

Universidad Complutense de Madrid
Departamento de Física Atómica, Molecular y Nuclear



DOCTORAL THESIS

**MEASUREMENTS OF THE ^{237}Np AND ^{240}Pu
NEUTRON CAPTURE CROSS SECTIONS
AT THE CERN N_TOF FACILITY**

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Ciemat
Centro de Investigaciones
Energéticas, Medioambientales
y Tecnológicas

A mi familia,
a Calia.

Madrid, 22 de octubre de 2008

Por la presente, el Dr. Daniel Cano Ott, Investigador Titular del Centro de Investigaciones Energéticas, Medioambientales y Tecnológicas CIEMAT certifica:

que la memoria titulada Measurements of the ^{237}Np and ^{240}Pu neutron capture cross sections at the CERN n_TOF facility / Medidas de las secciones eficaces de captura neutrónica del ^{237}Np y del ^{240}Pu en la instalación n_TOF del CERN, ha sido realizada por Carlos Guerrero Sánchez bajo mi dirección en la Unidad de Innovación Nuclear del CIEMAT, y constituye la Tesis que presenta para optar al grado de Doctor en Ciencias Físicas.

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Investigador Titular
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Lord of the Ring

Hay un anillo, bajo tierras suizas
y francesas, que es un secreto a voces
donde chocan partículas veloces
y se quedan, las pobres, hechas trizas.

Quieren saber, los físicos palizas
y un doctorando, de los más precoces,
tratando los actínidos a coces
qué le pasa al plutonio si le atizas.

Hadrones y leptones acelera
sin dejar núcleo radiativo entero.
Tomándole al neutrón la delantera,

se calza su calzado montañoero
y se sube al Mont Blanc de una carrera...
su nombre, sin dudar, Carlos Guerrero.

Geneva, 12-9-2004

Paco Vida

Abstract

In this work, the neutron capture cross sections of ^{237}Np and ^{240}Pu have been measured at the n_TOF/CERN facility in the energy range from 1 eV to 2 keV. Since the total absorption technique has been employed for at n_TOF for the first time in this work, a variety of analysis techniques and software tools for the data analysis had to be developed. These have been validated with experimental data and have been shown to provide a capture yield with an accuracy between 4% and 6%, depending on the particularities of the measurement.

The experimental data have been analyzed with the SAMMY code in combination with the most reliable transmission data. The results have been, for each isotope, a set of resonance parameters and the associated covariance matrix as well as the corresponding point-wise and average capture cross sections. The cross sections obtained in this work have been compared with all the previous data available as well as with the most recent evaluations. It has been shown that, for both isotopes, the data presented in this work are the most accurate and have the highest resolution to date.

An additional result consisted in the identification of a set of specific actions that will be implemented in future measurements at n_TOF to become even more accurate cross sections and to cover a wider energy range.

Resumen

En este trabajo se han medido, entre 1 eV y 2 keV, las secciones eficaces de captura neutrónica del ^{237}Np y del ^{240}Pu en la instalación n_TOF del CERN. Dado que este trabajo es el primero en que se utiliza la técnica de absorción total en n_TOF, se han desarrollado toda una serie de técnicas de reducción de datos y herramientas informáticas para el análisis de datos. Éstas han sido validadas con datos experimentales y han demostrado proporcionar un tasa de captura con una precisión de entre el 4% y el 6%, dependiendo de las características de la medida.

Los datos experimentales se han analizado con el código SAMMY en combinación con la datos de transmisión más fiables. Como resultado, se han obtenido para cada isótopo un conjunto de parámetros de las resonancias y las matrices de covarianza asociadas, así como las sección eficaces de captura neutrónica promedio y punto a punto. Las secciones eficaces obtenidas en este trabajo se han comparado con los valores experimentales previos disponibles, así como con las evaluaciones más recientes. Se ha demostrado que, para ambos isótopos, los datos presentados en este trabajo son los de mayor precisión y resolución hasta la fecha.

Un resultado adicional ha consistido en la identificación de un conjunto de acciones concretas que deberían llevarse a cabo para que la próxima medidas en n_TOF sea aún más precisas y cubran un rango de energía más amplio.

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Part I

INTRODUCTION

Chapter 1

Motivation and Objectives

This manuscript summarizes the work done on the measurement and analysis of the neutron capture cross sections of ^{237}Np and ^{240}Pu . Both isotopes are relevant to the management of the radioactive waste from conventional nuclear power plants and do not occur in nature.

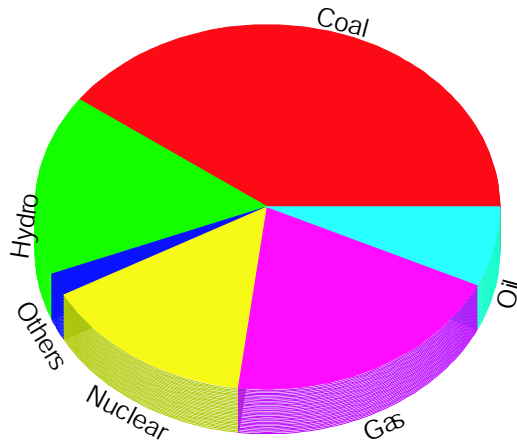
The aim of this chapter is to give an overview of the world situation concerning the consumption and demand of energy in general and nuclear energy in particular. Some of the main advantages and well as problems of nuclear energy are discussed, and the technology of Partitioning and Transmutation is presented as part of the solution for the long term management of the nuclear waste. Finally, the need of accurate measurements of basic nuclear data, such as the neutron cross sections of minor actinides, is discussed.

1.1 World Energy Outlook

From the industrial revolution in the 19th century to the rise of the new economies in the beginning of the 21st century, the economic growth of all countries has been correlated with the increase in energy demand [3]. In these two centuries, a wide variety of energy sources have been exploited, from fossil fuels such as coal, petroleum and gas to nuclear, hydro and other renewable sources such as solar and wind.

