shape (S1/S2/S3 and SA), present a sawtooth-shaped $\bar{\nu}(A)$ that accounts for the fact that two fragments with about equal masses have very different deformations. This is the case for the $\bar{\nu}(A)$ seen in, e.g., $^{235}\text{U}(n_{th},f)$, $^{239}\text{Pu}(n_{th},f)$ or $^{252}\text{Cf}(sf)$. Modes that produce a symmetric pre-scission shape of the FFs (e.g., SL and SS) give rise, instead, to a $\bar{\nu}(A)$ smoothly increasing with $A$. A more detailed discussion is provided in ref. [18].
It is now almost 30 years since this picture was introduced by Brosa, and during this time, new ideas have been explored for the modelling of fission. One of the successful approaches is the one introduced by P. Möller with the calculations of potential landscapes. Based on a five-dimensional parametrisation of the nuclear shapes, Möller calculated multi-dimensional potential energy landscapes for a large selection of nuclei [19–21]. These have been successfully used, e.g., in the random walk model proposed by Randrup [22] for the calculation of fission yields.

The ambitious endeavour of microscopic \textit{ab initio} calculations is still far from the needs of, e.g., nuclear data in terms of both precision and computing time [23].

Phenomenological models use existing nuclear data as input, or as a way to fix the values of model parameters. Though less rigorous, they are fast and able to reproduce data sufficiently well, and are thus frequently used in the nuclear data community. On the other hand, their power to extrapolate to different fissioning systems — where no data exist — is often limited.

1.1.6 Phenomenological models

The landscape of available models is extremely wide and has developed extensively in the last decade: from the first attempts, with the Madland-Nix Los Alamos Model [24] (that has since been extended [25], and included into Vladuca and Tudora’s PbP model [26, 27]), able to describe integral fission
properties; to the latest developments of Monte Carlo codes such as GEF [8], FREYA [28, 29], CGMF [30, 31] and FIFRELIN [32, 33], that simulate fission quantities on an event-by-event basis, and can then account for fluctuations of, and correlations between different observables.

A major distinction can be made among the models listed above.

One one side there is the GEF code, that is possibly the only one in the list that includes an explicit modelling of fission. GEF starts from the formation of the CN, and using a phenomenological depiction of the fission barrier height, defines two FFs for each fission event. The GEF code then calculates the total excitation energy $TXE$, based on the ideas reported in ref. [34].

The other codes, instead, typically start from a measured average kinetic energy distribution $TKE(A,Z)$, without introducing an explicit description of the evolution of the CN from creation to scission.

From this point forward, the main ingredients of all codes are essentially the same. The total energy available for the reaction is calculated. In case of neutron-induced fission, this is the sum of the incoming neutron energy $E_n$, the CN neutron separation energy $S_n^\text{CN}$, and the $Q$ value of the reaction:

$$E_{\text{tot}} = TXE + TKE = E_n + S_n^\text{CN} + Q, \tag{1.4a}$$

where $Q$ is defined, for a given mass split, as:

$$Q = M_{\text{CN}} - (M_{\text{FF1}} + M_{\text{FF2}}) \tag{1.4b}$$

Energy conservation is invoked, and from $TKE$ or $TXE$ the other quantity can be obtained using eqn. 1.4a.

The excitation energy is then shared between the two fragments. This step is possibly the most debated upon, as discussed in refs. [31, 35, 36]. It is here that some of the "magic" happens, and different models introduce ad-hoc corrections to the underlying physical assumptions in order to reproduce data.

The final step, once the physics parameters are fixed, is the extraction of the observables ($\bar{\nu}(A,Z,TKE), E_\gamma(A,Z)$, etc.).

The main focus of models is then, and rightly so, the definition of the properties of the FFs after scission. Authors then include some treatment of the de-excitation of FFs, needed in order to extract measurable observables that can be used to compare the model with experimental data. Fission quantities calculated by these codes appear then as the convolution of these two steps.

In chapter 4, I will describe a methodology to compare the models among each other and with experimental data, that is based on the main idea of detaching the fragments de-excitation from the codes and perform it in a standard, consistent way.

Here follows a list of some of the available fission codes. In each of the paragraphs, I will use a notation that is consistent with the publications authored by the codes developers, so that the same quantity might be defined differently throughout the rest of the thesis.
GEF

GEF (GEneral Fission)\(^2\), is a semi-empirical Monte Carlo model developed by K.-H. Schmidt and B. Jurado. It produces a variety of fission observables on an event-by-event basis. “The GEF code aims to provide a complete description including the entrance channel and the de-excitation of the fragments.” (General Description of Fission Observables. K.-H. Schmidt, B. Jurado and C. Amouroux [34].)

The GEF model uses many general theoretical ideas and, combining them with the available empirical information, implements them in a code that can, technically, be used to fission all heavy nuclei from polonium (\(Z = 84\)) to darmstadtium (\(Z = 100\)). The models implemented into GEF are largely phenomenological, and avoid the limitations of microscopical models (i.e., the need for heavy approximations and large computing power).

Unlike all the other models described in this chapter, the \((A, Z)\)-dependent parametrisation of the fission events included in GEF, makes it so that the model does not need to rely on experimental FY distributions or \(TKE\), which are instead an output of GEF. Parameters are optimised globally to reproduce experimental data.

The GEF code includes a model of the fission barrier height based on the idea pursued in the macroscopic-microscopic approach. The barrier height is the most critical input that affects, among others, the \(FY(A, Z)\) and the intrinsic excitation energy of the FFs \(E^{\text{int}}\). In ref. [34], the authors demonstrate how their model, with an empirical, mass-dependent correction to the barrier height, is able to accurately reproduce the experimental values for a variety of compound nuclei.

GEF calculates the excitation energy of the light and heavy fragments \(E^{*}_{L/H}\) as the sum of intrinsic, collective \(E^{\text{coll}}\), deformation \(E^{\text{def}}\) and rotational \(E^{\text{rot}}\) energy:

\[
E^{*}_{L/H} = E^{\text{int}}_{L/H} + \frac{E^{\text{coll}}_{L/H}}{2} + E^{\text{def}}_{L/H} + E^{\text{rot}}_{L/H}
\]  

The collective energy is shared equally between the two FFs (hence the \(\frac{1}{2}\) factor); \(E^{\text{def}}\) is calculated as the deformation energy of the nascent fragment at scission compared to a spherical shape; \(E^{\text{rot}}\) is the FFs rotational energy, calculated in a similar fashion as in FIFRELIN (see eqn. 1.12, later in this section). The \(TKE\) is obtained as the difference between the reaction Q-value and \(TXE\).

Other important ingredients for the nuclear model are the nuclear level densities, based on the constant-temperature description given by von Egidy and Bucurescu [37]. This choice in particular, differentiates GEF from all the other main fission codes, that usually assume a thermal equilibrium approach.

\(^2\)It has become customary in the nuclear data community to pronounce the name of the GEF code \(\text{[gef]}\), to avoid confusion with the Joint Evaluated Fission and Fusion File (JEFF), pronounced \(\text{[\#ref]}\).
The de-excitation of the FFs is calculated within a statistical model: neutron and photon emission compete above the neutron separation energy and their energy and momentum are sampled by Monte Carlo. Once the yrast line is reached, angular momentum is carried away by a $\gamma$-cascade.

**PbP**

The PbP (Point-by-Point) model is developed by A. Tudora and co-workers at the University of Bucarest [35, 38, 39].

It is a deterministic model, that produces “multi-parametric matrices” of fission-related quantities, expressed as a function of the fragment mass, charge and kinetic energy. These include, e.g., the fragments’ excitation energy at full acceleration $E^*_{\text{full acc.}}(A,Z,TKE)$, the prompt neutron multiplicity $\nu(A,Z,TKE)$, etc., quantities that are generically labelled $q(A,Z,TKE)$.

The matrices are calculated independently of the yield, but appropriate summing over one, or more, of the parameters and weighing with a yield distribution $Y(A,Z,TKE)$, produces average observables $\bar{q}(A), \bar{q}(Z), \bar{q}(TKE)$ or $\langle q \rangle$. The mass yield distributions as a function of $TKE$ can be obtained from any model or systematics, but experimental data-sets are generally preferred [40].

In order to obtain the matrices, the excitation energies of the fragments must be calculated. In the PbP model, the two FFs are represented at scission as heavily deformed, due to the residual attraction in the position of the snapping neck. In the first step of the model, the extra-deformation energies $\Delta E_{\text{def}}^{L/H}$ are calculated for both fragments, from the shell-corrected LDM, as the difference between the absolute deformation at scission and at full acceleration.

The available excitation energy at scission $E_{\text{sc}}^*$ is calculated then as the difference between the $TXE$ — obtained from energy conservation considerations — and the extra-deformation energy:

$$
E_r + E_n + B_n = TXE + TKE =
E_{\text{sc}}^* + \Delta E_{\text{def}}^L + \Delta E_{\text{def}}^H
$$

\hspace{1cm} \text{(1.6a)}

and

$$
E_{\text{sc}}^* = TXE - (\Delta E_{\text{def}}^L + \Delta E_{\text{def}}^H), \hspace{1cm} \text{(1.6b)}
$$

where $E_n$ and $B_n$ are the incoming neutron energy and the neutron binding energy, respectively; and $E_r$ is the energy release (or Q-value) of the reaction.

It is the available excitation energy at scission that is shared between the two fragments:

$$
E_{\text{sc}}^* = E_{\text{sc}}^L + E_{\text{sc}}^H, \hspace{1cm} \text{(1.7)}
$$

assuming statistical equilibrium, so that:

$$
\frac{E_{\text{sc}}^L}{E_{\text{sc}}^H} = \frac{a_{\text{sc}}^L}{a_{\text{sc}}^H}, \hspace{1cm} \text{(1.8)}
$$
where \( a_{sc}^{L/H} \) are the level density parameters of the nascent fragments in the Fermi-Gas regime. The excitation energy of the nascent fragments at scission and the level density parameters are obtained simultaneously by an iterative procedure under the condition of equal temperatures of the fragments.

Finally, for each fragment, the excitation energy at full acceleration \( (E_{FA}^{L/H}) \) is obtained as the sum of the excitation energy at scission obtained from eqn. 1.8 and the extra-deformation energy:

\[
E_{FA}^{L/H} = E_{sc}^{L/H} + \Delta E_{def}^{L/H}
\]  

(1.9)

Neutrons are evaporated from the fragments (in competition with photons) as long as the available excitation energy is larger than the neutron separation energy \( S_n \).

CGMF
The CGMF (CGM+FFD) code, developed at Los Alamos, is based on a Monte Carlo implementation of the Hauser-Feshbach model [41] for the de-excitation of nuclides [30,31,42]. It is focused on the emission of prompt neutrons and \( \gamma \)s and the result is a series of neutron emission histories, from which correlations can be extracted.

Mass distributions are provided as an input, using the best fit of available experimental data [31], while the charge distribution of fragments in a mass chain are extracted using Wahl’s systematics [43]. The \( TKE(A) \) is also an input, extracted from data using a lest-square fit.

The \( TXE(A) \) can then be calculated as the difference of the reaction Q-value and the \( TKE, \) at each event. Sharing of excitation energy at scission is, also here, a critical step: the assumption that the nascent fragments are in thermal equilibrium (and therefore the \( TXE \) is shared according to the level densities) is generally contradicted by observations, that show that more neutrons are emitted from the light fragment. For this reason, the authors introduce a scaling factor

\[
R_T = \frac{T_0^L}{T_0^H},
\]  

(1.10)

where \( T_0^L \) and \( T_0^H \) are the temperatures of the light and heavy fragments, respectively [31]. The value of \( R_T(A) \) in CGMF, is obtained as a fit of the ratio of the average neutron multiplicities of the light and heavy fragments, \( \bar{\nu_L}/\bar{\nu_H} \), measured experimentally as a function of mass.

FIFRELIN
FIFRELIN (FIssion FRagment Evaporation Leading to an Investigation of Nuclear data) is a code developed at CEA Cadarache with the aim of calculating fission observables. [40]

In the Monte Carlo approach implemented in FIFRELIN, the mass and total kinetic energy distributions are obtained from experimental data and sampled
to characterise every fission event. The nuclear charge is defined based on
the Wahl systematics. The excitation energy is calculated as the difference
between the reaction Q-value and the $TKE$ [33]:

$$TXE = Q - TKE$$

$$= E^{\text{int}} + E^{\text{rot}} + E^{\text{rot}}$$

$$= E^{\text{int}}_L + E^{\text{int}}_H + E^{\text{rot}}_L + E^{\text{rot}}_H$$

$$= E^*_L + E^*_H,$$

where $E^{\text{int}}$, $E^{\text{int}}_L$ and $E^{\text{int}}_H$ are the intrinsic excitation energies of the compound
nucleus, light and heavy fragments, respectively; $E^{\text{rot}}_L$ and $E^{\text{rot}}_H$ are instead the
rotational energies. The rotational energies $E^{\text{rot}}_{L/H}$ are approximated in FIFRE-
LiN using the rotating liquid-drop model:

$$E^{\text{rot}} = \frac{\hbar^2 J (J + 1)}{2 \mathfrak{I}},$$

where $J$ is the total nucleus’ angular momentum and $\mathfrak{I}$ stands for the moment
of inertia. Determination of $\mathfrak{I}$ is studied in some detail in ref. [33], where it is
found that the data are best described if the moment of inertia is equal to 50%
of the one of a rigid spheroid.

The sorting of the intrinsic excitation energy between the two fragments
is based on the observation that a constant-temperature ratio cannot properly
reproduce the experimental $\tilde{\nu}(A)$ sawtooth. The authors introduce, similarly
to CGMF, a mass-dependent temperature ratio $R_T$. From ref. [33], describing
the procedure for fission of $^{252}$Cf:

We consider three typical fragment configurations for which the ratio $R_T$ should
be lower, equal, or greater than 1:

(i) For symmetric fission we expect the same temperature for both comple-
mentary fragments and then $R_T = 1$.

(ii) For light mass number $A_L = 120$, $R_T$ is maximum because in the case
of $^{252}$Cf the complementary heavy fragment is nearly spherical with 132
nucleons (corresponding to $Z = 50$ closed proton shell and $N = 82$ closed
neutron shell). Consequently the light fragment $A_L = 120$ gains the major
part of the total excitation energy associated with a higher temperature
compared to its double magic complementary partner.

(iii) For very asymmetric fission the heavy fragment is more deformed than
the light fragment because the latter becomes shell stabilized. The light
fragment can be nearly spherical with a $Z = 28$ closed proton shell and an
$N = 50$ closed neutron shell, leading to a temperature lower than the
temperature of the heavy fragment ($R_T < 1$). […]

A simple composition of two linear laws is assumed between these three key
configurations.

(Litaize O. et al. Phys Rev C 82, 054616 (2010) [33])
De-excitation of FFs in FIFRELIN is treated using the Hauser-Feshbach formalism, in order to produce fission observables [44].

FREYA
The FREYA (Fission Reaction Event Yield Algorithm) code was first introduced to the community in 2009 [28, 29].

It is, like GEF, CGMF and FIFRELIN, a Monte Carlo-based approach aimed at producing complete fission events, focusing on neutron and photon emission. A detailed description of the code can be found in refs. [29, 45, 46]. Pre-equilibrium and pre-fission evaporation of neutrons is explicitly taken into account in neutron-induced fission.

The mass and charge distributions are modelled with a five-Gaussian fit of experimentally determined mass distributions, representing three distinct fission modes; the average prompt neutron multiplicity is used to convert the experimental FP yields into fission fragment yields needed by FREYA.

The $TKE(A)$ is also fit to experimental data and is used to extract the available $TXE$ for each event.

After accounting for the total rotational energy of the two fragments, $E^{\text{rot}}$, the total statistical fragment excitation:

$$E^{\text{stat}} = TXE - E^{\text{rot}} \quad (1.13)$$

is shared between the two fragments, with the assumption of thermal equilibrium corrected for the fact that observations suggest that “the light fragments tend to be disproportionately excited” (FREYA 2.0.2 User Manual [45]).

Once the fragments are assigned their excitation energy, de-excitation can proceed: first neutron evaporation — as long as energetically possible — followed by photon emission, the latter using both statistical (for the emission of statistical photons) and rotational excitation energies. “If the photon decay for a particular fragment is not in the RIPL-3 tables, the fragment must still dispose of its rotational energy. In this case, photons are emitted along the Yrast line […], after which the remaining energy is carried away by a single photon” (FREYA 2.0.2 User Manual [45]).

1.1.7 Yields to Understand Fission
Fission yields have been used as one of the first test-beds for fission models.

Figure 1.5 shows an overview of what is now experimentally known about mass yield distributions.

A systematic behaviour can be observed, with asymmetric fission prevailing from $^{227}$Ra to $^{256}$Fm, and the two peaks getting closer and gradually merging into one [34]. From the figure it is also possible to appreciate how the weight of the different modes (as from the picture suggested by Brosa) smoothly changes a function of the CNs, from a predominantly asymmetric fission for $^{234}$U, to a symmetric one in $^{218}$Th and $^{208}$Rn.
Since the early stages of fission theory, the shape of the yield distributions measured experimentally have helped steering the development of fission models, from the simple LDM description to more advanced pictures that use multi-dimensional potential energy landscapes of the CN to derive FF mass distributions.

Also now, a correct reproduction of FY(A) curves is a powerful test used to validate fission theories [47]. Especially when it comes to studies of the energy-dependence of FYs, data can provide valuable information to benchmark fission models.

In addition to independent mass and charge yields, Isomeric Yield Ratios (IYRs) are also of great interest. These can be expressed as the ratio between the yield of an isomeric state and the total yield for a given nuclide: \( \frac{Y_{is}}{Y_{is} + Y_{GS}} \), where \( Y_{is} \) and \( Y_{GS} \) are the yield (independent or cumulative) of the isomer and the GS, respectively. IYRs can be used to deduce the angular momentum of the FFs, which is, in turn, an important tool to look into the scission configuration.