Nowadays, the contribution of each energy source to the total electricity consumption varies significantly from country to country, even within the European Union (EU). Figure 1.1 shows the contribution of the different sources of energy to the world and EU electricity consumption [4]. It is observed that coal (40.3%) and gas (19.7%) cover more than half of the world electricity production, while in the EU the main contributions correspond to nuclear energy (31%) and coal (29%). Concerning renewable technologies, the maximum production is generated as hydroelectricity while the rest (solar, wind, waste, etc.) contribute less than 5% to the total.

WORLD 2005



European Union 2005

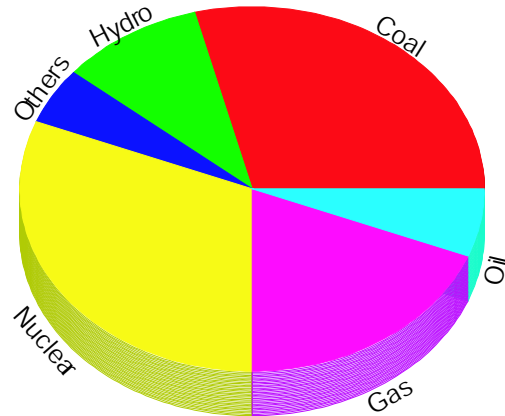


Figure 1.1: Contribution of the different sources of energy to the world (left) and EU (right) electricity consumption. Others: solar, wind, waste,...

In the forthcoming decades, the International Energy Agency (IEA - World Energy Outlook 2007 [4]) expects an annual increase of world demand on electricity to be 2.6%, led by the new non-OECD economies in such a way that their demand on electricity will overcome that of the OECD countries by 2015. This, together with the expected rise of oil prices, the geopolitical risks and growing competition associated to the production of oil and gas, and global warming, has triggered a worldwide debate about the future of energy production and demand.

In view of this scenario the EU has proposed in its report *A European strategic energy technology plan: Towards a low carbon future* [5], to reduce by 20% both the primary energy consumption and the emissions of CO₂, and reach the 20% production from renewable energies by the year 2020. These goals follow the lines discussed in the EU document *On Conventional energy sources and energy technology* [6] in which is stated clearly that nuclear energy is indispensable if basic energy needs are to be met in Europe in the medium term. This conclusion brings the need to increase efforts in the design of more sustainable nuclear technologies for both energy production and waste management.

1.2 Overview on Nuclear Energy

The development of nuclear energy took only a few years after the discovery of the neutron in 1932 by James Chadwick [7]. Six years later, Otto Hahn and Fritz Strassmann [8] observed in their study of the neutron interactions with matter that Uranium atoms would break-up into two fragments under neutron bombardment, emitting in the process two or three neutrons and

a significant amount of energy. The process was first explained by Meitner and Frisch in their work *Desintegration of Uranium by Neutrons: a New Type of Nuclear Reaction* [9], published by Nature in 1939, in which they gave it its actual name *nuclear fission*.

The development of the first nuclear reactor (Chicago Pile Number One) was lead by Enrico Fermi and Leo Szilard. Fermi had already worked on neutron induced reactions, for which he was awarded the Nobel Prize in 1938, and Szilard conceived and patented the nuclear chain reaction [10] (see Figure 1.2). Both patented together the neutronic reactor in 1944 [11], only twelve years after the discovery of the neutron.

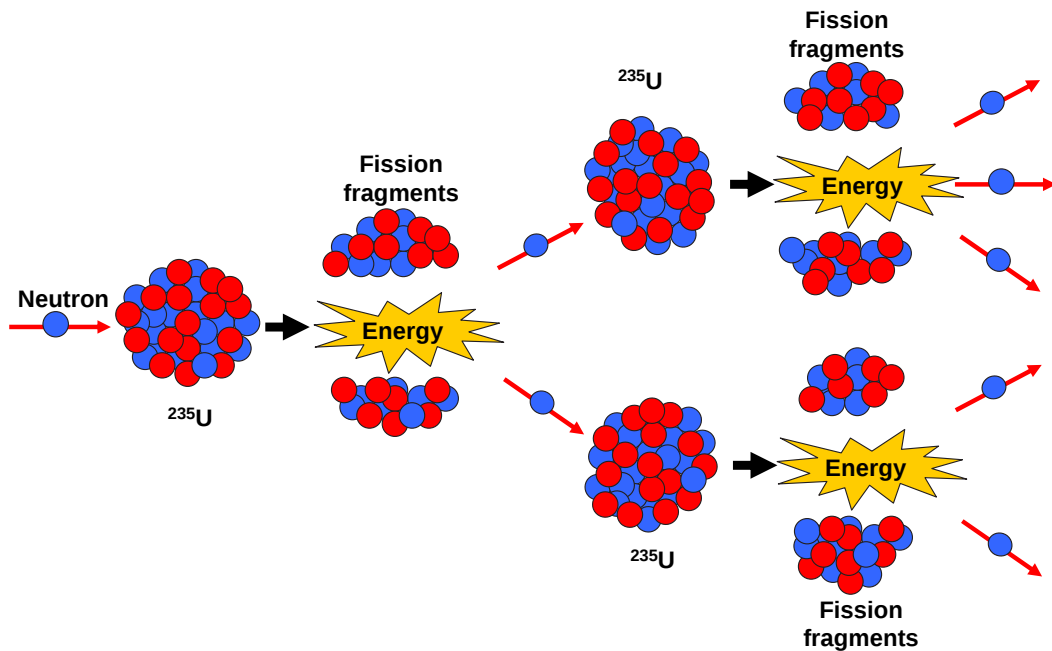


Figure 1.2: Simple scheme of the fission chain reaction: the fission reactions are induced by neutrons emitted in previous fission reactions.

1.2.1 Nuclear Energy in the world

In 1951, the Experimental Breeder Reactor EBR-I located in Idaho (USA) became the world's first electricity-generating nuclear power plant when it produced sufficient electricity to illuminate four 200-watt light bulbs. A few years later, in 1958, Calder Hall was inaugurated in Great Britain as the first commercial nuclear power plant. Since then several countries developed their own nuclear energy programs. Encouraged by the oil crisis in the early seventies, some industrialized countries such as Germany, Canada, Spain, France, Italy and Japan started their nuclear programs and were later followed by Mexico, Brazil, Taiwan and Korea. The construction of new nuclear power plants was reduced in the continuing decades, mainly due to the safety

concerns of the general public after the Chernobyl accident in 1986.

As of April 2008, there are a total of 439 Nuclear Power Reactors operating in 31 countries with a total installed power of 372 GWe. In the EU, a total of 197 nuclear power plants with an installed power of 170 GWe are in operation and 13 units with 12 GWe are under construction in five different countries. The number of nuclear power plants and the net nuclear power capacity of each country within the EU are listed in Table 1.1.

Country	In operation		In construction	
	#	MWe	#	MWe
Belgium	7	5,824	-	-
Bulgaria	2	1,906	2	1,906
Czech Rep.	6	3,523	-	-
Finland	4	2,696	1	1,600
France	59	63,260	1	1,600
Germany	17	20,470	-	-
Hungary	4	1,829	-	-
Lithuania	1	1,185	-	-
Netherlands	1	482	-	-
Romania	1	1,310	-	-
Russian Fed.	31	21,743	7	4,789
Slovakian Rep.	5	2,034	-	-
Slovenia	1	666	-	-
Spain	8	7,450	-	-
Sweden	10	8,974	-	-
Switzerland	5	3,220	-	-
Ukraine	15	13,107	2	1,900
United Kingdom	19	10,222	-	-
European Union	197	169,901	13	11,795

Table 1.1: *Nuclear power plants in the European Union and neighbouring countries, in operation and under construction, as of April 2008 [2].*

Concerning the share of nuclear energy in the overall energy production in each country, France holds the top position with a share of 79%, followed by Lithuania with 70%, Belgium and Slovakian Republic with 56% and Sweden with 47%. In Spain [1] a total of 6 nuclear power plants with 8 reactors are responsible for 20% of the total electricity demand, half of which is produced by strong CO₂ emitters, mainly coal and gas.

1.2.2 Nuclear Power Plants

The mechanism of energy production in most nuclear power plants is similar to that of conventional thermal plants (gas, coal and oil), in the sense that water is heated up in order to produce steam, directly or indirectly, and drive a turbine that generates electricity. The main

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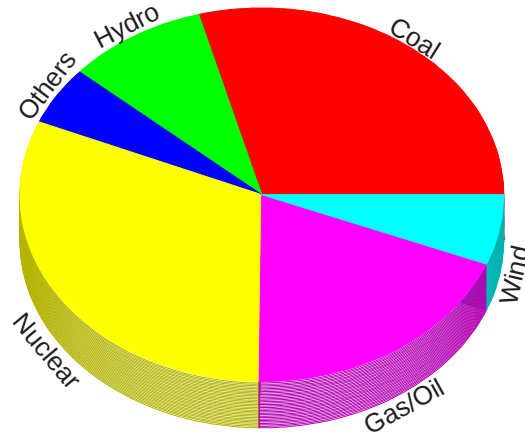


Figure 1.3: *Spanish distribution of electricity production by fuel.*

difference, which is indeed a very significant one, is that the heat is produced by a self sustained fission chain reaction.

The typical functioning of a Pressurized Water Reactor (PWR) can be followed in Figure 1.4: the core (12) of the nuclear reactor is built with fuel containing enriched uranium (3-5% content in ^{235}U) in which fission reactions take place releasing a large amount of energy and a few neutrons (2.3 neutrons/fission on average). The neutrons trigger additional fission reactions and sustain a chain reaction that is self-regulated by basic physical principles inserting and removing a series of *control rods* (1) made of strong neutron absorber materials. The energy released is used to heat the coolant (13) passing through the core. In an indirect cycle plant, as it is a PWR, the heat is then transferred to the water of a secondary circuit (11) through heat exchangers (3) in order to produce steam which is finally fed into a turbine (4), where the steams expands to very low pressure and ambient temperature, producing mechanical work (5) which is transformed into electricity (6). The water of the secondary circuit is cooled down in a heat exchanger with a cold water source (8,15) such as a river, a lake or the sea. A cooling tower is present, which sprays and cools the water down to ambient temperatures in order to be reused, except for the losses caused by evaporation (7) that are released to the atmosphere. In the case of BWR power plants, which use a direct cycle, the steam is directly produced in the core and then fed into the turbines.

Most of the existing nuclear power reactors (88%) use enriched ^{235}U fuel and light water as a moderator so that neutrons are slowed down (moderated) to thermal velocities (2200 m/s) at which the probability of inducing fission reactions in ^{235}U is enhanced. These are known as Light Water Reactors (LWR) and may work with pressurized (PWR) or boiling (BWR) water. The second most abundant type of nuclear reactor (10%) is the Canadian Pressurized Heavy Water Reactor (PHWR), which works with deuterated water and natural or slightly enriched

uranium fuel. Other reactor types are the Light Water Graphite Reactor (LWGR) designed by the Soviet Union, and the Advanced Gas Cooled Reactor (AGCR) designed by United Kingdom.