Furthermore, in fission induced by fast neutrons, very exotic nuclides far from the line of stability are produced, which are of interest, for example, in nuclear astrophysics for the understanding of the nucleosynthesis process [48].

1.1.8 Fission Yields for Applications

Fission yield data not only are important for basic understanding of the fission process, but become essential for some applications.

Less than four years after the fission process was identified and the possibility to start a chain reaction theorised, the first self-sustaining reactor was built in a Squash Court on the campus of the University of Chicago. The reactor,
with natural uranium as fuel and moderated by graphite, was slowly brought to criticality on December 2\textsuperscript{nd}, 1942. It was proved that the power level of the nuclear pile could be easily controlled by means of neutron absorber strips inserted in the reactor core [49].

Twelve years later, the Soviet APS-1 reactor at Obninsk became the first nuclear power plant connected to the electricity grid [50]. Since then, the use of nuclear power for electricity production has increased remarkably all over the world, reaching a total installed capacity of 391 GW(e), in 2016 [51].

Fission yields are among the quantities that are needed for the operation of nuclear power plants, and accurate nuclear data on FYs are important in applications for several reasons.

Today, more than 60 years of operation of nuclear reactors prove that the knowledge in terms of nuclear data is sufficient to predict reactor parameters for the everyday operation of power plants. The inventory of FPs in a light-water reactor, the kind used on large scale all over the world, is generally known with enough accuracy to ensure their safe operation.

This might not be true if the fuel burnup\textsuperscript{3} would be increased beyond the current levels, so that even FPs that have a relatively low probability to be produced (because they are in the tails of the FY curve, or produced in fission of minor actinides) could become important in absolute number. This will affect not only the FP inventory for storage, but also reactor kinetics, since a good knowledge of the delayed neutrons precursors and of neutron poisons\textsuperscript{4} is required for safe reactor operation.

The requirements in terms of nuclear data are similar for the new concepts of Generation IV Fast Reactors and Accelerator Driven Systems. In these designs, however, the neutron energy is higher and data become scarce.

1.2 Measuring Fission Yields

Several different techniques exist for the measurement of FYs. Some are more applicable to the production of chain yield curves, while others — thanks to their short measurement time — can be used to obtain Independent Fission Yields (IFYs). Here follows a brief review of the available methods, with an overview of the strong and weak points of each technique.

1.2.1 Radiochemical separation

Measurements of FPs started in the 1940s during the Manhattan project and were performed using radiochemical methods. The technique consists in dis-

\textsuperscript{3}Burnup measures the utilisation of uranium in a reactor (GWd/Metric tonne of U) and depends on the time the fuel is kept in the reactor core and what power the reactor is operated at.

\textsuperscript{4}A neutron poison is a nuclide with a large cross section for neutron absorption. When the amount of poisons becomes too large, the reactivity of the reactor is affected [52].
solving the longest lived isotope of a radioactive chain, after a waiting time that has allowed all precursors to decay, and then measure its activity. In the early days, before the development of detectors for high-resolution $\gamma$-ray spectroscopy, $\beta$-radiation was usually measured. This required many corrections and a difficult analysis procedure that resulted in large uncertainties on the final product yields. This methodology is now abandoned in favour of $\gamma$-ray spectroscopy, and the old data are heavily scrutinised by evaluators before they are inserted into nuclear data libraries [7].

1.2.2 Mass spectrometry

Mass spectrometry, as the name suggests, measures the mass of the nuclides in an evaporated and ionised sample and is usually a very precise technique. The need for a normalisation step to go from relative to absolute yields is the main source of uncertainty [7].

A disadvantage with this technique is that large fissile targets and long irradiation times are required to obtain sufficient amounts of FPs for the analysis, possibly resulting in changes in the fission yield distribution due to neutron capture in the FPs. Even so, the typical uncertainty of this technique for measurement of chain yields is below a few percent [53].

This technique can also be applied to measurements of IFYs. In this case, the fission target is usually connected directly to the ion source needed to produce the evaporated and ionised target for the analysis. The problems connected with long irradiation times do not apply, but other precautions must be taken in order not to introduce biases due to, e.g., different volatilisation properties of different elements.

1.2.3 $\gamma$-ray spectroscopy

This technique identifies the FPs by analysing their characteristic $\gamma$-rays. The data analysis requires a good knowledge of the decay properties of the nuclides that are to be measured. The big advantage of requiring only milligram-amounts of the fissionable nuclide, makes this technique largely applied for exotic targets of limited availability [7].

The uncertainty in chain yield determination is typically not better than 10%, and FPs with very low yields are not accessible without further chemical-separation.

Measurements performed with this technique at the ILL (Institut Laue-Langevin) have recently been reported for $^{239}$Pu(n$_{th}$,f) [54]. In this experiment, the Lohengrin mass separator (see also sect. 1.2.4), already used to measure yields with a high-resolution ionisation chamber, was complemented with a moving implantation-tape and a $\gamma$-spectrometry station to extend the measurement of isotopic yields to masses $A > 42$ [55].
Experimental efforts are also under way, e.g., at the Triangle Universities Nuclear Laboratory (TUNL) [56, 57], at the LICORNE fast neutron source [58] and at the University of Michigan’s Neutron Science Laboratory [59].

1.2.4 Measurement of unstopped fragments

All techniques that measure FFs at their full kinetic energy can be included in this category. The basic principle of this technique is to calculate the masses of the two fragments from their kinetic energy and/or velocity using momentum conservation. One important requirement is for the fission target to be as thin as possible, to limit energy losses that could hinder an accurate determination of velocity and/or energy [7, 60].

Notable examples of this technique are the Lohengrin mass separator [61] and the HIAWATHA instrument [62], that separate and identify single fragments; and Così fan tutte, that instead was used to measure both fragments in coincidence [63].

Mass spectrometers as the ones mentioned above provide in principle only the mass of the FPs. If such a system is equipped with a detector able to discriminate the nuclear charge, not only chain yields, but also IFYs, can be provided. The nuclear charge is typically measured estimating the stopping power $dE/dx$ of the ions. This technique allows to identify single nuclei up to $Z = 47$ [7].

Since FPs are identified on an event-by-event basis, this opens the way to multi-parameter experiments, where the mass of the FPs are correlated with other observables (such as $\gamma$ or neutron multiplicities).

In recent years, a revival of the $2E-2\nu$ technique, based on the idea behind Così fan tutte, is under way for studies of neutron-induced fission [64].

This is the case, for example, of the STEFF (SpectromeTer for Exotic Fission Fragments) setup, where the FFs are identified, and the emitted prompt-$\gamma$ energy and multiplicity are measured in coincidence. Experimental runs have been performed at ILL and the setup is currently installed at the Experimental Area 2 flight path of $n$-TOF, at CERN. The first experiment has started with the $^{235}\text{U}(n_{th},f)$ reaction.

Other examples are the SPIDER (Spectrometer for Ion DEtermination in fission Research) spectrometer, built at Los Alamos Neutron Science Center (LANSCE) [65], the VERDI (VElocity foR Direct particle Identification) setup, developed at JRC-Geel [66], and FALSTAFF (Four Arm cLover for the Study of Actinide Fission Fragments), to be installed at the NFS facility (GANIL) [67].

Experiments in inverse kinematics

New programs are also being developed exploiting the availability of new heavy ions beams to perform experiments in inverse kinematics. In this con-
cept, a beam of actinides or pre-actinides impinges on a heavy mass target, where it fissions.

The great advantage of this technique is the possibility to study the fission of very exotic and/or radioactive actinides, that are usually impossible or very difficult to produce in amounts and purities suitable for a neutron-irradiation experiment.

The SOFIA (Study On Fission with Aladin) experiment at GSI [68] has started a campaign to determine the FF masses of several fissioning systems. The SOFIA setup consists of a recoil spectrometer, built around the existing large acceptance dipole ALADIN. The spectrometer allows the simultaneous identification of both FFs in coincidence, and the determination of quantities such as isotopic yields, total kinetic energies and prompt neutron multiplicities. In the most recent experiment on the $^{236}$U($\gamma$,f) system, fragment mass distributions were determined with a mass resolution of 0.6 and 0.8 u for the light and heavy fragment group, respectively [69]. There are plans to extend the choice of systems to be investigated, by using a long-lived $^{242}$Pu primary beam, that would give access to heavier actinides up to Americium [64].

At GANIL, VAMOS (VAriable MOde Spectrometer) has been used to identify one FF per fission event with a mass resolution below 0.8 u [70]. The fissioning system, that can be selected among different neutron-rich actinides from $^{238}$U to Cm isotopes, is characterised in terms of mass and excitation energy. In particular, fission of $^{250}$Cf at an excitation energy of 45 MeV (produced in fusion of $^{238}$U and $^{12}$C) has been achieved, and provided the first data on fully identified FFs, emitted in the de-excitation process of a specific CN with a well-defined excitation energy in this high-energy range [64].

1.2.5 Available data and uncertainties

Despite the fact that most of the FYs used in application are known to a level that makes safe operation of nuclear power plants possible, further measurements are needed for a variety of fissioning systems [23].

It is interesting to notice that even for thermal fission of $^{235}$U, one of the most studied reaction in terms of IFY, only 106 out of 998 individual yield ratios tabulated in the ENDF/B-VII.1\(^5\) evaluation have a relative uncertainty smaller than 10% (fig. 1.6) [5, 71].

The situation worsens for other fissioning nuclides and higher neutron energies, so that only four mass chains have been measured for $^{241}$Pu at high energies ($E_\pi = 14$ MeV) [72].

\(^5\)ENDF/B-VII.1 (Evaluated Nuclear Data Files) [5].
1.3 Fission Yields at IGISOL-JYFLTRAP

In the framework of the AlFONS (Accurate FissiOn data for Nuclear Safety) project, the IGISOL-JYFLTRAP facility at JYFL (Department of Physics, University of Jyväskylä), was proposed as the location for neutron-induced fission yield measurements of various actinides.

The University of Jyväskylä has a long history of measurements of nuclear data and nuclear physics-related quantities. In particular, the Ion Guide and Isotope Separator OnLine (IGISOL) technique has been applied to FY studies. Separation of FPs with the IGISOL technique is a standard method to select isobaric chains that can then be analysed with γ-ray spectroscopy for isotope identification. This is, however, very sensitive to the accuracy with which decay schemes of FPs are known (as I will discuss in chapter 3, presenting the results of our measurements). As a way to deal with this limitation, in the early 2000s, the possibility to couple the IGISOL technique with a high resolution mass spectrometer, the JYFLTRAP Penning trap, was investigated and tested at JYFL for proton-induced fission [73].

The measurement of the FPs proceeds as follows (numbers correspond to the elements of the IGISOL-4 beam line highlighted in the sketch in fig. 1.7):

1. Fission is induced in an actinide target inside the fission chamber with a proton beam.
2. A flow of helium gas is used to stop the FPs and guide them towards the extraction electrode. Helium is also used to recombine the highly ionised nuclides so that a large fraction of them reaches a +1 state. A time of the order of 100 ms is needed to extract the FPs from the ion guide, which is enough to washout any time structure of the proton beam, making the radioactive ion beam basically continuous.
The ion beam, where still all masses are present, is then accelerated with a voltage of 30 kV towards a dipole magnet. It is here that the first selection, based on the ionic charge-to-mass ratio $Q/A$, is performed. The mass resolving power of the dipole magnet is $M/\Delta M \approx 5 \times 10^2$, which is enough to isolate a single isobaric chain. After the selection by the dipole magnet, all ions have the same charge, typically +1.

The beam of isobars is then accelerated towards the Radio FreQuency (RFQ) cooler and buncher. This step is needed to prepare the beam for the analysis in the Penning trap. The ions are accumulated over a period of a few hundred milliseconds and, during this time, lose energy in the helium contained in the RFQ cooler and buncher. In this way, the energy spread of the beam is reduced down to a few eV. The FPs, that have travelled as a continuous beam from the fission chamber, travel as a bunch after the RFQ cooler and buncher. A short bunch and a low energy spread are important for an efficient capture in the Penning trap and to increase the precision of the measurement.

In the trap, a sequence of dipole and quadrupole excitations selects the nuclides based on their $Q/A$ ratio with a maximum mass resolving power of $\approx 8 \times 10^5$. This mass resolution is enough to select single nuclides based on their mass and, in some cases, also isomeric states of the same nuclide.

The selected ions are ejected from the trap and counted in a Micro Channel Plate detector. The mass-dependent quadrupole excitation frequency in the Penning trap is varied to cover the mass range of interest (usually corresponding to the isobaric chain selected by the dipole magnet), and
Figure 1.8. Example of a frequency scan obtained with the JYFLTRAP Penning trap at mass $A = 101$ in the p-induced fission of $^{232}$Th. The peaks of $^{101}$Zr and $^{101}$Nb have been identified. No peak is observed for $^{101}$Cd and $^{101}$Ag since these two isotopes are in the proton-rich half of the chart of nuclides and are not produced in fission. The position of the peaks as identified from the banner is calculated from a rough frequency-to-mass calibration. Where a peak is found, the banner also shows the number of counts in the peak (e.g., 7131 for $^{101}$Zr) and the number of times the scan was repeated to obtain enough statistics (64).

A frequency spectrum can be produced with each peak corresponding to a specific nuclide (see fig. 1.8).

The main advantage of this technique is the possibility to unambiguously identify single nuclides within a few hundred milliseconds from their production, thus obtaining IFYs also for short-lived FPs.

The selection of the nuclides with the dipole magnet and the subsequent measurement in the Penning trap are chemistry-independent, making it possible to measure any element in the periodic table.

The background for this measurement is very low, as can be seen in fig. 1.8, so that even nuclides with very low yield can be measured, making this a competitive technique for measurements of FPs in the tails or the valley of the FY distribution. The possibility to automatically scan masses with the dipole magnet and in the Penning trap also allows a systematic measurement of all masses of interest within the same experiment, and in a relatively short time.

The main difficulty is the estimation of the efficiency with which the FPs stop and recombine in the helium gas inside the fission chamber. It has to be verified that no bias is introduced depending on the mass, energy or chemistry.
of the FPs, so that the number of ions measured in the Penning trap can be assumed proportional to the ones produced in the fission reaction. This effect is currently being investigated, more details can be found in refs. [74, 75].

Another disadvantage with this technique is that it is not possible to measure more than one observable at the same time, since the FPs are not identified on an event-by-event basis, but their production is integrated over several hundred milliseconds before the yield is measured.

1.3.1 Neutron-induced fission yield measurements

The installation at IGISOL-4 of a high-current MCC30/15 cyclotron, opened the way to new possibilities for fission studies.

The relatively high output current of the cyclotron, allows the use of protons to produce neutrons that will then induce fission in an actinide target.

The installation of a neutron source not only benefits application-focused measurements of neutron-induced FY, but is also interesting for fundamental research. Very neutron-rich nuclei can be separated with the IGISOL technique and studied in the Penning trap, or by other techniques.

For the production of a neutron beam suitable for FY measurements, we designed, characterised and tested a proton-neutron converter. This will be the subject of the next chapter.
2. A Neutron Source for IGISOL-4

One of the requirements to measure neutron-induced Fission Yields (FYs) is an appropriate neutron source. It needs to be intense enough to ensure quick collection of data; produce a neutron field with a suitable energy spectrum, in order to provide valuable nuclear data; and be constructed with materials that guarantee mechanical stability under continued operation.

The acquisition by JYFL-ACCLAB (Accelerator Laboratory, University of Jyväskylä) of a high-current MCC30/15 cyclotron in conjunction with the upgrade to IGISOL-4, made the development of such a source, based on a (p,nx)-converter, possible.

I will discuss the constraints that influenced the design and the final geometry of the converter, a 5 mm thick beryllium disc cooled by water on the backside, in sect. 2.1.

We performed the first characterisation measurement of a prototype of the neutron source in June 2012 at the PAULA (Protons At Uppsala) facility of The Svedberg Laboratory (TSL) in Uppsala, Sweden [76]. We used two techniques: an Extended-Range Bonner Sphere Spectrometer (ER-BSS) and a Time of Flight (TOF) system with a liquid scintillator. I will show the results of the measurement in sect. 2.2.

The proton-neutron converter was installed at IGISOL-4 in March 2014 during the BRIGIT (BReakdown Counters at IGIsol Target) campaign. We used Thin Film Breakdown Counter (TFBC) detectors and activation foils to determine the total yield of the neutron source. This measurement is presented in sect. 2.3.

2.1 n-IGISOL: an overview

The IGISOL-3 facility at the University of Jyväskylä, Finland, began operation in 2003 and saw progressive improvements in the ion beam quality and transmission compared to the earlier setups. Numerous studies of nuclides far from stability were successfully undertaken in an extensive program
of nuclear structure, nuclear astrophysics, fundamental studies and applications [73, 77, 78]. It was then realised that, in order to reach even more unstable nuclei, the ion guide method would need to meet stringent requirements [77].

After the last IGISOL-3 experiment with JYFLTRAP in 2010, the whole experimental line was dismantled and relocated to a new hall. This allowed, among other changes aimed at improving ion transport, to couple an MCC30/15 cyclotron1 to the new beam line [77].

The MCC30/15 is designed to deliver 30 MeV protons at currents up to 100 μA. Once fully operational, it will be mainly supplying the IGISOL-4 experimental line. The maximum current of the MCC30/15 is higher compared to the K130 cyclotron, previously the main source of light and heavy ions at JYFL (Department of Physics, University of Jyväskylä). This makes the new cyclotron a feasible tool for delivering proton beams that can be converted into neutrons for neutron-induced fission studies.

Advantages of neutron-induced fission are twofold: (1) for nuclear data applications, it allows measurements of neutron induced FYs both via direct ion counting, using the JYFLTRAP Penning trap, or via γ-spectroscopy (which was used in the first successful neutron-induced fission campaign at IGISOL-4, that I will describe in chapter 3); (2) for studies of nuclei far from stability with JYFLTRAP, that is the driving force of the research activity at JYFL, shifting from proton- to neutron-induced fission is expected to boost the production yield of more neutron-rich nuclei [79].