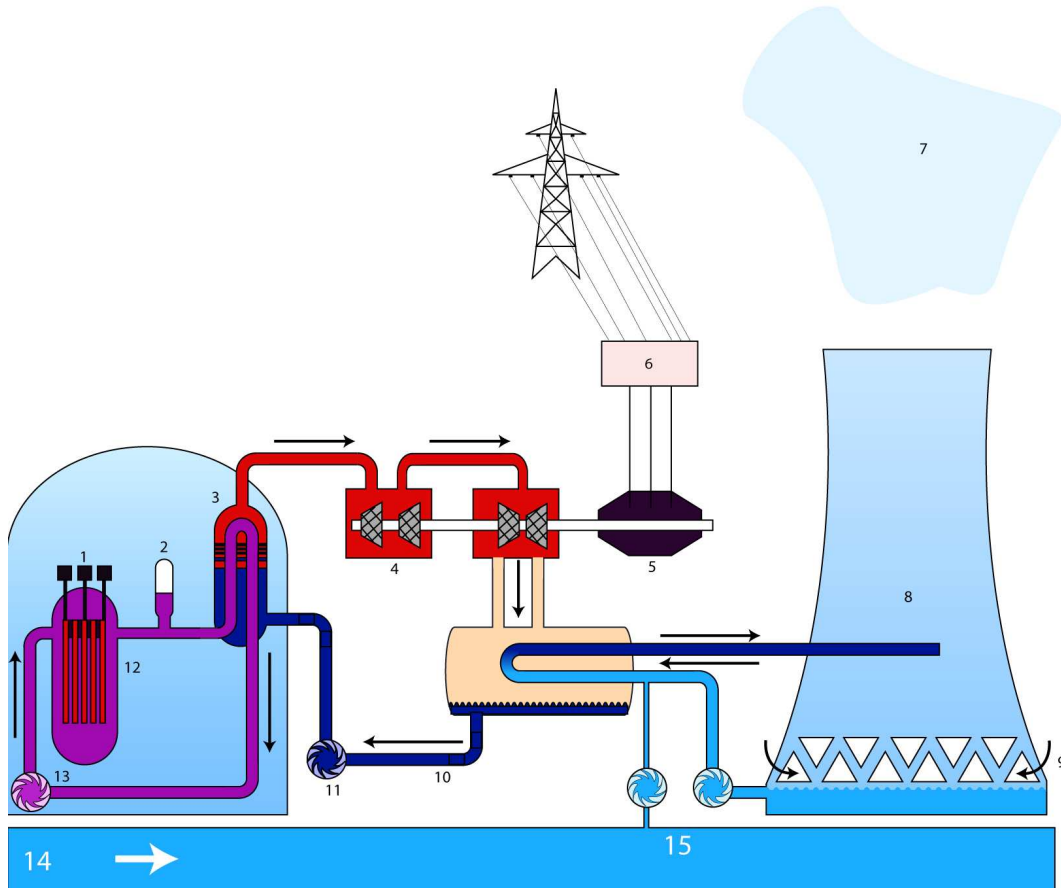


Figure 1.4: Nuclear power plant with Pressurized Water Reactor. (1) Control rods. (2) Pressurizer. (3) Heat exchanger. (4) Turbine. (5) Electric generator. (6) Transformer to high voltage. (7) Water vapour. (8) Refrigeration tower. (9) Air entrance. (10) Feed water. (11) Secondary pump. (12) Reactor vessel. (13) Primary pump. (14) Natural cold source. (15) Water supply.

1.2.3 Radioactive nuclear waste

During the normal operation of a nuclear power plant, radioactive nuclear waste is generated either by (i) fission reactions producing unstable fission fragments, (ii) by neutron induced reactions such as (n,γ) or (n,α) on uranium and higher atomic mass elements that give rise to transuranic elements, or (iii) by neutron activation of the water and structural materials. Among all, the activation products are mainly γ -ray emitters, while the fission fragments usually decay by β emission and the transuranic elements by both α and β emission with relatively long

half-lives.

The radioactive nuclear waste can be classified in multiple ways according to its physical or chemical form, specific activity, half-life, emitted radiation or radiotoxicity. Based on the thermal hazard and the requirements for disposal, the IAEA [12] distinguishes between two types of waste: high level (HLW), and low- and intermediate-level waste (LILW) with short and long lifetimes. The main characteristics that are the basis of their classification are summarized together with one possible strategy proposed for their management in Table 1.2.3

Category	HLW	LILW-LL	LILW-SL
Main character.	Highly radioactive waste, containing mainly fission products, as well as some actinides, separated during reprocessing of irradiated fuel.	Waste which, because of its radionuclide content requires shielding but needs little or no provision for heat dissipation during handling and transport.	Waste, which, because of its low radionuclide content, does not require shielding during normal handling and transport.
Heat generation	Any other waste with radioactivity levels intense enough to generate heat more than 2kW/m^3 by the radioactivity decay process.	$<2\text{kW/m}^3$	$<2\text{kW/m}^3$
Half-life		>30 years	<30 years
Other character.			Activity <400 Bq/g of long lived α emitters.
Management	Deep geological disposal	Geological disposal	Surface disposal

Table 1.2: *IAEA classification of the radioactive nuclear waste.*

Concerning the HLW, a solution reached by consensus of specialists consists in its geological disposal at several hundred meters underground, making use of the surrounding medium as a barrier to prevent the leakage of radioactive isotopes into the environment. However, this solution must be integrated among the energy policies of EU and many other countries that aim at achieving more sustainable energy production systems, mainly by the optimization of natural resources and the minimization of the long term legacy of hazards for future generations.

The principles of sustainability applied to the field of nuclear energy imply, mainly, to the minimization of the radiotoxicity (volume and dose) of the HLW to be buried in the geological disposal. This objective can be achieved by means of the *Partitioning and Transmutation* (P&T) technologies, which reduce significantly the risk and simplify the conditions of the final storage. This is the field of study of the RED-IMPACT project [13] of the EU, which is currently assessing the impact P&T is likely to have on the final geological repository requirements and the sustainability of nuclear energy.

An illustrative example of the generation of transuranic elements in nuclear reactors is given in Figure 1.5. It shows the isotopic composition of the fresh fuel of a PWR reactor (black) and that of the spent fuel after irradiation and cooling during 4 years (light brown). It is clearly observed how transuranic elements like Np, Pu, Am and Cm that were not present in the fresh fuel are produced after the irradiation. The third bar for each isotope (dark brown) corresponds to the composition of the nuclear fuel of a transmutation device such as an Accelerator Driven System (ADS), which is still at the design stage.

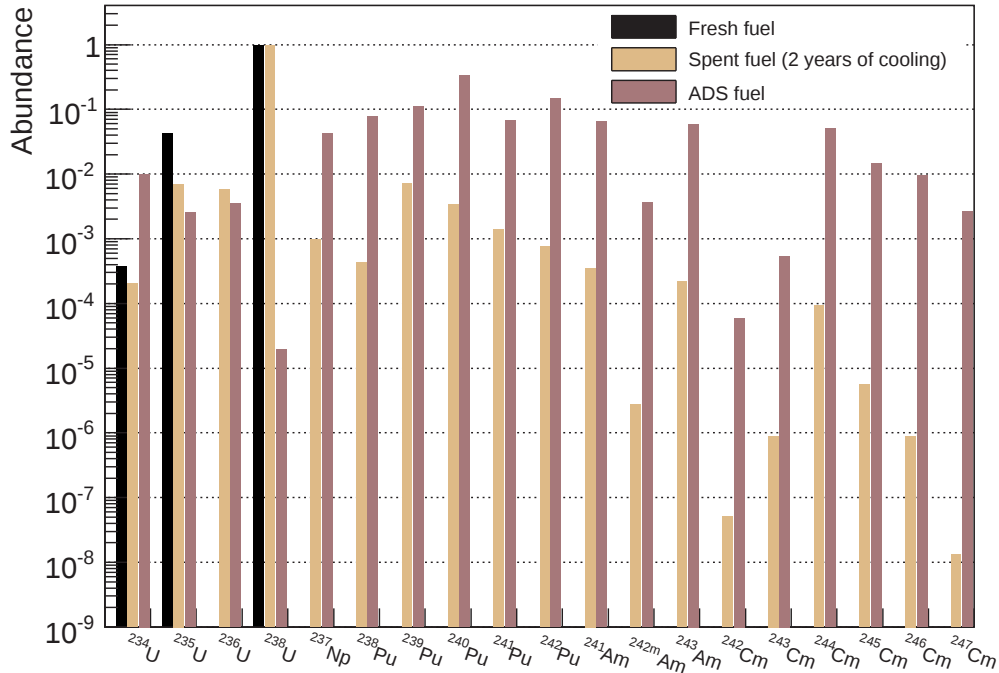
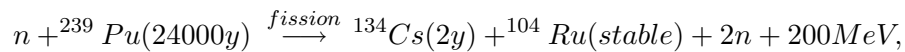


Figure 1.5: Isotopic composition of the nuclear fuel in different scenarios. Black: fresh fuel of a PWR. Light brown: spent fuel of a PWR after 2 years of cooling. Dark brown: fuel proposed for an ADS.

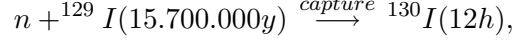
1.3 Transmutation of nuclear waste

The idea of P&T is to chemically process the HLW, isolating the different elements (partitioning) and to design a nuclear device capable of burning a minor actinide enriched fuel (transmutation) in such a way that the volume and radiotoxicity of the HLW is reduced. In recent years, several research projects have been approved by the different countries and international agencies related to the design and experimental tests of the future generation of reactors and transmutation devices. A complete review of the programs supported by the EU can be found in Ref. [14].

The key process in the transmutation is, as in conventional reactors, the neutron induced fission. Fission results in a distribution of fission fragments with a radioactivity that is high on average but remains for a shorter period than the transuranic elements. The transmutation would take place in a nuclear device with a fast neutron spectrum because the ratio of fission to absorption increases significantly for most actinide isotopes at high neutron energies. An example of transmutation by fission reactions is



where the half-life of each isotope is given in parenthesis. Transmutation can also be achieved by other neutron induced reactions such as neutron capture



or a combination of several reactions

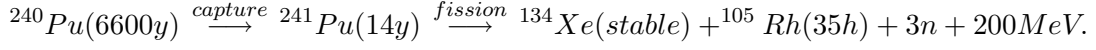


Figure 1.6 illustrates the evolution in time of the contribution of each transuranic isotope to the total radiotoxicity of the nuclear waste given in mSv per initial ton of Uranium. This information gives an idea of the importance of the transmutation of the different isotopes for the overall reduction of the radiotoxicity: in the first years ${}^{239}\text{Pu}$, ${}^{241}\text{Pu}$ and ${}^{244}\text{Cm}$ are the major contributors to the radiotoxicity, while in the time span between 50 and 2000 years after unloading, the radiotoxicity is mainly determined by ${}^{241}\text{Am}$. At longer times ${}^{240}\text{Pu}$, ${}^{239}\text{Pu}$ and ${}^{243}\text{Am}$ dominate up to 10^5 years, when the radiotoxicity of the spent fuel reaches the value corresponding to that of the fresh fuel. Despite its small contribution at short times, neptunium is of concern for the long-term disposal of the spent nuclear fuel due to its long half life (2.1×10^6 yr), build-up from the decay of ${}^{241}\text{Am}$ and its potential mobility in groundwater. Neptunium is therefore considered of great importance [15] in the overall risk assessment of the long-term disposal of the spent nuclear fuel.