2.1.1 How to build a neutron source: a wish-list

Considering the two main uses of the neutron source, specific considerations were put forward regarding the neutron energy spectrum, and the fluence.

- **Total neutron yield**: the first requirement, in terms of number of neutrons on target, is to have a fission rate comparable to the one achieved in proton-induced fission. In previous experiments, a reaction rate of $3 \times 10^9$ fissions/μC was reached.

- **Fast neutron fraction**: the yield of neutron rich nuclei far from the stability line is enhanced with fast neutrons. For this reason a goal of $10^{12}$ fast ($E_n > 1$ MeV) neutrons per second on the fission target was set. This value is also enough to ensure a competitive fission rate for fertile nuclei.

- **Neutron Spectra**: for studies of neutron-induced FYs for applications, the neutron spectrum should ideally resemble that of a typical fast reactor or of accelerator driven systems. For the sake of comparison with existing data and validation of the technique, a thermalised spectrum would also be desirable. The latter would also be useful for measurements of

1obtained from the Efremov Institute, St. Petersburg.
fission products with very low yield in the tails of the FY distributions of well-studied fissile targets, such as $^{235}$U. Finally, in order to extract the energy dependence of FYs, (quasi)mono-energetic neutron spectra would be ideal.

- **Heat removal:** at the maximum nominal current and energy of the MCC30/15, the proton beam will carry a power of 3 kW that will largely be deposited in the target in the form of heat. This heat needs to be removed, preferably with a simple water-cooling system, able to keep the target and all other elements of the proton-neutron converter well below their phase-change temperatures (melting or boiling points). An additional complication is the fact that, in order to limit the energy loss of the protons impinging on the target, the target must also act as a vacuum window between the beam line to the cyclotron and the fission chamber.

Trying to consider all the requirements mentioned above, we made a first selection on the materials suitable for a proton-neutron converter target. Looking in particular at the heat dissipation issue and the desire to avoid too tight margins on the operation of the neutron source, we chose to investigate tungsten and beryllium as two of the most promising materials, given their high melting points (3422 °C and 1287 °C, respectively) and high thermal conductivities (173 W/(m·K) and 200 W/(m·K) at 20 °C, respectively).

Monte Carlo simulations performed with FLUKA [80, 81] and MCNPX [82] were used to determine the best geometry in order to maximise the neutron yield. This resulted in full-stop targets for both materials. We also considered versions of the converter with an additional moderation stage, in order to produce thermal reactor-like spectra. The main findings of these early studies are summarised in refs. [83, 84]. In particular, we found that using beryllium instead of tungsten can increase the fast neutron flux with about a factor of seven, and the total neutron flux with about a factor of four. Regarding the cooling requirements, the higher thermal conductivity of beryllium would compensate for its lower melting point.

It should be mentioned that the MCC30/15 can not only deliver protons at 30 MeV, but also 15 MeV deuterons; the neutron yield per incoming deuteron at 15 MeV is larger than the yield for protons at 30 MeV. From design specifications, however, the deuteron current from the MCC30/15 is only half the one for protons, which reduces the effect of the improved yield. In addition to that, the lower energy of the beams that would be produced in the $^9$Be(d,nx) is expected to reduce the production of very neutron-rich fission products, which is one of the motivations for the development of this source at IGISOL-4 [79]. For these reasons, in the first phase of development of the source we chose to pursue the $^9$Be(p,nx) design, to have a more versatile source that can accommodate different needs.
Figure 2.1. Exploded view of the proton-neutron converter installed at IGISOL-4. 30 MeV protons (red arrow) impinge on a 5 mm thick beryllium disc (A, orange) cooled by water on the backside (B, cyan). A 3 mm thick aluminium plate (C, red) closes the target assembly and isolates the flowing water from the fission chamber. In this design, the beryllium disc is also used as a window to the vacuum of the beam line to the cyclotron (D, green). All dimensions are in mm. Figure courtesy of D. Gorelov

2.1.2 The final design

Further factors came into consideration once the final design had to be chosen. Learning from the experience of the Low Energy Neutron Source (LENS) developed for the University of Indiana Cyclotron facility [85–87], we pursued the idea of making the target thickness slightly less than the stopping range of protons. In the improved design, the protons go through the converter material and eventually stop in a layer of water placed on the backside of the converter.

There are two advantages that come with having protons stopping in the cooling water: the main effect is that no hydrogen accumulates inside the converter, thus avoiding the consequences experienced at LENS, where the beryllium target blistered due to hydrogen pressure building up inside the material [87]; in addition to that, having a fraction of the proton energy deposited directly in the cooling water greatly simplifies the heat removal.

The preferred design was then a 5 mm water-cooled beryllium converter, shown schematically in fig. 2.1.

A drawback of this design is that it results in a reduction of the neutron yield compared to a full-stop converter of approximately 5 - 10%.

Calculations show that about 10 MeV of the kinetic energy remain, after 30 MeV protons go through the converter. Several low-threshold Be(p,nx) reactions could contribute to an increased total neutron yield, if the protons were fully stopped inside the converter volume [88]. Production of neutrons in wa-
ter is not expected to make up for this loss, since the thresholds for $^{16}\text{O}(p,n\alpha)$ reactions is $>16.7 \text{ MeV}$.

We considered this an acceptable compromise, especially if the converter is used at the maximum current available from the MCC30/15 cyclotron.

**Heat Removal**

In order to verify that it is possible to ensure that all the elements of the converter are below their phase-change points, a 3-dimensional calculation using the finite element software COMSOL MultiPhysics\(^2\) was performed. According to this model, at the maximum nominal beam power of 3 kW, a water flow of 4 L/min would keep the water below 90 °C and the hottest regions of the beryllium disc around 500 °C.

The result of this calculation is shown in fig. 2.2: 20 °C water is flowing from top to bottom in the figure. Lines represent the water stream and colour coding is used to show the steady-state temperatures.

![Temperature (degC) Streamline: Velocity field](image)

*Figure 2.2. Results of the COMSOL MultiPhysics calculation: 3 kW of power are deposited in the beryllium converter and the cooling water. A flow of 4 L/min is enough to keep the water and the beryllium below their phase-change points. Distances are in cm, temperatures in °C.*

\(^2\)COMSOL Multiphysics v. 5.2., Comsol AB, Stockholm, Sweden.
Neutron Energy Spectrum

The energy spectrum of the field produced by the neutron converter was the object of intense studies, with the Monte Carlo technique. We used the MCNPX\(^3\) and FLUKA codes and compared their results.

The neutron energy spectrum obtained simulating the geometry used in the TSL characterisation, where 29.6-MeV protons impinge on a 5-mm thick beryllium converter, are presented in fig. 2.3. The neutrons, produced in \(^9\)Be(p,nx) reactions, are transported in the geometry and detected at a distance of 200 cm. I will describe the experimental setup in more detail in sect. 2.2.

To describe the \(^9\)Be(p,x) reactions, FLUKA uses internal models for the reaction cross-sections that are not accessible to the user.

In MCNPX it is instead possible to select nuclear data libraries: the simulations shown in fig. 2.3 uses the ENDF70PROT (for proton-induced reactions), the ENDF70 and the LA150 data libraries, based on ENDF/B-VII.0 evaluations. For those materials for which evaluated libraries are not available, internal models are used.

\[\text{Energy (MeV)}\]
\[
\begin{array}{cccccc}
10^{-1} & 10^{-2} & 10^{-3} & 10^{-4} & 10^{-5} & 10^{-6} \\
10^{-7} & 10^{-8} & 10^{-9} & 10^{-10} & 10^{-11} & 10^{-12} \\
\end{array}
\]

\[\text{dE (n/(sr C))}\]
\[
\begin{array}{cccccc}
10 & 20 & 30 & 40 & 50 & 60 \\
70 & 80 & 90 & 100 & 110 & 120 \\
\end{array}
\]

\[\Omega\]

\[\text{EdN/d}\]

\[\times 10^{15}\]

\[\times 10^{16}\]

\[\times 10^{17}\]

\[\times 10^{18}\]

\[\times 10^{19}\]

\[\times 10^{20}\]

\[\text{Energy (MeV)}\]

\[\text{MCNPX simulation}\]

\[\text{FLUKA simulation}\]

Figure 2.3. Results of the double-differential neutron yield from the 5 mm beryllium converter simulated with the MCNPX and FLUKA Monte Carlo codes.

The predictions of the two codes do not agree within the quoted uncertainties. In particular, MCNPX shows a harder energy spectrum with an enhanced high energy (>5 MeV) yield compared to FLUKA.

From further studies, it appears evident that the difference is not in the neutron transport, but rather in the production in the beryllium converter [89].

If sorting out this difference is important when the neutron source is used to produce very neutron-rich Fission Products (FPs), since different neutron spectra will produce different yields; it becomes crucial in the scope of nu-

\(^3\)The calculations with the MCNPX code presented throughout this thesis were performed by A. Solders.
clear data, where we need to know and report on the fissioning system under measurement, in order to provide high-quality data.

We therefore conducted a measurement at TSL to characterise the neutron source before its installation at IGISOL-4.

2.2 Characterisation measurement at TSL

The measurement on a mock-up of the converter was performed in June 2012, at the PAULA facility of TSL [76]. We used two different techniques to characterise the neutron source: a TOF system based on a liquid scintillator; and an Extended-Range Bonner Sphere Spectrometer (ER-BSS).

In this section I will discuss in more detail the experimental setup and the TOF system. More details on the characterisation of the neutron detector and on the analysis procedure can be found in ref. [88]. A description of the ER-BSS system and its results can instead be found in Paper I and in ref. [90].

2.2.1 Experimental Setup

We installed the 5 mm thick beryllium converter in a \((20 \times 20)\) cm\(^2\) aluminium support. In order to reproduce as accurately as possible the conditions in which the source would be used at IGISOL-4, we designed the holder with a 1 cm thick layer of water along the beam direction to resemble the water needed to cool the converter.

**Proton beam**

A 1.015 mm thick aluminium degrader reduced the initial energy of the \((37.3 \pm 0.5)\) MeV protons to obtain an energy comparable to the one available at IGISOL-4. The final energy of the protons was estimated to be 29.6 MeV at the Actual User Position (AUP), where the converter was placed [91].

Graphite collimators, with a \((10 \times 10)\) mm\(^2\) aperture, were used for a first shaping of the proton beam in the x- and y-directions. The size of the beam spot was finally determined by an additional collimator with a cylindrical aperture, 15 mm in diameter. The distance of the elements on the beam line up to the AUP, is shown in fig. 2.4.

The intensity of the proton beam was monitored in real time with a pair of thin scintillators (proton telescope), which detected protons scattered at an angle of approximately 45° off a stainless steel foil at the end of the beam pipe, upstream of the degrader and the collimators.

The telescope "count price", *i.e.*, the conversion factor between counts in the proton telescope (Monitor Units (MU)) and the number of protons on the beryllium converter, was provided by the TSL staff: \(1.89 \times 10^6\) protons/MU [92].
Neutron detector

The TOF setup consisted of a 3.3 litre BC-501 liquid scintillator from the NORDBALL array [93]. The scintillator was read out by a digital acquisition system based on an SP Devices ADQ412 high-speed digitiser. We extracted the TOF as the time difference between the signal coming from the scintillator, acting as a trigger, and a signal synchronous with the proton bunch, given by the radio frequency of the cyclotron. Both signals were recorded as digital pulses by the ADQ412 on an event-by-event basis.

The liquid scintillator was calibrated using multiple $\gamma$-sources, and its response function was determined with Monte Carlo calculations using Geant4 [94–96]. The calibration and more details of the Data AcQuisition (DAQ) are discussed in ref. [88].

The measurement with TOF focused on the high-energy region of the neutron spectrum, where the discrepancy between the two simulated neutron spectra is larger. It was important to obtain the best possible energy resolution up to 30 MeV, by measuring at the furthest possible distance. The frame-overlap, or wrap-around\(^4\) (WA), on the other hand, tends to make shorter distances preferable.

In order to optimise the energy resolution and minimise the effect of the WA, we collected data at three distances: 1.2 m, 2.1 m and 4.9 m.

Taking into account the uncertainty in the determination of the arrival time of the neutrons (due to the detector time resolution and the time spread of the proton bunch hitting the neutron converter) and the distance (mostly due to the unknown interaction point of the neutrons inside the sensitive volume of the

\(^4\)The WA is the superposition of flight times of the neutrons due to the repetition rate of a pulsed neutron source. This causes ambiguities in the determination of the energy, since a slow neutron may be mistaken for a fast neutron from a subsequent pulse.
scintillator), we estimated the uncertainty on the final energy as a function of the measurement position and energy (fig. 2.5).

In the data analysis, we implemented a pulse-height cut, with a different energy threshold for each distance, to remove the contribution of WA neutrons. We extracted the response function of the liquid scintillator to neutrons of different energies for each pulse-height threshold level. The result of this study is shown in fig. 2.6 and is explained in greater depth in ref. [88]. The uncertainties plotted in fig. 2.6 are dominated by the uncertainties on the light output functions [88]. They give an idea of the error on the response function, when we compare counts acquired at the three different thresholds.

A realistic estimate of the uncertainty on the absolute value of the efficiency can only be obtained looking at the difference between simulated and experimental data, if the efficiency is measured directly. We did not perform a direct measurement of the efficiency for our detector; in ref. [88], however, we used the experimental efficiency data measured by Drosg [97] and compared it with the results from a model of that experimental setup, using the same procedure used to extract the curves in fig. 2.6. From this comparison, we estimated the uncertainty on the absolute efficiency for our experiment to be of the order of 20%.

We then combined the results from the measurements at the three different distances over the energy range between 6 and 30 MeV, by scaling for the change in solid angle covered by the detector. We used runs with shadow cones to subtract the contribution of scattered neutrons.
Figure 2.6. Scintillator detector efficiency curves for the thresholds used in the TOF measurement at a distance of 1.2 (black), 2.1 (red) and 4.9 m (green). The uncertainties are indicated with a shade of the same colour.

2.2.2 Results

Figure 2.7 shows the energy spectrum above 5 MeV obtained with the TOF technique compared to the MCNPX and FLUKA calculations. Included are also the spectrum measured with the ER-BSS and the result of a TOF measurement by Johnsen [98] on a similar setup. To normalise the ER-BSS and TOF measurements we used the count price from the proton-telescope.

It is worth noticing that in fig. 2.7, unlike fig. 4 in Paper I, all spectra are in absolute scale. Also, the error bars on the TOF spectrum also accounts for the uncertainty originating from the estimation of the absolute efficiency of the liquid scintillator.

2.3 The BRIGIT campaign: installation and total yield measurement at IGISOL-4

The converter was first installed at IGISOL-4 in spring 2014. After that, a validation measurement, the BRIGIT campaign, was performed with TFBCs (Thin Film Breakdown Counters) using the TOF technique.

The main objectives of this characterisation measurement were: (1) to obtain a realistic estimate of the neutron flux at the fission target position, and (2) to verify that the spectra measured at TSL were compatible with what is mea-

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Johnsen’s measurement was performed at the the University of California, Davis in 1978. In his experiment, 30 MeV protons impinged on a 1.27 cm thick full-stop beryllium converter.
Figure 2.7. Comparison of the neutron energy spectra from the TSL measurement with the TOF and the ER-BSS, and Monte Carlo simulations in FLUKA and MCNP and Be(p,nx) data measured by Johnsen [98].
Figure 2.8. Schematic view of the working principle of TFBC detectors. An electric field is applied between the two electrodes: this is sufficient to induce a breakdown once a heavy ion from the fission target creates free charges in the dielectric layer. The high current generated in the process will produce a detectable signal.

2.3.1 Experimental Setup

Thin Film Breakdown Counters are detectors sensitive to highly ionising particles. As the name suggests, TFBCs exploit the phenomenon of electric breakdown in a semiconductor induced by heavy ions. A TFBC is built as a sandwich of different layers (as shown in fig. 2.8): two electrodes are interleaved with a dielectric layer.

An electric field is applied between the two electrodes: this is sufficient to induce a breakdown once a heavy ion from the fission target creates free charges in the dielectric layer. The high current generated in the process will produce a detectable signal and - at the same time - evaporate a portion of the electrode, interrupting the short-circuit. The portion of the electrode that evaporates will not be able to detect any further ions, slightly reducing the efficiency of the detector [100, 101].

If a fissionable material is placed on the face electrode, heavy ions can be produced in a fission reaction as fission products, making it possible for TFBCs to detect neutrons, as in the measurements described in refs. [102, 103].
Figure 2.9. Experimental setup used during the BRIGIT campaign. The proton beam enters from the bottom-right corner of the picture and the proton-neutron source is attached to an extension of the cyclotron beam line. TFBC detectors are positioned at a distance of 25.5 cm from the backside of the converter in the direction of the beam.

The count rate (R) of the TFBC thus depends on the detector efficiency $\tilde{\varepsilon}$ (which, in turn, is determined by its active surface and by how many events it has already detected), and on the number of fission events per unit time. The latter depends on the target applied to the TFBC detector (the fissionable material used and its thickness $\rho$, in g/cm$^2$), as well as on the neutron flux $j$ and energy spectrum. These factors are summarised in equation 2.1:

$$R = \tilde{\varepsilon} \frac{N_{av}}{A} \frac{j}{\sigma_F},$$

(2.1)

where $N_{av}$ is Avogadro’s number, $A$ is the mass number of the target, and $\sigma_F$ is the spectrum-averaged cross section that depends on the target’s fission cross section and the neutron energy spectrum.