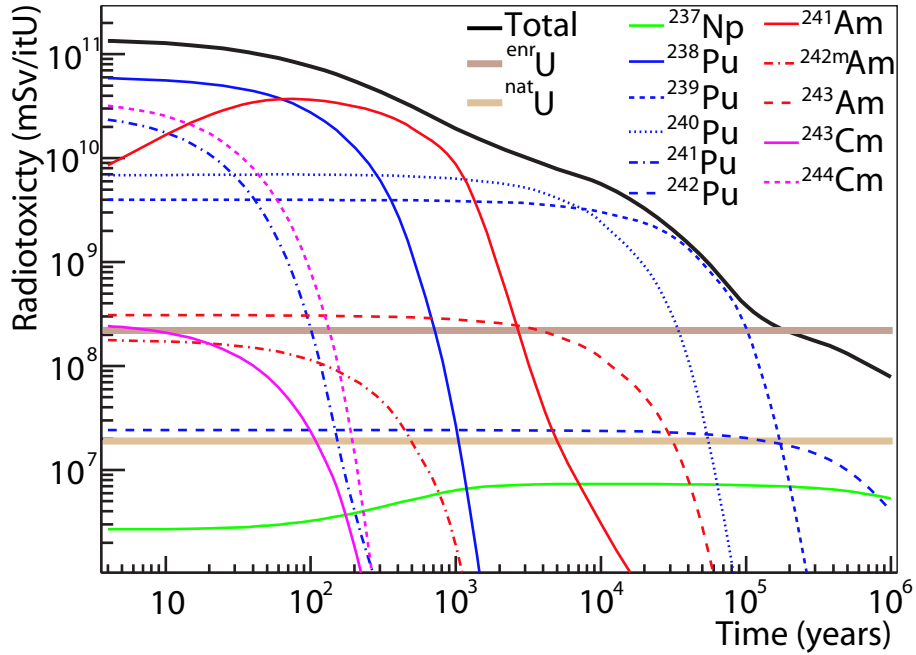


Figure 1.6: Contribution of each isotope to the radiotoxicity of the nuclear waste.

Among the different scenarios of implementation of the P&T strategy, there are two options that have received special attention in recent years:

- The first option consists in the use of fast critical reactors of the Generation IV type (Gen-IV). The primary goals of the Gen-IV reactors are to improve the nuclear safety, improve the proliferation resistance, minimize the waste and the utilization of natural resources, and to reduce the cost to build and run such plants. In addition, some of the Gen-IV type of reactors will be loaded with a small content of Minor Actinides from the nuclear waste produced in thermal reactors in order to eliminate part of the existing HLW.
- The second option consists in the use of a subcritical nuclear device with a fast neutron spectrum. In a subcritical reactor the fission chain-reaction is sustained by an external source of neutrons consisting in a proton accelerator coupled to a spallation target. In this way, whenever the neutron source is turned off, the reaction ceases. This device is known as an Accelerator-Driven System (ADS). The subcritical operation allows increasing the flexibility of the system, and hence to load fuels with a high concentration of MA compared to those of fast critical reactors.

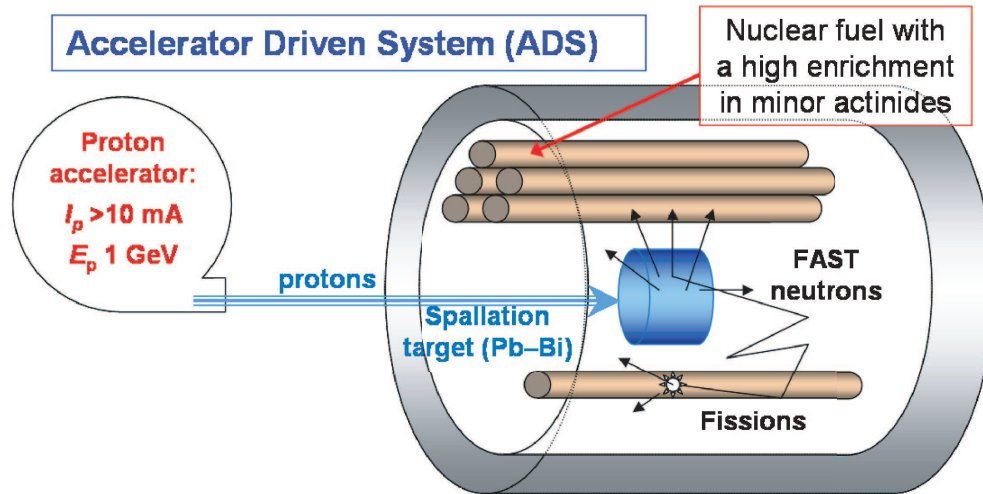


Figure 1.7: Schematic view of the different components of an ADS.

The conceptual design of the ADS [16] has been widely discussed in the recent years, see Figure 1.7 for a simplified scheme, no prototypes are yet available. At this stage the construction of an experimental ADS is possible and considered [17], but there are several issues that must be further investigated before the industrial scale ADS is reached [18, 19]:

- The measurement of accurate nuclear data for neutronic calculations of kinetic and dynamic core behaviour for better modelling of core physics, safety, radiation shielding and so on in order to reduce the design uncertainties.
- Research on the materials for fuel and structural materials (cladding for the fuels with high content of actinides, vacuum window between the accelerator and the spallation target, etc.) in various coolants and in radiation fields, including protons.
- The partitioning technology making use of the aqueous- as well as pyro-reprocessing.

- The performance assessment for a geological disposal in order to clarify the overall cost-benefit analysis of a P&T scenario.

It is interesting to underline that many of such developments are common for subcritical and critical systems.

1.4 Nuclear data needs

The design of a new nuclear device, including the future ADS and Gen-IV reactors, rely on the validity of the results given by dedicated codes of neutron transport, heat transfer, material damage, etc. Concerning neutron transport, the main ingredient of the calculation codes are basic nuclear data such as neutron cross sections, decay rates, emission probabilities of secondary particles (multiplicities, energies, angular distributions), etc. Hence, the impact of neutron cross section uncertainties is very significant on the inventory of the final repository as well as on the safety assessment and the economic evaluation of a dedicated core.