The neutron flux can be extracted from equation 2.1, as:

$$j = \frac{R \cdot A}{\tilde{\varepsilon} \cdot \rho \cdot N_{av} \cdot \sigma_F},$$

(2.2)

We positioned the TFBC at a distance of 25.5 cm downstream of the converter; the centre of the TFBC was located at 1.2 cm and 3.2 cm to the right and top, respectively, from the centre of the beryllium disc.
We used an 80 μg/cm² isotopically pure $^{238}$U foil as the TFBC fission target. The detector was irradiated for 907 s at a proton beam current of 2.1 μA. The efficiency $\tilde{\epsilon}$ of the TFBC was measured to be $(0.176 \pm 0.009)$ cm².

### 2.3.2 Results

Because of the short flight-path of the neutrons between production and detection, we chose to compare the result of this measurement in terms of TFBC counts in the TOF-frame. This reduces the uncertainty that would be introduced if the spectrum measured with the TFBC had to be corrected for the energy-dependent response function.

We then convoluted the neutron energy spectra calculated with Monte Carlo codes, or measured with other experimental techniques, with the TFBC response function ($\epsilon(E)$). Including the information on the integrated proton current ($Q$), we converted the incident fluence at the irradiation position ($\Phi(E)$) into the number of events that would be detected by a TFBC exposed to such a neutron field ($TFBC_{eq}$):

$$TFBC_{eq}(E) = Q \cdot \Phi(E) \cdot \epsilon(E).$$

(2.3)

The result of the MCNPX calculation were extracted as a spectral neutron fluence in TOF, so that it could be directly compared to experimental data, after including the corrections mentioned above. We had instead to convert the other spectra from the energy- to the TOF-frame in the position of the TFBC. A Gaussian spread, added on the simulated spectra, accounted for the time resolution of the TOF system (estimated to 1.7 ns FWHM).

Figure 2.10 shows the result of the TOF measurement with the TFBC, compared to the activation measurement and the Monte Carlo simulations of the same experimental setup, in the TOF-frame. The spectrum from the TSL characterisation is also shown.

### 2.4 Discussion

In presenting the results of our characterisation campaigns, we compared the measurements of the neutron fields performed at TSL and JYFL despite the different techniques and conditions used in the measurements (e.g., background, neutron scattering, and distance from the source). We, in fact, expect to see the most prominent effect of these differences in the lower energy range, a portion of the spectrum that is of limited interest for application of the source in neutron-induced fission of fertile nuclei (like $^{238}$U and $^{232}$Th), which are targets commonly used at JYFL.

From the results in fig. 2.7, we see that the shape of the TOF measurement at TSL is well reproduced by the MCNPX simulation. Also in absolute scale,
considering the uncertainties on the determination of the absolute efficiency, the TOF spectrum is best described by MCNPX, though the measurement appears to over-estimate the total yield. This difference can be due to additional systematic errors, e.g., on the count price measured by the proton-telescope, that could not be accounted for. The TOF result is also in reasonable agreement with the measurement by Johnsen, performed on a similar setup. The difference in shape observed above 25 MeV can be attributed to the different energy resolution of the two detectors (approximately 3 MeV at 30 MeV for Johnsen’s [98] vs 0.8 MeV of our system). Note that, in fig. 2.7, the energy resolution of Johnsen’s data is not included in the error bars, that only represent the uncertainty on the differential neutron yield attributed by Johnsen to the efficiency calibration [98].

The spectrum measured with TFBCs at JYFL (fig. 2.10) agrees better, even in absolute scale, with the MCNPX simulation. The MCNPX-simulated neutron flux also reproduces well the result of the activation measurement [79,99].

The inconsistency of the TOF data from the TSL campaign is not entirely surprising, and reflects the over-estimation of the MCNPX neutron spectrum I discussed for fig. 2.7. While the shape of the curve does indeed reproduce the one measured with TFBCs (see also fig. 5 in Paper I), the offset in the absolute scale could be explained, also in this case, with a systematic error, e.g., on the telescope count price, whose uncertainty was not included in the error bars.

It can therefore be concluded that the MCNPX code describes sufficiently well the high-energy region of the neutron energy spectrum from the $^9$Be(p,nx) reaction, while FLUKA underestimates it.
Having a code that has been successfully benchmarked against experiments is very beneficial if minor modifications are introduced to the converter design: in that case we would not need to repeat the same extensive characterisation, and a realistic estimate of the neutron flux will come directly from Monte Carlo calculations. This is what happened in the first campaign of FY measurements at JYFL, where instead of the 5 mm version of the converter characterised in this work, we used a thicker full-stop design. This will be described in more detail in chapter 3.

**Thermal Flux**

In both characterisation campaigns, estimating the thermal component of the neutron flux has never been the focus of attention.

In the TSL measurement, the minimum energy detected with the TOF setup was 6 MeV; the thermal peak detected with the ER-BSS (see Paper I) was instead dominated by the scattering of neutrons in the experimental hall.

In addition to that, it is reasonable to assume that the thermal and epithermal components of the neutron spectrum would be highly dependent on the amount and type of material surrounding the neutron detector. For this reason, had the measurement at TSL been able to estimate the thermal fraction, it still would not have been directly transposable to the conditions at IGISOL-4.

In the BRIGIT campaign, on the other hand, neither technique (*i.e.*, TOF with TFBCs or activation foils) was able to measure thermal neutrons due to the thresholds of the selected reactions ($^{238}$U(n,f), $^{27}$Al, $^{58,59}$Ni, $^{115}$In, $^{209}$Bi [79]). At this stage, the best estimate we have of the thermal fraction, comes from Monte Carlo calculations and is presented in tab. 2.1.

In the future, a direct measurement of the thermal contribution needs to be performed for the source to be used to measure high-quality nuclear data. In fact, even in the measurement of FYs of fertile nuclei, available targets are usually contaminated with traces of other nuclides.

This is the case, for example, of the measurement performed in December 2016 and described in chapter 3, where we used a $^{nat}$U target.

<table>
<thead>
<tr>
<th>Energy Range</th>
<th>FLUKA (%)</th>
<th>MCNPX (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\leq$1 eV</td>
<td>0.26 ± 0.15</td>
<td>0.211 ± 0.001</td>
</tr>
<tr>
<td>1 eV – 1 keV</td>
<td>1.72 ± 0.06</td>
<td>1.297 ± 0.003</td>
</tr>
<tr>
<td>1 keV – 1 MeV</td>
<td>31.32 ± 0.02</td>
<td>18.668 ± 0.014</td>
</tr>
<tr>
<td>1 MeV – 10 MeV</td>
<td>46.06 ± 0.02</td>
<td>52.423 ± 0.021</td>
</tr>
<tr>
<td>$\geq$10 MeV</td>
<td>20.63 ± 0.02</td>
<td>26.641 ± 0.016</td>
</tr>
</tbody>
</table>

Table 2.1. Percentage contribution from different energy ranges to the total neutron spectra simulated using FLUKA and MCNPX for the IGISOL-4 setup.
2.5 Conclusion

In this chapter, I have described our efforts to design and characterise a neutron source for neutron-induced fission yield studies.

The source is based on the Be(p,nx) reaction and will use 30 MeV protons from the high-current MCC30/15 cyclotron installed at the IGISOL-4 facility at JYFL. The neutron yield was calculated with the Monte Carlo codes FLUKA and MCNPX; we verified that a water cooling loop at 4 L/min is enough to ensure thermal stability under continued operation at the maximum available current delivered by the MCC30/15.

We performed a characterisation measurement at TSL, Uppsala, where we produced neutrons by irradiating a mock-up of the proton-neutron converter with 29.6 MeV protons. The energy spectrum was measured with two techniques: an ER-BSS to measure from thermal energies up to 30 MeV; and a TOF system, to focus on the high-energy part of the spectrum (above 6 MeV).

The neutron field produced by the final design of the neutron source was also measured at JYFL during the BRIGIT campaign, with TFBC detectors and activation foils. These results are in reasonable agreement with the MCNPX simulation and confirmed that the shape of the energy spectrum is compatible with the one measured in the first characterisation at TSL.

Overall, we conclude that three different measurements (TOF with liquid scintillator, TOF with TFBCs and activation foils), as well as the earlier measurement by Johnsen, support the harder neutron-energy spectrum suggested by MCNPX.

According to the measured fission rates in the TFBCs at JYFL (more details on the calculation are discussed in ref. [88]), a total neutron flux between 2 and $5 \times 10^{12} \text{n/sr/s}$ can be obtained with this setup, at an incoming proton current of 100 μA. Of these, between 2 and $3 \times 10^{12} \text{n/sr/s}$ are fast neutrons ($E_n > 1 \text{ MeV}$). Both these numbers fulfil the design goal in terms of total and fast neutron flux and allow neutron-induced fission studies at IGISOL-JYFLTRAP.

Dwelling on the past . . .

After the first results of the calculations showed the discrepancy between the two Monte Carlo codes, we realised that a characterisation of the neutron source was needed, before the converter was installed.

We started planning for the measurements at TSL in the fall of 2011. Now, six years later, we know of the things that could — and should — have been done differently. Not only for what concerns the experiment at TSL, but also for the BRIGIT campaign.

An updated plan for TSL

If I was asked, today, to plan the characterisation campaign at TSL, these are some of the things I would consider:
A **cleaner source**: one of the big, insurmountable, difficulties of the TSL campaign at PAULA was the initial proton energy. The energy of the protons had to be lowered from 37.3 MeV to about 30 MeV. In the approach we followed, a 1.015 mm aluminium slab and about 1.4 m of air did the job. This caused, however, a very large beam divergence, that in turn had to be remedied by the multi-stage collimation described in sect. 2.2.

From both measurements and Monte Carlo simulations, it was shown that many neutrons reaching the detector were produced in the collimators. This, in the end, did not invalidate the result of the TOF measurement, thanks to the shadow cone runs used to estimate and subtract the background. However, placing the last cylindrical collimator a few tens of centimetres upstream the converter and reducing its aperture would have reduced the background contribution at the measurement position.

A **real-time current monitor at the converter position**: one additional difficulty at PAULA coming from the need to degrade the beam, was the fact that it was hard to estimate the absolute current impinging on the beryllium disc. The final value, only recently obtained from TSL, required an intense effort from the staff at the laboratory, and has an uncertainty that is hard to quantify. An alternative monitor, or an easier geometry for the calibration, would have been beneficial.

A **reference detector**: we spent a large part of the time for the analysis of data from the TOF system on the estimation of the response function of the liquid scintillator. The modelling of the light output, the choice of the best libraries to describe the interaction of neutrons on carbon (especially above 20 MeV), the comparison with cases in which the efficiency was measured directly, etc.

Despite these efforts, we were still left with an uncertainty on the absolute neutron flux of about 20%. The most realistic and feasible way to reduce this uncertainty could have been to perform a reference measurement using a detector with a known response function, such as a TFBC. TFBCs, have a limit on the maximum rate that would make a complete characterisation with this technique very time-consuming. A reference measurement, however, would have allowed to rescale the absolute neutron spectrum with less uncertainty and much less effort.

A **better DAQ**: a series of problems with the digital DAQ software made it difficult to associate the counts in the detector to the real-time proton-telescope counts, in order to correct for the initial current. The problem with the DAQ was solved after the run, and a similar software was used in some of the measurements of the BRIGIT campaign.

**BRIGIT-3**

Some issues were also encountered in the BRIGIT campaigns. The first and most successful campaign is the one we reported on in Paper I and in sect. 2.3.
The second, that was intended to improve statistics and use several fission targets, did not provide conclusive results. A summary of the measurements and the results is reported in ref. [104].

The main problem in that case was that, unlike in the first measurement, we were forced to use the K130 cyclotron instead of the MCC30/15, which was under repair. The older K130 cyclotron has a time-width of the proton pulse that is too large for a good time resolution at such a short distance. This makes it unsuitable for the kind of TOF measurements we performed with TFBCs.

...and dreaming of the future

At this stage, it is not thinkable to repeat a measurement similar to the one done at TSL, but rather measure the neutron spectrum in situ, at IGISOL-4.

In a future BRIGIT-3 campaign, the objective should be a characterisation of the thermal component of the neutron field and a better determination of the total yield.
3. Neutron-induced Fission Yields Measurement at IGISOL-4

The IGISOL facility in JYFL-ACCLAB (Accelerator Laboratory, University of Jyväskylä), has been used in the past for measurements of proton-induced Fission Yields (FYs) with the JYFLTRAP Penning trap [78, 105].

Following the installation of the neutron source, in 2015 we made several attempts to measure neutron induced FYs, but it was not until December 2016 that we successfully determined relative yields from neutron-induced fission of $^{235}$U, in the first systematic measurement at the upgraded IGISOL-4 facility. During this campaign (n-IGISOL), we measured the yields using $\gamma$-spectroscopy.

In sect. 3.1, I will briefly discuss what we did and what we learned in the first attempts. I will then present, in sect. 3.2, the experimental technique, the setup and the detector characterisation used in the latest n-IGISOL campaign. In sect. 3.3, the data analysis and the results will follow, which I will discuss and comment on in sect. 3.4.

3.1 The first neutron-induced Yield Measurement Campaigns

After the construction of the converter and a series of characterisation measurements, it was finally time to put the neutron source to good use.

Many unforeseen (and unforeseeable) delays, however, built up along the way: the MCC30/15 cyclotron, installed in 2010 and successfully used in 2014 for the BRIGIT campaign (described in sect. 2.3), suffered a series of technical glitches that made it, de facto, unusable. All the measurements that followed were then performed using protons delivered by the K130 cyclotron.

The main difference between the two cyclotrons, besides the maximum available proton current, that in the case of the K130 is about an order of magnitude lower, is availability. The MCC30/15 is essentially dedicated for use at the IGISOL-4 beam line. The K130 cyclotron, instead, serves the entire JYFL-ACCLAB, resulting in a very busy schedule and limited beam-time available for development and tests.
3.1.1 Spring 2015: first tests

The first experiments to measure neutron-induced FYs of natU were performed in April and June, 2015; 30 MeV protons were delivered at an intensity ranging from 1 to 11 μA. The measurements are discussed in ref. [79], where it is also concluded that:

The intensity of any mass separated beams after the dipole magnet was so low that it was impossible to transport the beam towards the spectroscopy station and perform on-line measurements. (D. Gorelov. PhD Thesis (2015) [79])

The presence of Fission Products (FPs) was confirmed with an off-line γ-spectroscopy analysis of an implantation foil positioned before the mass-separating dipole magnet. The delay between the implantation and the spectroscopy measurement, as well as the absence of any chemical or physical separation of the FPs, made it impossible to obtain any useful yield distributions.

The measurement allowed, however, to estimate the fraction of FPs extracted from the ion guide:

Due to waiting time of about 18 hours before the γ-measurement the isotopes $^{135}$Xe, $^{143}$Ce, $^{133}$I and $^{97}$Zr with shorter half-lives were chosen to calculate extraction efficiency. In all these cases the fraction of isotopes observed on the aluminium foil after the extraction from the ion guide is about $\approx 0.01\%$. [...] In comparison a typical total efficiency for proton induced fission ion guide is about 10 time larger. (D. Gorelov. PhD Thesis (2015) [79])

3.1.2 Lessons learned

Realising the loss in extraction efficiency, most likely attributable to the different geometry of the fission targets in the ion guide between proton- and neutron-induced fission [79], we started planning a new campaign for December 2016. Not being able to rely on the MCC30/15, all the other parameters that could improve the product yields with a reasonable effort, were optimised:

- **Source-to-Target distance**: the easiest way to improve, not the efficiency, but the number of FPs, is to increase the fission rate. This can be done by increasing the solid angle covered by the fission targets. We thus placed the ion guide as close as possible to the proton-neutron converter assembly.

- **Amount of fissionable material**: exploiting the fact that neutrons diffuse over a much larger volume than protons, the fission rate can be also increased by increasing the area of the fissionable targets. The ambition was to use a large natU target (25 cm$^2$) that, however, was not obtained in
time for the experiment. Instead, we used two $(1 \times 5) \text{cm}^2$ natU targets, which were also employed in the earlier runs.

- **Beam line transport optimisation**: one of the difficulties of a neutron-induced fission run is that the low current of radioactive ions extracted from the fission ion guide makes it problematic to tune the parameters of the beam line. For this reason, a 3-day proton-induced fission run was scheduled right before the run with neutrons, in order to optimise the extraction and transport of the radioactive beam.

- **Neutron source geometry**: in order to achieve the maximum possible fission rate, we also changed the neutron source. A 1 mm beryllium disc was added after the 5 mm converter, effectively changing the design to a full-stop target. After validating the MCNPX code, we used Monte Carlo simulations to estimate the neutron energy spectrum and neutron yield at the fissionable target position. This new geometry, besides a slight increase of the total flux, also reduces the activation in the cooling water. Hydrogen build-up was not considered a serious danger at these low currents.

3.1.3 Let’s get real...

Once the major bottlenecks were identified and worked on, we set realistic objectives for a first systematic study of neutron-induced FYs. The possibility to study FPs in the Penning trap was ruled out: despite the increased fission rate, transport of the ions to the RFQ cooler and buncher and the trap, even with a total efficiency of 60 $\%$, would risk another unsuccessful campaign.

It was decided, instead, to identify the products as close as possible to the mass separation step. The beam line was therefore equipped with a tape-implantation station positioned after the switch-yard. We used an HPGe detector, operated in $\beta$-coincidence mode to suppress the background, in order to collect $\gamma$-rays from the mass-separated FPs. The aim was a systematic study in the mass range $A = 128 - 133$, where FPs have convenient half-lives and decay radiation, as well as a reasonably high yield.

3.2 Experimental Method

Figure 3.1 shows a schematic view of the beam line as it appeared during the neutron-induced fission run. Circled numbers mark the elements of the setup for beam extraction and transport; lower/upper case circled letters are the detection stations for offline/online analysis, respectively.

30 MeV protons from the K130 cyclotron impinged on the 6 mm thick beryllium converter (①). The resulting white neutron field was used to induce fission in two $(1 \times 5) \text{cm}^2$ and 15 mg/cm$^2$ natU foils (②) located at a distance
of 2.5 cm from the back of the neutron source, and installed in a specifically
designed ion guide [99].

The FP stopped in helium gas at a pressure of 400 mbar (with about 4% stopping efficiency [106]) and were extracted from the ion guide gas cell using a SextuPole Ion Guide (SPIG) (3) [107] and differential pumping. The ions were then accelerated to 24 keV and separated online based on their charge-over-mass ratio using a dipole magnet (4) with a mass resolving power $M/\Delta M$ of about 500.

Mass-separated ions were then transported to the tape-implantation station (5), at the end of a dedicated beam line.

We monitored the neutron energy spectrum and its intensity with a series of activation foils attached to the outer wall of the fission ion guide (a). We used removable aluminium implantation foils (b), positioned on the back of the $^{235}$U targets, to measure the accumulated activity of the FPs over the entire run. A titanium implantation foil (c), placed on the interior wall of the fission ion guide, facing the fissionable targets, monitored the activity of the FPs that escaped the targets, but did not stop in the helium buffer gas. Fission products were also accumulated at an implantation station before mass separation (d): these represent the ions that successfully stopped in the helium buffer gas and were extracted by the SPIG, and can include several ionisation states. This is the same position as the one used in 2015 [79].

We have not yet analysed the data from the implantation foils b-d, but these will eventually be valuable in validating the simulation of the stopping efficiency of the fission ion guide developed by A. Al-Adili, K. Jansson, et al. [74, 75, 106].

Silicon detectors (E and F) were used to monitor the activity of the ion beam before and after mass-separation. The $\beta$-activity of the mass-separated FPs was detected with a plastic scintillator (G) surrounding the movable implantation tape. We also used this detector as a trigger for the detection of the characteristic $\gamma$-rays from the FPs with the HPGe detector (H).

3.2.1 The neutron source and the fissioning system

As mentioned in sect. 3.1, we used a slight modification of the neutron source characterised in the TSL and BRIGIT campaigns. After the validation measurements, MCNPX was elected the code of choice to obtain the reference neutron spectrum in the fissionable target position.

The Monte Carlo calculation, whose result is reported in fig. 3.2, shows that the total yield obtained with a full-stop converter is $1.8877(6) \times 10^9$ n/cm$^2$/$\mu$A, approximately 8% larger than with the 5 mm configuration. The shape of the neutron energy spectrum does not change appreciably between the two geometries.
Figure 3.1. Schematics of the beam line used in the neutron-induced fission run (n-IGISOL) in December 2016. Circled numbers mark the elements of the setup for beam extraction and transport; lower/upper case circled letters are the detection stations for offline/online analysis, respectively. A detailed description of all elements is provided in sect. 3.2.
Using the MCNPX simulated spectrum in fig. 3.2, we were able to deduce some information on the fissioning system. The isotopic composition of \( ^{nat}U \) is in large majority \(^{238}U\) (99.3\%); \(^{235}U\) represent the remaining 0.7\%, but can become important in virtue of its high fission cross section, reaching about 580 b at thermal energies. Convoluting the neutron energy spectrum with the two cross sections, we found that fission of \(^{238}U\) accounts for (98 ± 1)\% of the total fission events. The average energy weighted with the fission cross-section of \(^{238}U\) is (12.4 ± 8.8) MeV.

The energy of the incoming neutrons is so high that there is a large probability of emitting one or more neutrons before the compound nucleus undergoes fission. We used the GEF (GEneral Fission) [34] code and the MCNPX simulated spectrum to estimate the probability of multi-chance fission for \(^{238}U\), resulting in about 55\%, 31\%, 8\% and 6\% for first, second, third and fourth chance fission, respectively.

GEF was also used to estimate the FYs (cumulative and independent) from fission of \(^{nat}U\). To do so, GEF yields were calculated for various incoming neutron energies (31 values for \(^{238}U\) and 47 for \(^{235}U\)), distributed between 0 and 30 MeV. The yields for each energy were then weighted with the fission rate, obtained as the convolution of the neutron energy spectrum with the nuclides’ fission cross-section, and summed. In sect. 3.3, we will compare the results of this calculation with our experimental data.
3.2.2 The HPGe detector

The detector used for the γ-spectroscopy measurement is a Canberra GC7020 70% coaxial p-type HPGe detector, with an energy resolution at 1332.5 keV of 3.1 keV (vs a nominal resolution of 2 keV).

We operated the detector in coincidence with a plastic scintillator coupled to a photomultiplier tube. The scintillator surrounded the implantation tape, and was used to detect the electrons from the β−-decay of the FPs.

A full-energy peak efficiency calibration was done using an 152Eu source. Data were acquired for 900 s (live time) and the areas of the characteristic γ-lines were extracted using FitzPeaks [108]. The efficiency $\varepsilon_i$ can be obtained as:

$$\varepsilon_i = \frac{A_i}{I_i(\gamma)} \quad (3.1)$$

where $A_i$ is the integrated number of counts of the peak at energy $i$, $I_i(\gamma)$ is the intensity of the γ-line at energy $i$.

We used the function

$$\ln(\varepsilon) = a + b \cdot \ln \left( \frac{E}{7} \right) + c \cdot \ln^2 \left( \frac{E}{7} \right) + d \cdot \ln^3 \left( \frac{E}{7} \right) + e \cdot \ln^4 \left( \frac{E}{7} \right) \quad (3.2)$$

to interpolate the data-points.

In eqn. 3.2, $\varepsilon$ is the relative detection efficiency, and $E$ the energy of the photons. The best values of the parameters obtained from the fit are:

- $a = -29.0 \pm 0.2$
- $b = 11.89 \pm 0.05$
- $c = -1.501 \pm 0.007$
- $d = (3.51 \pm 0.09) \times 10^{-2}$
- $e = (2.4 \pm 0.1) \times 10^{-3}$
- $f = (8.3 \pm 1.2) \times 10^{-1}$

The result of the fit, along with the efficiency data-points, is shown in fig. 3.3.

3.2.3 Data Acquisition procedure

A γ-spectrum was acquired for each mass between $A = 128$ and $A = 133$. In order to reduce contamination from different mass chains, the implantation tape was moved after modifying the settings of the dipole magnet (i.e., every time a new mass chain was selected).

Data were measured for 7200 s, except for mass $A = 133$ for which the spectrum was acquired for 3200 s.

Before starting the acquisition, FPs were accumulated on the tape for times ranging between 60 s and 15 min. This was done in order to get as close as possible to the saturation condition for the nuclides that dominate the γ-spectra.
In most mass chains, we could not recognise more than 2-3 elements from the pulse-height spectrum. In some cases, we could distinguish isomeric states based on their characteristic $\gamma$-lines. The spectrum acquired for mass $A = 130$ is shown, as an example, in fig. 3.4, where some of the most prominent peaks are labelled with the nuclide they correspond to.

In tab. 3.1, I present an overview of the nuclides identified in the measurement. The colour-coding qualitatively shows how clearly each nuclide was identified in the spectrum, from red (‘not seen’), to green (‘easily recognisable’). A white box is used when the nuclide (isomer) was not expected. Given the overall picture outlined in tab. 3.1, we focused our attention only on those elements for which a more systematic study could be attempted: tin (Sn) and antimony (Sb).

The peaks were identified based on the information retrieved from nuclear data libraries. Besides the energy, the most important information necessary to extract the integrated activity over the measurement time is the intensity of each $\gamma$-transition.

In the analysis, we selected the peak positions based on the $a$-priori knowledge of the energy of the strongest $\gamma$-rays of the nuclides of interest extracted from the nuclear data tables (hence the low counts - compatible with background - in some of the peaks). Corrections for the detector efficiency were included using the function in eqn. 3.2. In ref. [109], we report all the $\gamma$-ray energies, intensities, integrated counts and detector efficiencies used to extract the yields reported in the following. Corrections for the acquisition time (growth and decay of radionuclides) were added in a subsequent step, once the integrated activity was extracted for all nuclides.
Figure 3.4. Typical spectrum from the $\gamma$-spectroscopy measurement during the n-IGISOL campaign. The spectrum refers to mass $A = 130$ and some of the most prominent peaks are identified.

Table 3.1. Qualitative overview of the nuclides identified in the $\gamma$-spectroscopy measurement. The colour-coding shows how clearly each nuclide was identified in the spectra, from red = 'not seen', to green = 'easily recognisable': a white box is used when the nuclide (isomer) was not expected. HL and IT in a red box are used to mark nuclides that were not identified because of the half life and the decay through internal transition, respectively.

<table>
<thead>
<tr>
<th>Element</th>
<th>Mass Nr.</th>
<th>128</th>
<th>129</th>
<th>130</th>
<th>131</th>
<th>132</th>
<th>133</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cd-m</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cd</td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>In-m</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>In</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sn-m</td>
<td></td>
<td>IT</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sn</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sb-m</td>
<td></td>
<td></td>
<td>IT</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sb</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Te-m</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Te</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>I-m</td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>I</td>
<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
</tr>
</tbody>
</table>
When more than one peak was visible for a certain nuclide, we used the weighted average to obtain a value for the integrated activity:

$$\text{avg}_w = \frac{\sum_i A_i (1/\sigma_i^2)}{\sum_i (1/\sigma_i^2)},$$  \hspace{1cm} (3.3)

where \(A_i\) is the activity extracted from the \(i\)-th peak and \(\sigma_i\) its uncertainty.

Peaks common to more than one nuclide (typically, ground and isomeric states of a nuclide) were discarded from the analysis.

**From nuclide activity to fission yields**

If the precursors of the nuclide under analysis have reached the saturation condition when the measurement starts, we can extract the integrated activity \(A_{\text{int}}\), over the measurement time \(T\) as:

$$A_{\text{int}} = \int_T P R \cdot \left(1 - e^{-\lambda t}\right) \, dt = P R \cdot \left[T - \frac{1 - e^{-\lambda T}}{\lambda}\right],$$  \hspace{1cm} (3.4)

so that we can calculate the production rate \((PR)\), or Cumulative Fission Yields (CFYs), as:

$$PR = \frac{A_{\text{int}}}{T - \frac{1 - \exp(-\lambda T)}{\lambda}}.$$  \hspace{1cm} (3.5)

where \(\lambda\) is the decay constant of the studied nuclide.

Equation 3.5 also allows to extract CFYs for longer-lived nuclei, without any knowledge of the yield of the precursors, as long as these precursors have significantly shorter half-lives. The assumptions needed to derive eqn. 3.4 are satisfied for all isotopes of Sn, and eqn. 3.5 was used to extract the CFYs.

In the analysis of Sb data, eqn. 3.4 is not always valid, given the longer half-lives of the precursors (up to 59.1 min in the case of \(^{128}\text{Sn}\), decaying to \(^{128m}\text{Sb}\)). However, we could use the CFYs measured for Sn and subtract the contribution of precursors to the yield of Sb, thus extracting Independent Fission Yields (IFYs).

### 3.3 Results

Following the procedure outlined in sect. 3.2.3, we obtained the relative yields for five isotopes of Sn (128 to 132) and six isotopes of Sb (128 to 133). The yields are plotted in fig. 3.5. For five nuclides – three isotopes of Sn and two of Sb – we observed both ground and isomeric states. In these cases, we plot the sum of the two yields. Detailed results of the observed Isomeric Yield Ratios (IYRs) are presented in tab. 3.2.
Figure 3.5. Relative CFY of Sn (a) and Relative IFYs of Sb (b) produced in high-energy neutron-induced fission of natU. Solid symbols are used to report the sum of the isomeric and GS. The half-filled symbol (●) is used for $^{128}$Sn, where only the GS was seen. Open symbols (○) are used for $^{128}$Sb and $^{129}$Sb, where only the isomeric states were observed due to the longer half-life of the GS.
Table 3.2. IYRs measured in this experiment, compared to evaluated data libraries for fission of $^{238}$U induced by 14 MeV neutrons ($^{238}$U($n_{14}$,f)) and to GEF calculations weighed with the simulated neutron spectrum. IYRs are reported as $Y_{IS}/(Y_{IS} + Y_{GS})$. The values reported are cumulative IYRs and independent IYRs for Sn and Sb, respectively.

<table>
<thead>
<tr>
<th>Z</th>
<th>$J^\Pi_{IS}$</th>
<th>$J^\Pi_{GS}$</th>
<th>$E_{IS}^*$ (keV)</th>
<th>This Work</th>
<th>GEF</th>
<th>ENDF B-VII.1</th>
<th>JENDL</th>
</tr>
</thead>
<tbody>
<tr>
<td>129Sn</td>
<td>$^{(11/2)^-}$</td>
<td>$(3/2)^+$</td>
<td>35.15(5)</td>
<td>0.37(13)</td>
<td>0.63</td>
<td>0.15(12)</td>
<td>0.68(43)</td>
</tr>
<tr>
<td>130Sn</td>
<td>7$^-$</td>
<td>0$^+$</td>
<td>1946.88(10)</td>
<td>0.64(48)</td>
<td>0.49</td>
<td>0.48(49)</td>
<td>0.70(46)</td>
</tr>
<tr>
<td>131Sn</td>
<td>$^{(11/2)^-}$</td>
<td>$(3/2)^+$</td>
<td>65.1(3)</td>
<td>0.43(19)</td>
<td>0.69</td>
<td>0.48(45)</td>
<td>0.81(51)</td>
</tr>
<tr>
<td>130Sb</td>
<td>$(4,5)^+$</td>
<td>8$^-$</td>
<td>4.80(20)</td>
<td>0.81(43)</td>
<td>0.68</td>
<td>0.43(41)</td>
<td>0.43(28)</td>
</tr>
<tr>
<td>132Sb</td>
<td>8$^-$</td>
<td>4$^+$</td>
<td>200(30)</td>
<td>0.25(10)</td>
<td>0.49</td>
<td>0.61(41)</td>
<td>0.61(20)</td>
</tr>
</tbody>
</table>

3.4 Discussion

The FPs that reach the tape-implantation station for $\gamma$-spectroscopy are only a fraction of the ions produced in the fission reaction. Losses occur in recombination processes in the fission ion guide, during extraction and transport, and finally in the mass selection in the dipole magnet. Also, of the ions decaying on the tape, some are not detected because of the efficiency of the scintillator used for the $\beta^-$-coincidence condition.

The exact transport efficiency of the setup is not known experimentally. Based on previous experience, we can safely assume that the efficiency does not change within the same atomic number and does not change significantly for neighbouring masses. We were not able to refer the measured activities to the absolute number of fissions, hence we report only relative yields in fig. 3.5. The sum of the yields from ENDF/B-VII.1 (extracted for 14 MeV neutron-induced fission of $^{238}$U) and of the ones from the GEF calculations (weighed with the simulated neutron spectrum) were rescaled to the sum of the measured ones.

3.4.1 Charge States

The Ion Guide and Isotope Separator OnLine (IGISOL) technique is able to produce all chemical elements and is considered element-independent [79], but the total transport efficiency will ultimately depend on the $Z$ of the FPs. The mechanisms of ion recombination of the FPs down to charge +1, the one we generally choose to select with the dipole magnet, in fact, depend on the chemistry of the produced elements and on the impurity concentration in the helium stopping gas [110].

We investigated in detail the ratio of singly- and doubly-charged ions at $A = 132$. The results show that the ratio of the production rates of the two charge states $PR(+1)/PR(+2)$ goes from $1.19 \pm 0.17$ for $^{132}$Sn to $2.78 \pm 0.42$ for
$^{132}\text{Sb}$. This suggests that comparing absolute values of the production rates of the two elements is not relevant, until a more complete understanding of the element-dependent ion recombination in the ion guide is reached.

Nevertheless, we can (and should) subtract the CFYs of Sn in order to obtain IFYs for Sb. We can in fact assume, based on the half-lives, that most of the decays of Sn to Sb happen on the tape, i.e., after the extraction from the ion guide and the θ/λ selection by the dipole magnet.

### 3.4.2 Tin

The precursors of Sn in all mass chains have a half-life shorter than 1.5 s, allowing us to use the approximations discussed in sect. 3.2.3. The $7^-$ isomer of $^{128}\text{Sn}$ decays through internal transition with a probability of 100% and it was not visible due to the $\beta$-coincidence condition used in this experiment.

The cumulative yields of Sn exhibit a staggered behaviour around mass $A = 131$, that is neither observed in the ENDF/B-VII.1 evaluated data library, nor in the GEF calculation.

We should say, however, that decay information for $^{131}\text{Sn}$ was not available in ref. [111] and incomplete in ref. [112]. Also, in ref. [112], no conclusive decay data were found to discriminate between $^{131}\text{Sn}$ and $^{131m}\text{Sn}$; this information comes instead from ref. [113]. This fragmented information may have caused an incorrect estimation of the activity.

The large uncertainty at $A = 130$ is dominated by counting statistics. For $^{130m}\text{Sn} (7^-)$, the only visible peak is at 898.5 keV, that – with an intensity of 8% – produces only $31 \pm 19$ counts in the peak, in a measurement time of 7200 s.

### 3.4.3 Antimony

We were not able to see the ground states of $^{128}\text{Sb}$ and $^{129}\text{Sb}$, due to their half-lives of 9.01 h and 4.4 h, respectively. The isotopes of Sb show an increasing trend from masses $A = 128$ to 132, that contradicts the one seen in the ENDF/B-VII.1 library. Our data seem instead to confirm the calculations performed with the GEF code, that present a distribution peaked as many as two mass units above the one from ENDF/B-VII.1.

### 3.4.4 Isomeric Yield Ratios

IYRs are another important quantity that can be extracted from fission: they help in determining the average angular momentum of the highly excited fission fragments after scission. They also represent an important test for fission models and are of interest, e.g., for experiments related to antineutrino spectra generated by nuclear reactors [114].
The study of isomeric yields would require a dedicated and extensive work, that is outside the scope of this measurement and of this thesis. A new method to extract the Fission Fragments (FFs) angular momenta is currently being developed by V. Rakopoulos and the first results were recently presented in ref. [115]. In this section, I will only comment and compare our results to evaluated data libraries and the GEF model.