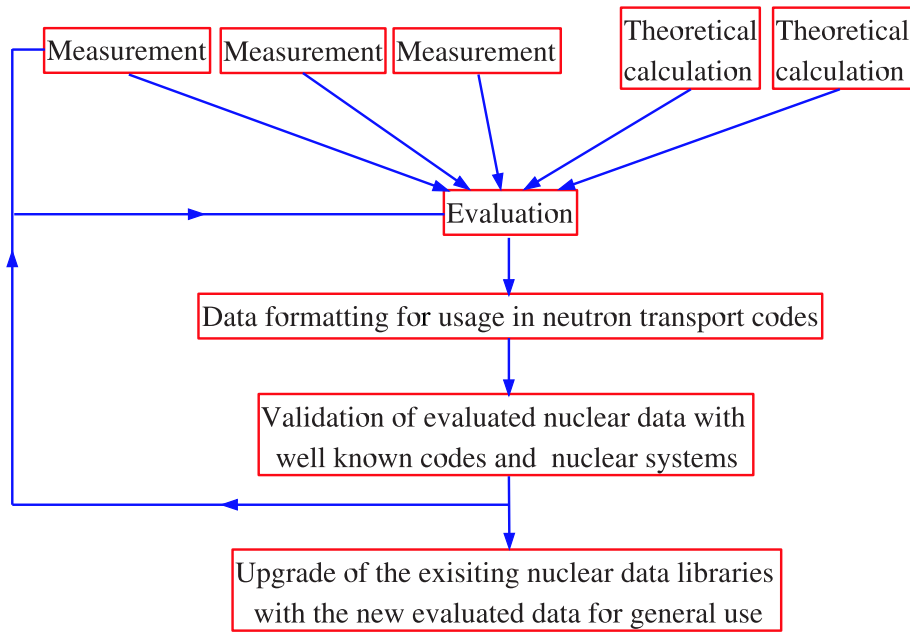


Figure 1.8: *Conceptual scheme of the different steps between the measurement of nuclear data and the upgrade of the general purpose Evaluated Nuclear Data Files.*

The measured neutron cross sections follow a complex process of evaluation before becoming available for the nuclear data users community, as sketched in Figure 1.8. For each specific isotope, the evaluators combine the experimental data available with theoretical models and produce a coherent set of evaluated cross sections suitable for reactor calculations. After a complete validation process with well known neutron transport codes and benchmarked integral

experiments, the evaluated cross sections are made available to the public for applications in the form of *Evaluated Nuclear Data Files* (ENDF) [20], which are published by the different nuclear energy agencies. The most used libraries are JEFF (EU), ENDF (USA), JENDL (Japan), BROND (Russia) or CENDL (China), which can be found in [21].

At present, the accuracy of the evaluated neutron cross sections is very different from one isotope to another. Such differences are related to the fact that the actual nuclear power plants are designed to work with a thermal neutron flux and enriched uranium fuel. For instance, the accuracy of the fission and capture neutron cross sections ^{235}U and ^{238}U at thermal energies is better than 1%, while those of MA range between 5 and 50% depending on the energy range and the reaction channel [22]. These large uncertainties are related to the facts that:

- the available data sets are not being accurate enough, are incompatible in some cases, do not cover the necessary neutron energy range and the resolution provided is not sufficient;
- the available evaluated cross sections do not agree between themselves. This is related to the fact that the evaluators are forced to question and re-investigate the accuracy of specific data sets in order to reach a self-consistent result in their evaluation for all the reaction channels. Thus, the result on the evaluation depends strongly on the decisions adopted by the evaluator.

In order to set priorities in the measurement of neutron cross sections it is important to identify those playing a major role in the design of an advanced fuel cycle involving ADS. This is the aim of *sensitivity analysis* studies in which the uncertainty of a given cross section is propagated to determine its impact in the calculation of macroscopic quantities used in reactor calculations such as κ_{eff} , β_{eff} , $\Delta\rho^{Doppler}$, $\Delta\rho^{Void}$, etc. As part of the worldwide effort for the development of transmutation technologies, the NEA Nuclear Data High Priority Request List (HPRL)[23] is a continuously upgraded compilation of the most critical nuclear data with the purpose to provide a guide for those planning new measurements and evaluations.

Recent works on uncertainty and target accuracy assessment of nuclear data for ADS [24, 25, 26] has evidenced the need of determining the neutron capture and fission cross sections of a set of minor actinides (^{237}Np , $^{238,239,240,241,242}\text{Pu}$, $^{241,243}\text{Am}$ and ^{245}Cm) with an accuracy better or equal to 5% in the energy range from thermal energies up to 1 MeV. A selection criterion based on the impact on the transmuter neutronics and the availability of capture cross section data allows to set a lower priority to the capture measurements of $^{239,241}\text{Pu}$. In addition, the extreme difficulties involved in the capture measurements of the highly radioactive isotopes like ^{238}Pu ($T_{1/2} = 87.74$ years) and ^{244}Cm ($T_{1/2} = 18.1$ years) reduces further the list of possible measurements to ^{237}Np , $^{240,242}\text{Pu}$, $^{241,243}\text{Am}$ and ^{245}Cm .

These measurements can be performed at the n_TOF facility at CERN [27] with the required accuracy due to the following reasons:

- The neutron energy range between 1 eV and 1 MeV is covered in one single measurement with an excellent energy resolution.

- The Total Absorption technique that will be applied for the measurements is particularly well suited for low mass and radioactive samples.
- The sources of background at n_TOF are very well characterized and can be largely suppressed in combination with the Total Absorption Calorimeter.

For the reasons discussed, the measurement of the neutron capture cross section of ^{237}Np , $^{240,242}\text{Pu}$, $^{241,243}\text{Am}$ and ^{245}Cm at n_TOF was proposed [28] in the range from 1 eV to 1 MeV with an accuracy of 5% and an energy resolution in the unresolved resonance region better than 5%. Such measurements were part of the scientific program of the contract FIKW-CT-2000-00107 between the European Commission and the n_TOF Collaboration participating institutes.