In particular, what catches the eye in tab. 3.2, is the large discrepancy that exists among the different models. In this case we also include the data from the JENDL (Japanese Evaluated Nuclear Data Library). What we learn by studying the evaluated data (even besides the few reported here), is that IYRs in data libraries are approximate values, that present very large uncertainties and that give, in some cases, only an estimate of this quantity. Experimental data that can nail the evaluations are in fact very scarce, especially at higher energies.

Our data, though overlapping with ENDF/B-VII.1 and JENDL (partly due to their enormous uncertainties), follow only slightly the values extracted from GEF.

What we can say, however, is that, by looking at the angular momenta of the nuclei in their isomeric and ground states, we see some consistency that gives us confidence in the truthfulness of these results. This is especially true for the $^{130}\text{Sb}/^{132}\text{Sb}$, and $^{129}\text{Sn}/^{131}\text{Sn}$ pairs. The first two nuclides have opposite angular momenta assignment between the ground and isomeric states, and result in reciprocal IYRs. The second, with the same angular momenta, have IYRs that are compatible within uncertainties.

It is worth pointing out that, in some of these cases (i.e., $^{129}\text{Sn}$, $^{131}\text{Sn}$ and $^{130}\text{Sb}$), these IYRs are not accessible in measurements with JYFLTRAP, because of the low excitation energy of the isomeric state.

3.4.5 Proton current & other uncertainties

We did not record the proton current as a function of time throughout the run. A monitor, based on the charge collected on the beryllium converter, did however provide this value, that was manually logged during the experiment. From these recorded values, we can claim that the current remained stable within $\approx 5\%$ during the neutron-induced campaign, around an average of $10\,\mu\text{A}$. This uncertainty is not included in the error bars of fig. 3.5.

A sudden reduction of the proton current could easily explain the staggered behaviour in proximity of $^{131}\text{Sn}$. However, if this was the case, we would expect a reduction also of the yields of $^{131}\text{Sb}$, which was not observed.

Other uncertainties we did not include in the error bars are those on the intensities of the $\gamma$-lines.
3.5 Summary & Outlook

In this chapter, I presented the first results of neutron-induced FY measurements at the IGISOL-4 facility. We used a white neutron field with an average energy of $(12.4 \pm 8.8)$ MeV to induce fission in a $^{238}\text{U}$ target. Relative CFYs of five Sn isotopes and relative IYFs of six Sb isotopes were extracted using the IGISOL technique and $\gamma$-spectroscopy. IYRs of five nuclei were also measured.

CFYs of Sn show a staggered behaviour with what seem to be odd-even effects around mass $A = 131$. This behaviour is neither reproduced by the ENDF/B-VII.1 evaluated data libraries, nor by the GEF model. For the IYFs of Sb we find a mass distribution peaked around $A = 132$, in agreement with the trend from GEF, but about two mass units above the peak observed in the ENDF data library.

This campaign was the crowning achievement of a work that lasted more than 6 years, and included the development and characterisation of the neutron source, as well as few previous attempts at the determination of the yields. Besides its intrinsic significance (highlighted in sect. 3.4), I cannot help but think of it as a successful proof-of-principle, and another important stepping stone towards a more comprehensive study of the yields, using the MCC30/15 cyclotron and JYFLTRAP.

3.5.1 A plan for studies of Fission Yields at IGISOL-4

The upgrade of the IGISOL-4 facility is now complete, and the JYFLTRAP Penning trap has already been extensively tested and used in numerous experimental campaigns. Hiccups with the MCC30/15 cyclotron, still not conclusively resolved, remain today the only big technical limitation for neutron-induced FY studies.

During this time, we have gained experience in the analysis of Penning trap data and refined our understanding of the IGISOL technique. With this knowledge, we can outline a realistic plan of measurements, that will eventually lead to the promised high-quality nuclear data that we have been chasing for all these years.

**Determining the ion extraction efficiency**

One of the main obstacles for the extraction of IFY distributions is the $Z$-dependent ion recombination in the ion guide. In the latest measurement at IGISOL-4, in December 2016, along with the study of proton- and neutron-induced FYs, we also measured the activity of FPs at different locations along the extraction line. These data have not yet been analysed, but they will eventually offer reference values to adjust and validate the simulations of the slowing-down and ion recombination of the FPs in the ion guide.
A conclusive answer, however, will probably only come with a reference measurement. Measuring FYs with JYFLTRAP for a well known case, such as the reaction $^{252}\text{Cf}(sf)$, will provide efficiency factors for all elements and masses, that we will be able to use in the measurement of other compound nuclei.

A comparison of the FP yields extracted from JYFLTRAP with previous high-quality measurements is probably the first stop towards a full characterisation of the facility for FY studies.

**Exploring the Chart of Nuclides**

Once the mechanisms of ion recombination in the ion guide, as well as the extraction and mass selection efficiency of the dipole magnets are understood, the first studies of new mass yield distributions can start.

The first fission experiment will likely be fission of $^{238}\text{U}$ induced by a neutron field with a spectrum similar to the one used for the experiment in December 2016, and characterised in the campaigns described in chapter 2.

The availability of a large target and the intensity of the source, makes this an ideal case to test the capabilities of neutron-induced fission in the facility, even if the MCC30/15 was still not available or not working at its full potential. The data from the JYFLTRAP Penning trap will extend and update the ones obtained from $\gamma$-spectroscopy (presented in this chapter and in Paper IV), that were contingent upon (sometimes incomplete) decay data.

A successful $^{238}\text{U}$ campaign would pave the way to studies on one of the targets already available at JYFL (e.g., $^{232}\text{Th}$), or any other fissionable material that can be produced in a sufficient amount (e.g., $^{235}\text{U}$).

Thinking a few years ahead, a very interesting improvement for the facility would be to explore fission induced by different incoming neutron spectra. By reducing the thickness of the neutron converter, as it was reported in ref. [116], or by changing the initial proton energy, we could start studying the energy dependence of the yields.
4. **DEℓFIN**: a code for the De-Excitation of Fission Fragments

While working on the developments for the measurement of neutron-induced Fission Yields (FYs), it became obvious that fission models had to be included in the discussion, in order to interpret our results.

We then started considering what model(s) to choose and how to judge their reliability in regions for which experimental data are scarce or non-existent, as is the case for the high-energy fission we intend to study at IGISOL-4.

The large part of the models that nowadays can successfully calculate fission observables, mostly provide a phenomenological description of some parts of the fission process. As I discussed in chapter 1, often these models focus on defining the properties of the Fission Fragments (FFs) after scission. Model parameters in the codes can be tuned, depending on the compound nucleus and the available energy, in order to reproduce experimental knowledge.

In this chapter, I describe a methodology that is used to study the models performances and their respective assumptions. The method, that resulted in the development of the **DEℓFIN** (De-Excitation of Fission Fragments) code, detaches the modelling of the FFs state at scission from the de-excitation, that is needed to produce measurable observables.

This work - summarised in Paper II and III - is far from complete, and a series of extensions would be needed to allow for a more thorough study of different observables. As of today, it focuses on the average prompt neutron multiplicity distributions, $\bar{\nu}(A)$.

After having introduced the most important available codes for the simulation of fission observables in chapter 1, I will start the discussion with an overview of our approach to comparison and validation in sect. 4.1. This was implemented in the **DEℓFIN** code, that I will present in sect. 4.2. The first results of the comparison will be shown in sect. 4.3 and discussed in sect. 4.4. Finally, I will present some conclusions and an outlook, in sect. 4.5.

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The human world - it’s a mess. Life under the sea is better than anything they got up there.

Sebastian, *The Little Mermaid*
4.1 A new approach to comparison and validation

From an experimentalist’s point-of-view, it is often desirable to have models that can be compared to freshly obtained experimental results. Sometimes, these same models are instead needed at an earlier stage, in the analysis and interpretation of data (e.g., the estimation of fission chances at a specific $E_{CN}^\ast$).

Then the wide landscape of available codes that can be used to calculate fission observables opens up. The choice sometimes becomes a question of whether the code is freely downloadable, and possibly easy to run, without deeper considerations on the specific models assumptions.

How can we really understand and compare different models? While using different assumptions to describe the state of FFs at scission, all the models generally seem to be able to reproduce experimental data. This can be a confirmation of their trustworthiness, but can also be due to the fact that the parameters in the codes are tuned to the same experimental data used to benchmark them, i.e., there can be compensatory effects. In addition to that, what can be compared are fission observables: neutron emission rather than $E^\ast$, Isomeric Yield Ratios (IYRs) rather than the angular momentum of the FFs, etc.

Eliminating any variability in the way the final observables are extracted could help highlight the differences in the models assumptions and their consequences on fission quantities.

In this work, we detach the FFs de-excitation step from the models, and we handle it in a consistent way for all cases. The quantities defined right after scission by the model (typically the mass, charge and excitation energy of the FFs), are provided as input to the TALYS reaction code [117], that we use to carry out the de-excitation of the FFs and to extract the desired observables.

We bundled the preparation of the TALYS inputs, starting from the $E^\ast$ distributions calculated by the models under test, with the extraction of the final observables in a single code: $\text{DE}/\text{FIN}$.

We developed the work in various steps. The first phase made extensive use of the GEF code [118] to validate the methodology behind $\text{DE}/\text{FIN}$. In a second phase, we performed the actual comparison of the models.

Prompt neutron emission distributions, $\tilde{\nu}(A)$, are one of the first quantities we studied in this work. We considered them a good starting point to gain an overall perception of the capabilities of the method, since $\tilde{\nu}(A)$ is a quantity tightly related to the excitation energy provided as input, and that has been measured in many independent experiments.

$\text{DE}/\text{FIN}$ was also used to extract other quantities from the TALYS output. We performed tests for IYRs, but we soon realised that: (1) very few codes model the fission process at a sufficient level of detail to provide $J(A,Z,E^\ast)$ and $FY(A,Z)$, quantities needed for a meaningful comparison among the codes; and (2) experimental data are scarce and sparse, so that benchmarking codes to experiments can only be done for specific nuclides, often missing the big picture.
4.1.1 Why TALYS?

TALYS is a code for the simulation and reproduction of nuclear reactions. It was developed by Arjan Koning and co-workers at Petten, and the first version was launched in 2004 [119].

TALYS can simulate proton, neutron and particle-induced reactions in the 1 keV-200 MeV energy range, for nuclides heavier than $A = 12$; it includes numerous nuclear reaction models, that can be accessed from a single interface. Since its launch, TALYS has been increasingly used in fundamental as well as applied nuclear physics, and it is at the core of the TALYS Evaluated Nuclear Data Library TENDL [120].

The reason for choosing TALYS-1.8 as a tool to be implemented in DE/FIN not only has to do with the general trustworthiness of its models, but also with the fact that it is available, extremely well documented and open source.

Its transparency makes it very easy to include in a larger project that relies on its output. TALYS also allows the user to have full control on the reaction parameters. It should be noted that, for the use made of the code in this phase of the development of DE/FIN, we have always kept the default model parameters unchanged.

One of the key technical features of TALYS, that makes it perfect for this kind of application, is the possibility to run de-excitation of a compound nucleus using the Hauser-Feshbach model embedded in the code [41, 119]. This can be done providing TALYS with a matrix of $(E^*, J)$ distributions of the chosen compound nucleus (the primary FF, in our case).

4.1.2 Why GEF?

The GEF code, developed by K.-H. Schmidt, is — as already mentioned — one of the main fission codes available.

It is a Monte Carlo code, which allows to obtain information, even-by-event, on the FFs. Like TALYS, GEF is also well documented and open source.

It can simulate proton-\(^1\), neutron- and compound nucleus fission of a wide range of heavy nuclei from $A \gtrsim 180$, taking also multi-chance fission into account.

Contrary to the majority of the other codes, GEF does not require any experimental input information (e.g., on the yields as is the case of FREYA, or $TKE$ as for PbP). This makes it a self-contained tool, that can be used to simulate also fissioning systems for which experimental data are incomplete or non-existent.

\(^1\)Starting from the latest version, 2016/v1.2
4.2 DE\textsubscript{FIN}: idea and implementation

The principle followed in the development of DE\textsubscript{FIN} is transparency: we tried not to make arbitrary assumptions on the quantities needed in the calculations. The process is completely reproducible and — as mentioned before — the default values of the physics parameters in the TALYS code were not modified. If a value needed for the calculation is not provided by the models themselves (e.g., FF yields, \(Z(A)\) distributions, \textit{etc.}) we turn to well-known systematics, such as Wahl’s \cite{43}.

DE\textsubscript{FIN} can read simple input files with information on the FFs coming from any model. It needs the FF excitation energy \(E^*\) and, if available, the spin. These values can be averaged over mass (\(E^*(A)\)), or on a nuclide basis (\(E^*(A,Z)\)). The de-excitation of the FFs is then performed in TALYS-1.8.

The principle is schematically shown in fig. 4.1: in order to obtain \(\bar{\nu}(A)\), DE\textsubscript{FIN} runs a TALYS calculation for each of the FFs in an isobaric chain with mass \(A\). The number of prompt neutrons \(\bar{\nu}(A)\) is obtained by averaging the number of neutrons emitted by each FF in the chain.

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**Figure 4.1.** Schematics of the methodology used to extract \(\bar{\nu}(A)\) distributions: a TALYS calculation is run for each FF (red box) in an isobaric chain. The production cross-section of the fission products (green boxes) is used to determine the probability of emission of 1, 2, …, \(n\) neutrons. An average weighted with the relative isobaric yield \(Y(Z|A)\) allows to extract \(\bar{\nu}(A)\). The same procedure is then repeated for all the isobaric chains of interest.

DE\textsubscript{FIN} automatically repeats the procedure for all isobaric chains of interest. Information on the relative charge yield of each fragment for a given mass \(A\), \(Y(Z|A)\), is needed. If the mass yields \(FY(A)\) of the isobaric chains are also known, DE\textsubscript{FIN} provides the total average neutron multiplicity \(\bar{\nu}\).
4.2.1 Validation of the methodology using the GEF code

Not all codes for the simulation of fission observables provide the same amount of information on the FFs. Deterministic codes often give only quantities averaged over a mass-chain; but even most Monte Carlo codes do not provide an output as detailed as the one available from GEF. In order to validate the methodology, it was necessary to demonstrate that a comparison between codes that provide different amount of information was fair and sound.

The first step in our study was then to evaluate how much the input could be simplified \((i.e.,\) how little information would be required from the model, for instance by taking the average \(E^*\) instead of the whole distribution), before the results would start deviating considerably from the original calculation.

![Graph](image)

*Figure 4.2.* Validation of the methodology using the GEF code for the reaction \(^{235}\text{U}(\text{nth},\text{f})\) reaction. Details of all calculations are discussed in the text.

The GEF code proved to be a valuable tool in this phase, for its ability to provide extensive information on the FFs on an event-by-event basis. We performed a validation of the methodology comparing the results of DELFIN calculated using inputs with less and less information. The result of this sequence of calculations for the \(\bar{V}(A)\) of \(^{235}\text{U}(\text{nth},\text{f})\) is shown in fig. 4.2, where:
(a) GEF | E_distrib(A,Z): is the $\tilde{\nu}(A)$ distribution obtained running the standalone\(^2\) GEF code.

(b) DE\(/\text{FIN} | E_distrib(A,Z): is the $\tilde{\nu}(A)$ distribution extracted from a calculation where we provide the complete $E^*$ distribution as input. The yields were obtained directly from the GEF code.

(c) DE\(/\text{FIN} | E(A,Z): as the previous step, but instead of the entire $E^*$ distribution, we used only the average of the distribution as input.

(d) DE\(/\text{FIN} | E(A,Z_{\text{Wahl}}): as the previous step, but instead of the yields from GEF, the $Z(A)$ distribution was obtained from the Wahl systematics. The mass chain yields are obtained from GEF.

(e) DE\(/\text{FIN} | E(A_{\text{Wahl}},Z_{\text{Wahl}}): as the previous step, but we extract both $Z$ and $A$ yields from the Wahl systematics. This step justifies the use of a simple $E^*(A)$ distribution, without any information provided by the model on the $Z$ of the FFs. This is the last simplification that we can do before the results are no longer comparable to the ones obtained using all the information provided by the model.

(f) DE\(/\text{FIN} | E(A): in this case DE\(/\text{FIN} runs only one TALYS calculation for each isobaric chain $A$. We select the charge of the FFs based on the Unchanged Charge Distribution model. Comparing this curve with the curve in case (b), we concluded that this simplification is too crude to produce acceptable results for our study.

This extensive testing of the methodology proves that the use of the average excitation energy for the isobaric chain (cases (c) to (e)) gives results that are in very good agreement with those obtained using the full distribution of excitation energy $E^*_\text{distrib}(A,Z)$, provided that several FFs in a mass chain are included in the calculation.

In the calculation in case (e), we included all the isobars for which Wahl’s systematics predicts a non-zero yield in the averaging process. Using only one isotope per mass chain (as it was done in case (f)) produces very staggered data points that do not reproduce the original $\tilde{\nu}(A)$ distribution.

In addition to this, the generally good agreement between TALYS’ de-excitation and GEF standalone, reassured us on the choice of model parameters for the FFs used in the TALYS reaction code.

**A word of caution: $E^\text{rot}$**

The GEF code calculates the excitation energy $E^*$ available at scission for each of the FFs as the sum of different contributions, reported in eqn. 1.5. After scission, the FFs relax to a less deformed shape and the deformation energy $E^\text{def}$ will convert into intrinsic energy [35, 118].

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\(^2\)In the following of the chapter and in the figures, I will talk about the "standalone" codes when discussing the results obtained directly from the codes, without the use of DE\(/\text{FIN}.
The rotation energy $E_{\text{rot}}$, however, will not be available to the FFs statistical excitation and thus for de-excitation through neutron emission. $E_{\text{rot}}$ will instead have an important role in the emission of non-statistical photons.

In order to take this into account, we modified the original event-by-event output of GEF and subtracted $E_{\text{rot}}$ from $E^*$ before the excitation energy distributions were used in $\text{DE}^{\text{FIN}}$. These (very minor) changes in the source code of GEF are reported in Appendix B.

4.3 Results

Here follows an overview of the results obtained from a variety of sets of $E^*$ processed using the methodology described above. The plots presented in this chapter are very dense of information, which makes them hard to read, at times. For a less compact representation of the results, I refer the reader to appendix A.

The sets of excitation energies $E^*(A)$ from the models were found in the literature, obtained from the authors by private communication, or by running the codes directly.

In addition to the GEF, FREYA and PbP codes studied in Paper II, we were kindly given access to sets of $E^*$ from CGMF and FIFRELIN, that are presented in Paper III.

It is not surprising that all the fissioning systems tested in this phase are coming from the most studied reactions ($^{235}\text{U}(n,f)$, $^{239}\text{Pu}(n,f)$ and $^{252}\text{Cf}(sf)$), as many of these codes use input from experimental data.

4.3.1 $^{235}\text{U}(n_{\text{th}},f)$

The thermal neutron-induced fission of $^{235}\text{U}$ was studied using $\text{DE}^{\text{FIN}}$ with FREYA, PbP, FIFRELIN and GEF. The results, shown in fig. 4.3, are compared to the Wahl systematics and to the $\bar{\nu}(A)$ distributions from the standalone codes (refs. [28,32,121]). The mass yields from ref. [122] are shaded behind the $\bar{\nu}(A)$ curves, and were used to calculate the total $\bar{\nu}$ and the $\bar{\nu}$s for the light and heavy fragments ($\bar{\nu}_{\text{LF}}$ and $\bar{\nu}_{\text{HF}}$, respectively) reported in tab. 4.1.

Information on the FF yields does not come directly from FREYA, FIFRELIN and PbP, but was extracted from the Wahl systematics. The GEF code does indeed provide information on the independent fragment yields, but for consistency we used the same yields as in the other cases. As I discussed in sect. 4.2.1 and showed in fig. 4.2, this choice does not affect the final result significantly.
$\bar{\nu}(A)$ distributions from the $^{235}\text{U}(n_{th},f)$ reaction, as obtained from DE$\ell$FIN using input from GEF, PbP, FIFRELIN and FREYA. DE$\ell$FIN results are compared to the distributions from the standalone codes and to the Wahl systematics. The mass yields from ref. [122] are shaded behind the $\bar{\nu}(A)$ curves.
4.3.2 $^{239}$Pu(n$_{th}$,f)

The results for thermal neutron-induced fission of $^{239}$Pu are shown in fig. 4.4 for the GEF, FIFRELIN and FREYA codes. We compare the calculations from $\text{DE/FIN}$, to the standalone codes (refs. [28, 40]) and to the Wahl systematics. Also in this case, the FY mass distribution shaded behind the $\bar{\nu}(A)$ curves was obtained from ref. [122], and was used to calculate the $\bar{\nu}$s in tab. 4.1.

4.3.3 $^{252}$Cf(sf)

The $\bar{\nu}(A)$ for spontaneous fission of $^{252}$Cf is shown in fig. 4.5 for GEF, FIFRELIN and CGMF. The results of $\text{DE/FIN}$ are compared to the standalone codes (refs. [40, 123]) and to experimental data from ref. [124]. The FY mass distribution shaded behind the $\bar{\nu}(A)$ curves was also obtained from ref. [124]. In addition to using the Wahl systematics, CGMF was also run with the $Z(A)$ distributions obtained from the code itself. The calculation in $\text{DE/FIN}$ is then labeled “DE/FIN with CGMF (FY CGMF)” in fig. 4.5.

4.4 Discussion

In the work for this thesis, we studied five of the most used models for the simulation of fission observables: GEF and FIFRELIN (for all reactions), FREYA (for $^{235}$U(n$_{th}$,f) and $^{239}$Pu(n$_{th}$,f)), PbP (for $^{239}$Pu(n$_{th}$,f)), and CGMF (for $^{252}$Cf(SF)). In this section, I will outline the main observations we can make from the $\bar{\nu}(A)$ distributions.

Since this is a study on the models, rather than the specific reactions, I will try to generalise, and present some remarks for each code.

Before we proceed, it is important to mention how to (and not to) interpret the results from this study. As described, in $\text{DE/FIN}$ the FFs de-excitation step is detached from the model codes and handled using TALYS. This implies that the probabilities of neutron emission, in competition with $\gamma$ de-excitation in each of the simulated FFs, comes from the parametrisation used in TALYS. The parameters in the code have not been optimised and it is not in the scope of this work to try to reproduce experimental data by changing them. Therefore, when commenting on the results from $\text{DE/FIN}$ (for the $\bar{\nu}(A)$ distributions in fig. 4.3-4.5, or for the total $\bar{\nu}$ in tab. 4.1), we are studying the difference with the standalone codes, rather than comparing $\text{DE/FIN}$ to experiments or systematics.

4.4.1 GEF

The hump observed in the GEF-standalone $\bar{\nu}(A)$ of $^{235}$U (fig. 4.2), $^{239}$Pu (fig. 4.4) and $^{252}$Cf (fig. 4.5) around mass $A = 140$ is not found in experimental
Figure 4.4. $\nu(A)$ distributions from the $^{239}$Pu(n,f) reaction as obtained from DELFIN using input from GEF, FIFRELIN and FREYA, compared to the results of the standalone codes and to the Wahl systematics. The mass yields from ref. [122] are shaded behind the $\nu(A)$ curves.
Figure 4.5. $\bar{\nu}(A)$ distributions from $^{252}$Cf(SF) as obtained from DELFIN using input from GEF, CGMF and FIFRELIN compared to the results from the standalone codes and experimental data (ref. [124]). The mass yields from ref. [124] are shaded behind the $\bar{\nu}(A)$ curves.
data. Neither is it reproduced when the same excitation energy distributions are processed in \texttt{DE/FIN}. This suggests that this is a peculiarity of the de-excitation process built into GEF rather than a consequence of the $E^*$ partition at scission.

The same is not true for the double-humped structure around the low-mass peak of $^{235}\text{U}$ and $^{239}\text{Pu}$, that appears to be a direct consequence of the $E^*$ distribution. The shape is present not only in GEF, but in most of the models, although the positions of the humps and the minima differ. This structure is not replicated in experimental data, however uncertain these might be in proximity of the symmetric fission mass range due to the low yield.

In general, except for these two peculiarities that are worth exploring further with the code’s developers, the agreement between \texttt{DE/FIN} and the standalone version of GEF is very good. We notice a slight over-estimation of the $\bar{\nu}(A)$ from \texttt{DE/FIN} for all fissioning systems around $A = 100$ and $A > 155$. This, however, could be the result of the choice of parameters in TALYS, and cannot undoubtedly be attributed to features of the GEF model.

It is worth reminding that the rotational energy of the fragments has been subtracted by the $E^*$ provided by GEF. The agreement between \texttt{DE/FIN} and GEF also suggests that the fission model is able to estimate the fraction of rotational energy with rather good accuracy. This should yield, e.g., a rather good estimate of the $\gamma$ de-excitation with non-statistical photons, the ones responsible for most of the angular momentum loss by the FF.

4.4.2 CGMF

The study of CGMF with \texttt{DE/FIN} was limited to $^{252}\text{Cf}$. Given the close affinity between the de-excitation process implemented in CGMF and the one used in TALYS, we did not expect large changes.

In addition to the standard calculation performed in \texttt{DE/FIN} for all the other codes, CGMF also provides $FY(Z,A)$ distributions. This study is similar to the one we used to validate the methodology with GEF (discussed in sect. 4.2.1), and the overall consistency between the two treatments is confirmed also in this case. The only significant differences appear in the vicinity of mass $A = 122$ (inset in fig. 4.5). This region corresponds to mass chains with a very low yield, so that the Monte Carlo nature of CGMF can lead to an inaccurate estimation of the $FY(A,Z)$.

Compared with the standalone code, \texttt{DE/FIN} consistently, though only slightly, overestimates the $\bar{\nu}(A)$. This is enough, however, to cause the total $\bar{\nu}s$ to differ by about 0.3 neutrons (see tab. 4.1).
4.4.3 FIFRELIN

FIFRELIN, like TALYS and CGMF, treats de-excitation of FFs with the Hauser-Feshbach model. On the other hand, as shown in eqn. 1.11, the excitation energy provided for each of the FFs is the sum of both intrinsic and rotational energy.

Since DE/FIN uses the whole E* as available energy for de-excitation through neutron emission this, unsurprisingly, yields a substantial over-estimation of the $\tilde{V}(A)$ all over the mass range. The only exception is seen in proximity of mass $A = 132$, where fragments are nearly spherical.

Angular momenta distribution from FIFRELIN were not available for this study, so we could not perform an exact subtraction of the rotational energy contribution using eqn. 1.12, as we did for GEF. We could, however, reconstruct how the J is estimated, following the description given by the developers of FIFRELIN in ref. [33]. Once the $E_{rot}$ is subtracted using our simplified treatment, the $\tilde{V}(A)$ from DE/FIN (“DE/FIN with FIFRELIN (w/o E_{rot})” in figs. 4.3-4.5) agrees almost perfectly with the one reported for FIFRELIN, except for nuclei close to $A = 132$, and its complementary mass, $A = (A^{CN} - 132)$.

4.4.4 FREYA

FREYA is also a Monte Carlo code, but unlike the others presented so far, uses a simplified description of the de-excitation of the FFs.

From the analysis in DE/FIN, the trend between $\tilde{V}_{HF}$ and $\tilde{V}_{LF}$ is consistently the opposite of what is observed from the standalone version of FREYA. The difference is evident if we calculate the average prompt $\tilde{V}$ for the light and heavy fragments, as presented in tab. 4.1. Such a substantial difference in the $\tilde{V}/\tilde{V}_{H}$ between DE/FIN and FREYA standalone is an indication that there might be some compensating effects in the code. This is also discussed in ref. [29], where the developers explain the introduction of an adjustable parameter $x$, necessary to redistribute the $E^*$ of the fragments and obtain an adequate description of $\tilde{V}(A)$.

The results shown in figs. 4.3 and 4.4 for FREYA use the distributions of $E^*$ and $\tilde{V}(A)$ from ref. [28] which reports on an earlier version of the code. As an outlook of this study, it would be interesting to repeat the same calculation with $E^*$ distributions obtained from the latest version, FREYA 2.0.2, released just a few months ago [45, 46].

4.4.5 PbP

The results of DE/FIN with PbP for $^{235}$U(n_{th},f) show a striking difference in the height of the sawtooth with respect to the standalone code and to the other models, especially for the light FFs and the total $\tilde{V}$. This is most likely due to the fact that the $E^*(A)$ at full acceleration includes also a fraction of rotation
energy that should not be available for neutron emission. Since this model, unlike FIFRELIN, does not provide a detailed description for the estimation of $E^{\text{rot}}$, we were not able to subtract this contribution [125]. It is interesting that this code does not explicitly calculate $E^{\text{rot}}$, as it is a very important part of the non-statistical $\gamma$ emission.

4.4.6 Total $\bar{\nu}$

The average prompt $\bar{\nu}$ for the light and heavy fragments, as well as the total prompt $\bar{\nu}$ for all the analysed cases are reported in tab. 4.1. In this case, the fragment mass distribution is important in the extraction of this value. To ensure a fair comparison, we used the same FY($A$) for all models for a specific reaction, i.e., the distributions from ref. [122] for $^{235}\text{U}(\text{n}_{\text{th}},f)$ and $^{239}\text{Pu}(\text{n}_{\text{th}},f)$, and the one from ref. [124] for $^{252}\text{Cf}(\text{sf})$.

The DE$\ell$FIN de-excitation tends to, only slightly, over-estimate the total $\bar{\nu}$ in the cases where $E^{\text{rot}}$ is correctly taken into account, while largely over-estimating it when $E^{\text{rot}}$ is available for de-excitation through neutron emission. Except for the differences already discussed for FREYA, the $\bar{\nu}_L/\bar{\nu}_H$ values from DE$\ell$FIN generally follow the trends of the standalone codes.

4.5 What have we learned?

The work of looking into and trying to find the "secrets" behind the many codes now available was a very instructive and rewarding experience. One general observation, that also helps to understand how closely related the evolution of the model codes is to experimental data, can be drawn comparing the $\bar{\nu}(A)$ for $^{252}\text{Cf}$ (fig. 4.5), with the ones from the other two reactions (figs. 4.3 and 4.4). When good quality data are available, the spread between the results from model calculations decreases significantly.

Also, within the same reaction, regions of low yield (i.e., where the uncertainties on the measurements increase) are the ones where the models show the larger discrepancy. Another way to look at it is that, in order to reproduce the total $\bar{\nu}$, models cannot deviate too much from each other and experimental data or systematics in regions of high yields. This is, of course, a strong advocacy for new, better measurements, but should also be seen as a warning that it is risky to rely on these models in regions where data are lacking.

When it comes to the lessons learned about the codes themselves, our observations can only be based on the few cases we have studied. Double-magicity at $A = 132$ seems to be a difficult test for all models. Most often it is here that the theoretical $\bar{\nu}(A)$ distributions show their minimum, a few mass numbers above the one that is expected from experimental data. Inconsistencies with data, and with the DE$\ell$FIN results, are also found at the mass of the complementary fragments.
Codes based on the Hauser-Feshbach formalism (CGMF and FIFRELIN) seem to behave more consistently in this study. Of course, the fact that TALYS also implements the same model does not exactly make this conclusion unexpected. Nevertheless, this is a verification that three implementations of the same model generally seem to produce good and consistent results. By not treating explicitly $E_{\text{rot}}$ (like the PbP model), or by using a simplified model for the emission of neutrons in competition with $\gamma$ de-excitation (like FREYA and GEF), codes are disregarding phenomena that can be important for a correct reproduction of experimental data.

By studying and comparing the $\bar{\nu}(A)$ distributions of the various models, we have also gained a better understanding of our methodology. We learned, e.g., how sensitive the results from $\text{DE}^{\ell}/\text{FIN}$ are to the inclusion of $E_{\text{rot}}$. At the same time, by properly using this information, our study could be extended to include gamma emission and production of isomeric ratios to compare with experimental data.

In conclusion, we now have a tool that, with only minor modifications, can be used to extract all sorts of information from FFs de-excitation.

A similar methodology, that uses TALYS to obtain IYRs is being used by V. Rakopoulos to extract the angular momentum of fission fragments, starting from measured isometric yields ratios from proton-induced fission of $^{238}\text{U}$ and $^{232}\text{Th}$ [115].
Table 4.1. Comparison of the evaluated average prompt $\bar{\nu}_{\nu_{\ell}}$ for the different reactions studied using DE$\ell$FIN with inputs from GEF, FREYA, PbP, FIFRELIN and CGMF. The DE$\ell$FIN calculation is compared to the standalone codes.

<table>
<thead>
<tr>
<th>Reaction</th>
<th>$\bar{\nu}<em>{\nu</em>{\ell}}$</th>
<th>ENDF B-VII.1</th>
<th>GEF</th>
<th>FREYA</th>
<th>PbP</th>
<th>FIFRELIN</th>
<th>DE$\ell$FIN (w/o Erot)</th>
<th>DE$\ell$FIN (FY CGMF)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{235}$U(n,f)</td>
<td>2.44 ± 0.08</td>
<td>2.48 ± 0.12</td>
<td>2.52 ± 0.14</td>
<td>2.54 ± 0.12</td>
<td>2.45 ± 0.14</td>
<td>2.39 ± 0.09</td>
<td>2.47 ± 0.09</td>
<td>––</td>
</tr>
<tr>
<td>$^{239}$Pu(n,f)</td>
<td>2.87 ± 0.13</td>
<td>2.84 ± 0.13</td>
<td>2.90 ± 0.13</td>
<td>2.91 ± 0.13</td>
<td>3.02 ± 0.14</td>
<td>2.83 ± 0.10</td>
<td>2.83 ± 0.10</td>
<td>––</td>
</tr>
<tr>
<td>$^{252}$Cf(SF)</td>
<td>3.76 ± 0.06</td>
<td>3.75 ± 0.11</td>
<td>3.91 ± 0.26</td>
<td>––</td>
<td>––</td>
<td>3.69 ± 0.15</td>
<td>3.94 ± 0.12</td>
<td>4.08 ± 0.24</td>
</tr>
</tbody>
</table>
5. Summary & Outlook

Never settle for something less.
Something’s better than nothing, yes!
But nothing’s better than more, more more

Breathless Mahoney, Dick Tracy

In this thesis I have presented my work, developed in the framework of AlFONS (Accurate FissiOn data for Nuclear Safety), a project supported by SSM (Swedish Radiation Safety Authority) and SKB (Swedish Nuclear Fuel and Waste Management Company). AlFONS aimed at measuring high-quality neutron-induced fission of various actinides at the IGISOL facility at JYFL (Department of Physics, University of Jyväskylä).

Accurate nuclear data for neutron-induced fission yields are desirable to better understand the fission process. Of particular interest are the isotopic yields of short-lived nuclei far from stability, for which data are very scarce, especially for fission induced by high-energy neutrons.

The IGISOL-4 facility at JYFL was proposed as the location for these studies of various actinides, where the IGISOL (Ion Guide and Isotope Separator OnLine) technique, coupled to the JYFLTRAP Penning trap, has been applied in the past to measure proton-induced fission yields.

The neutron source

In the first part of my project, I participated in the design, development and characterisation of a neutron source for IGISOL-4.

The source is based on a 5 mm thick, water-cooled beryllium converter, and will be used in conjunction with the MCC30/15 high-current cyclotron installed at JYFL, able to deliver 30 MeV protons at 100 μA.

We characterised the neutron source in two distinct campaigns at TSL (The Svedberg Laboratory) and at JYFL. Both measurements confirmed that MCNPX, one of the Monte Carlo codes used in the design phase, reproduces the neutron energy spectrum and the total yield sufficiently well in the energy range above approximately 1 MeV.

We also verified that, at the maximum current available from the MCC30/15, the source will produce between 2 and $5 \times 10^{12} \text{n/sr/s}$ on target.
This is enough to perform fission yields measurements at IGISOL-4 using the JYFLTRAP Penning trap.

Neutron-induced yields at IGISOL-4

After installing the neutron source, we measured, for the first time at IGISOL-4, product yields from high-energy neutron-induced fission of nat\(\text{U}\) in a dedicated campaign at JYFL.

We obtained data on the relative cumulative yields of tin and relative independent yields of antimony in the mass range \(A = 128-133\). Despite the limited amount of studied nuclei, and the wide energy spectrum of the neutron source, these data represent an important addition in this energy region, where available data are very scarce.

Our results for tin show even-odd effects that contradict what is reported in the evaluated nuclear data libraries for fast fission. Also for antimony, the measured data show a difference of as much as two mass units in the position of the mass peak, with respect to the evaluations.

Uncertainty in the decay data, especially for \(^{131}\text{Sn}\), call for a better determination of the yield with a technique that is not sensitive to these quantities. This can be achieved in a mass spectroscopy measurement with direct ion-counting using JYFLTRAP.

De-excitation of fission fragments with \texttt{DE\ell\textsc{FIN}}

In parallel with the experiments discussed above, we also started a work aimed at comparing and analysing phenomenological fission models, that culminated in the development of the \texttt{DE\ell\textsc{FIN}} (De-Excitation of Fission fragments) code. \texttt{DE\ell\textsc{FIN}} takes the excited fission fragments, the properties of which are defined by the models under test, and de-excites them in a reproducible and standardised way, using the physics models in the TALYS reaction code.

In this process, we achieved a better understanding of the ideas used in the development of fission models, and we found interesting features in the \(\bar{\nu}(A)\) distributions in some of the codes, that will be worth discussing further with the developers. Maybe even more importantly, we developed a tool that can easily be adapted in order to extract quantities that can be compared to our experimental results.

What’s next?

This is the first PhD thesis at Uppsala University on the scope of fission yield measurements at IGISOL-4, but it will not be the last. More work needs to be done, on many fronts, to reach the long-sought goal of a systematic measurement of yields with JYFLTRAP.
• **Neutrons at IGISOL-4:** when it comes to the neutron source, we have reached a rather good understanding of its yield and energy spectrum, thanks to the characterisation campaigns we performed at TSL and JYFL. There is however work to be done, for example, estimating the thermal neutron flux that, until now, has escaped our grasp. Also, a way to properly monitor the neutron flux online needs to be implemented. An idea currently being considered is to install TFBC detectors in proximity of the fission chamber at IGISOL. These detectors, once properly calibrated, will be an easy and reliable way to log the changes in the absolute neutron flux in real-time.

• **Efficiency studies:** one of the main limitations of the IGISOL experimental technique is that the stopping, extraction and transport efficiency of the fission products at various locations along the beam line has not been fully studied. It must be verified that the dependence of the total efficiency on the ions’ charge (Z) and mass (A) is negligible or, should this not be the case, proper correction factors must be estimated. This has currently been done with a Monte Carlo simulation, but needs to be verified with experimental data.
  Some of these data have already been acquired during the n-IGISOL campaign and now need to be analysed. The best way to finally address this problem, however, will likely be a dedicated measurement that uses a well known reaction (e.g., \(^{252}\text{Cf(sf)}\)) to obtain A- and Z-dependent efficiency factors.

• **New challenges for DEℓFIN:** the work on the fission models done with DEℓFIN is certainly worth continuing. In particular, now that we have a working framework, it would be interesting to study other quantities besides \(\bar{\nu}\). We have learned how sensitive the \(\bar{\nu}(A)\) distributions are to the rotational energy, it would now be interesting to study how \(\gamma\) emission is affected.
  Also, the same idea is now being applied to obtain a consistent, transparent and reproducible method to extract the angular momentum of the fission fragments based on experimental isomeric yield ratios.
Fissionsutbytet, det vill säga hur mycket av en viss kärna som skapas vid klyvning av en atomkärna, är en av flera viktiga observabler inom fissionsforskning. God kunskap inom detta område är nödvändig i alla steg av bränslecykeln för kärnkraftsreaktorer, exempelvis för reaktivitetsberäkningar, uppskattningar av (det använda) bränslets sammansättning och för beräkningar av resteffekt. Kunskap om fissionsutbyten leder också till bättre förståelse av själva fissionsprocessen eftersom de kan härledas tillbaka till den kluvna kärnans tillstånd vid klyvningsögonblicket. Data över fissionsutbyten är också viktiga inom exempelvis astrofysik, för att förstå nukleosyntes, och för skapandet av radioaktiva jonstrålar, vilka för närvarande är under utveckling vid flera forskningsanläggningar runtom i världen.

Fissionsutbytesdistributionen som funktion av massa, eller laddning, beror inte bara på vilken kärna som klyvs utan också på hur mycket energi som finns tillgänglig för reaktionen. För fission orsakad av neutroner är detta direkt kopplat till den inkommande neutronens rörelseenergi.

Detta avhandlingsarbete har genomförts inom ramen för AlFONS-projektet med stöd av Strålsäkerhetsmyndigheten, SSM, och Svensk Kärnbränslehantering AB, SKB. Projektet har till syfte att mäta neutroninducerade, oberoende fissionsutbyten för olika aktinider av relevans för ett geologiskt slutförvar respektive eventuell separation och transmutation av det använda kärnbränslet. Mätningarna planerades i samarbete med fysikinstitutionen vid universitetet i Jyväskylä (JYFL), Finland, och genomfördes vid deras IGISOL-4-anläggning vid universitetets acceleratorlaboratorium. IGISOL har tidigare använts vid mätningar av protoninducerade fissionsutbyten genom att räkna de joniserade fissionprodukterna med Penningfällan JYFLTRAP, som kan användas som en väldigt precis mass-spektrometer.

Avhandlingens första del beskriver utvecklingen och karaktäriseringen av en lämplig neutronkälla för installation vid IGISOL-4. Källan baseras på reaktionen $^9\text{Be}(p,\text{nx})$. Protoner med energin 30 MeV, från en av cyklotronerna vid JYFL, kolliderar med en 5 mm tjock berylliumplatta, vilket skapar ett flöde av snabba neutroner. Intensiteten och energifördelningen för neutronerna har studerats med hjälp av Monte Carlo-simuleringar och två mätningar, en vid The Svedberg-laboratoriet i Uppsala, den andra på plats vid IGISOL. Beräkningarna med en av simuleringskoderna (MCNPX) visade sig stämma väl överens med uppmätta data. Beräkningarna bekräftar också att vid den maximalt tillgängliga protonströmmen från MCC30/15-cyklotronen i Jyväskylä kommer ett neutronflöde på mellan 2 och $5 \times 10^{12}$ neutroner /cm²/s kunna uppnås, vilket var ett av huvudmålen vid designen av neutronkällan.
Neutronkällan har använts vid IGISOL-4 i en första mätning av utbytet från neutroninducerad fission av $^{nat}$U vid höga energier. Denna mätning presenteras och diskuteras i avhandlingen andra del. Fissionsproduktarna separerades med hjälp av en dipolmagnet, och isobarer i massområdet $A = 128 - 132$ valdes ut för insamling och identifiering med hjälp av gammaspektrometri. I avhandlingen rapporteras data för isotoputbyten för tenn och antimon. Även isomeriska utbyten, det vill säga hur stor andel av en viss kärna som skapas i ett metastabilt tillstånd, mättas för fem olika kärnor.

I det studerade massområdet påvisar de uppmätta resultaten signifikanta skillnader jämfört med evaluerade databaser. Dessa databaser representerar vår nuvarande kunskap om fission och resultaten visar på behovet av mer omfattande mätningar. Utbytet för tenn-isotoperna uppvisar ett betende omkring masstalet $A = 131$ som markant skiljer sig från data i den viktiga databasen ENDF/B-VII.1. Även det uppmätta utbytet för antimon-isotoperna avviker från evaluerade data, men överensstämmer med beräkningar gjorda med fissionsmodellkoden GEF. Stora osäkerheter i sönderfallsdata, framför allt för $^{131}$Sn, påkallar nya mätningar med en metod som inte är beroende av dessa, exempelvis masspektrometri med JYFLTRAP.

Den tredje delen av denna avhandling beskriver en studie av olika koder som använder kärnmodeller för att beräkna och beskriva fissionsprocessen. Syftet med dessa koder är att uppnå bättre kunskap om fragmentens tillstånd precis efter klyvningen, på en så kort tidsskala att den inte är direkt tillgänglig för experimentella mätningar. Utbytet för tenn-isotoperna uppvisar ett betende omkring masstalet $A = 131$ som markant skiljer sig från data i den viktiga databasen ENDF/B-VII.1. Även det uppmätta utbytet för antimon-isotoperna avviker från evaluerade data, men överensstämmer med beräkningar gjorda med fissionsmodellkoden GEF. Stora osäkerheter i sönderfallsdata, framför allt för $^{131}$Sn, påkallar nya mätningar med en metod som inte är beroende av dessa, exempelvis masspektrometri med JYFLTRAP.

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Metoden, som är implementerad i koden DE/FIN, går ut på att ta de exciterade fissionsfragmenten, som de definieras i respektive modell, och de-excitera dem på ett reproducerbart och standardiserat sätt med hjälp av kärnmodellkoden TALYS. Genom att på detta sätt eliminera alla tänkbara modellberoenden i hur de slutgiltiga observablerna beräknas, blir det möjligt att fokusera på de enskilda modellernas antaganden.

DE/FIN har hittills använts för att jämföra de prompta neutronernas fördelning från fission olika kärnor. Jämförelsen har gjorts för fem av de mest använda modellerna för beräkningar av fissionsobservabler. Förhoppningen är att denna studie kommer främja en ökad förståelse för den fysik som är inbyggd i de olika fissionsmodellerna. För vissa av koderna upptäcktes intressanta egenskaper i de beräknade fördelningarna som kommer leda till fortsatta diskussioner med modellutvecklarna. Ett viktigt resultat av arbetet var själva utvecklandet av DE/FIN, ett verktyg som enkelt kan användas för att extrahe
 olika storheter vilka kan jämföras med experimentella resultat, till exempel från fissionsutbytesmätningar vid IGISOL-4.
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A. Complete DE\ell FIN results

So long, so long and thanks for all the fish.

The dolphins,

*The Hitchhiker’s Guide to the Galaxy*
Figure A.1. $\bar{\nu}(A)$ distributions from the $^{235}\text{U}(n_{th},f)$ reaction, as obtained from \textsc{De/Fin} using input from GEF. \textsc{De/Fin} results are compared to the distributions from the standalone codes and to the Wahl systematics. The mass yields from ref. [122] are shaded behind the $\bar{\nu}(A)$ curves.

Figure A.2. $\bar{\nu}(A)$ distributions from the $^{235}\text{U}(n_{th},f)$ reaction, as obtained from \textsc{De/Fin} using input from PbP. \textsc{De/Fin} results are compared to the distributions from the standalone codes and to the Wahl systematics. The mass yields from ref. [122] are shaded behind the $\bar{\nu}(A)$ curves.
Figure A.3. $\bar{\nu}(A)$ distributions from the $^{235}\text{U}(n_{\text{th}},f)$ reaction, as obtained from DE/lFIN using input from FIFRELIN. DE/lFIN results are compared to the distributions from the standalone codes and to the Wahl systematics. The mass yields from ref. [122] are shaded behind the $\bar{\nu}(A)$ curves.

Figure A.4. $\bar{\nu}(A)$ distributions from the $^{235}\text{U}(n_{\text{th}},f)$ reaction, as obtained from DE/lFIN using input from FREYA. DE/lFIN results are compared to the distributions from the standalone codes and to the Wahl systematics. The mass yields from ref. [122] are shaded behind the $\bar{\nu}(A)$ curves.
Figure A.5. \( \bar{\nu}(A) \) distributions from the \(^{239}\text{Pu}(n_{th},f)\) reaction as obtained from DE\textsc{fIN} using input from GEF, compared to the results of the standalone codes and to Wahl systematics. The mass yields from ref. [122] are shaded behind the \( \bar{\nu}(A) \) curves.

Figure A.6. \( \bar{\nu}(A) \) distributions from the \(^{239}\text{Pu}(n_{th},f)\) reaction as obtained from DE\textsc{fIN} using input from FIFRELIN, compared to the results of the standalone codes and to Wahl systematics. The mass yields from ref. [122] are shaded behind the \( \bar{\nu}(A) \) curves.
Figure A.7. $\tilde{\nu}(A)$ distributions from the $^{239}$Pu(n$_{th}$,f) reaction as obtained from DE/FIN using input from FREYA, compared to the results of the standalone codes and to Wahl systematics. The mass yields from ref. [122] are shaded behind the $\tilde{\nu}(A)$ curves.
Figure A.8. $\bar{\nu}(A)$ distributions from $^{252}\text{Cf}(SF)$ as obtained from DELFIN using input from GEF compared to the results from the standalone codes and experimental data (ref. [124]). The mass yields from ref. [124] are shaded behind the $\bar{\nu}(A)$ curves.

Figure A.9. $\bar{\nu}(A)$ distributions from $^{252}\text{Cf}(SF)$ as obtained from DELFIN using input from CGMF compared to the results from the standalone codes and experimental data (ref. [124]). The mass yields from ref. [124] are shaded behind the $\bar{\nu}(A)$ curves.
Figure A.10. $\bar{\nu}(A)$ distributions from $^{252}$Cf(SF) as obtained from DELFIN using input from FIFRELIN compared to the results from the standalone codes and experimental data (ref. [124]). The mass yields from ref. [124] are shaded behind the $\bar{\nu}(A)$ curves.
B. Changes in the GEF source code

Here follows a comparison of the changes between the original GEF code [118] (that I will call GEF-original.bas, and mark in red), and the one I used to create the E* files I input to DE\ell FIN (GEF-4-DE\ell FIN.bas, in green).

In both versions the Total Excitation Energy $TXE$ of a fission fragment is defined as:

\[
\begin{align*}
5377: & \quad TXE_{\text{light}} = E_{\text{exc,light}} + E_{\text{rot,light}} \quad \text{\' final values} \\
5378: & \quad TXE_{\text{heavy}} = E_{\text{exc,heavy}} + E_{\text{rot,heavy}} \quad \text{\' " "}
\end{align*}
\]

\textit{i.e.}, as the sum of the excitation energy $E_{\text{exc,light/heavy}}$ and $E_{\text{rot}}$.

In GEF-original.bas, the lines I then modified read:

\[
\begin{align*}
2833: & \quad \text{If PXE = 1 Then Print } \#\text{foutlmd,} \\
& \quad \text{"* Eexc1: Excitation energy of first fragment [MeV]"} \\
2834: & \quad \text{If PXE = 1 Then Print } \#\text{foutlmd,} \\
& \quad \text{"* Eexc2: Excitation energy of second fragment [MeV]"} \\
\end{align*}
\]

\[
\begin{align*}
2833: & \quad \text{If PXE = 1 Then Print } \#\text{foutlmd,} \\
& \quad \text{"* Eexc1: Excitation (TXE1 - Erot1) energy of first fragment [MeV]"} \\
2834: & \quad \text{If PXE = 1 Then Print } \#\text{foutlmd,} \\
& \quad \text{"* Eexc2: Excitation (TXE2 - Erot2) energy of second fragment [MeV]"}
\end{align*}
\]
The information in output is no longer $TXE_{\text{light/heavy}} = E_{\text{exc_light/heavy}} + E_{\text{rot_light/heavy}}$, but the excitation energy $E_{\text{exc_light/heavy}}$. 
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List of Acronyms

**ACCLAB** Accelerator Laboratory, University of Jyväskylä.
**AlFONS** Accurate FissiOn data for Nuclear Safety.
**AUP** Actual User Position.

**BRIGIT** BReakdown Counters at IGIsol Target.

**CFY** Cumulative Fission Yield.
**CGMF** CGM+FFD.
**CN** Compound Nucleus.

**DAQ** Data AcQuisition.
**DE/FIN** De-Excitation of Fission fragmeNts.

**ENDF** Evaluated Nuclear Data Files.
**ER-BSS** Extended-Range Bonner Sphere Spectrometer.

**FALSTAFF** Four Arm cLover for the Study of Actinide Fission Fragments.
**FF** Fission Fragment.
**FIFRELIN** Fission FRagment Evaporation Leading to an Investigation of Nu-clear data.
**FP** Fission Product.
**FREYA** Fission Reaction Event Yield Algorithm.
**FY** Fission Yield.

**GEF** GEneral Fission.
**GS** Ground State.

**IFY** Independent Fission Yield.
**IGISOL** Ion Guide and Isotope Separator OnLine.
**IYR** Isomeric Yield Ratio.

**JEFF** Joint Evaluated Fission and Fusion File.
**JENDL** Japanese Evaluated Nuclear Data Library.
**JYFL** University of Jyväskylä, Department of Physics (Jyväskylän Yliopisto Fysiikan Laitos).

**LANSCE** Los Alamos Neutron Science Center.
**LDM** Liquid Drop Model.

**MM-RNR** Multi-Modal Random Neck Rupture.
PAULA Protons At UppsaLA.
PbP Point-by-Point.

SKB Swedish Nuclear Fuel and Waste Management Company (Svensk Kärnbränslehantering AB).
SPIDER Spectrometer for Ion DEtermination in fission Research.
SPIG SextuPole Ion Guide.
SSM Swedish Radiation Safety Authority (Strålsäkerhetsmyndigheten).
STEFF SpectromeTer for Exotic Fission Fragments.

TFBC Thin Film Breakdown Counter.
TOF Time of Flight.
TSL The Svedberg Laboratory.

VAMOS VAriable MOde Spectrometer.
VERDI VElocity foR Direct particle Identification.
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