This manuscript is devoted to the first measurements of the capture cross section of minor actinides carried out at n_TOF in 2004 and corresponding to ^{237}Np and ^{240}Pu .

Chapter 2

Neutron cross sections

This chapter introduces the concept of neutron cross sections and presents the theory of the Compound Nucleus and the $R - matrix$ formalism for their description. It also presents different techniques for the measurement and analysis of neutron cross sections, with emphasis on the *time-of-flight* measurements of neutron capture cross sections.

2.1 Introducing neutron cross sections

The microscopic neutron cross section (σ) is a physical quantity that describes the interaction probability of a neutron with a specific nucleus. The wide varieties of nuclear reactions that may occur when a neutron interacts with a given nuclide (capture, scattering, fission, etc.) give rise to the definition of partial cross sections. The total cross section σ_t is defined as the sum of all partial cross sections:

$$\sigma_{total} = \sigma_{capture} + \sigma_{scattering} + \sigma_{fission} + \dots \quad (2.1)$$

The concept of neutron cross section can be understood easily from the experimental point of view. Let's consider a beam of neutrons with intensity $I(neutrons/cm^2/s)$ incident on a very thin plate of a given isotope with area $A(cm^2)$, density of nuclei $N(nuclei/cm^2)$ and thickness $\Delta x(cm)$. In this case, the collision reaction rate $R(reactions/s)$ must be proportional to the intensity of the neutron beam (I) and the number of target nuclei ($N \cdot \Delta x \cdot A$), with proportionality constant σ :

$$R = \sigma \cdot I \cdot (N \cdot \Delta x \cdot A), \quad (2.2)$$

which has the dimension of an area (cm^2). Indeed, if neutrons were classical point particles and nuclei were solid spheres, σ would simply be the cross-sectional area of each nucleus. The cross sections are usually expressed in units of barns ($1 \text{ barn} = 10^{-24} \text{ cm}^2$), which is approximately the cross-sectional area of an uranium nucleus.

The partial cross sections are very different depending on the specific reaction and the energy of the incident neutron. Furthermore, neutron cross sections are completely different from one isotope to another because they are related to the nuclear structure of the nucleus under neutron bombardment.

As an example, Figure 2.1 shows the neutron capture (n,γ) and fission (n,f) cross sections of ^{238}U on an energy scale spanning more than ten decades. At first sight it is observed that there is a very wide range of fourteen orders of magnitude between the smallest and largest cross section values. Furthermore, even within a small energy interval there are strong resonant structures where the peak to valley ratio can be as large as eight orders of magnitude.

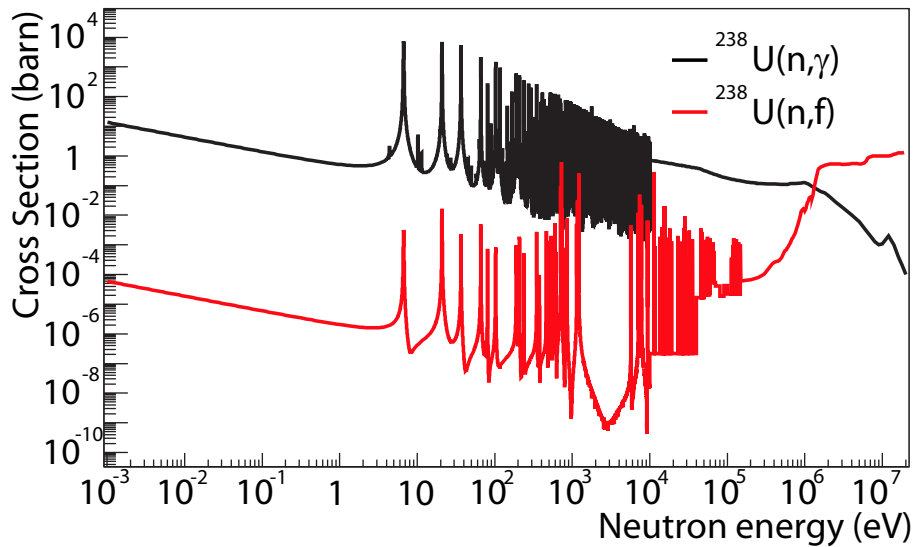


Figure 2.1: Neutron fission and capture cross section of ^{238}U .

The examination of the figure allows distinguishing up to four well differentiated energy regions attending to the shape of the neutron cross sections:

- Thermal and epithermal regions. Between 0.025 eV (thermal point) and the first resonant structure in the eV region, the cross sections are found to be smooth and inversely proportional to the square root of the energy ($\sigma \sim 1/\sqrt{E} \sim 1/v$), that is proportional to the time that the incident neutrons spend in the vicinity of the target nucleus.
- Resolved Resonance Region (RRR). Between a few eV and several keV, depending on the specific isotope, the cross sections show well resolved resonances with large peak to valley variations. The nature of these resonant structures is related to the existence of quasi-stationary levels of the Compound Nucleus ($n + {}^A_Z \rightarrow {}^{A+1}_{Z^*}$) and is discussed in the next section.
- Unresolved Resonance Region (URR). With increasing excitation energy the resonances start to overlap, because their intrinsic widths become comparable to the distance between

neighbouring resonances.

- High energy region. As the neutron energy increases, the distance between resonances is much smaller than their intrinsic widths and resonant structures can not be observed any more. In addition, more and more reaction channels corresponding to threshold reactions open up.

The point wise cross sections displayed in Figure 2.1 correspond to the ENDF/B-VII evaluated data library [21] and consist of 211.752 and 23.306 data points for (n,γ) and (n,f) , respectively. Usually, the cross sections are not provided in a point-wise basis but instead are parameterized in the appropriate theoretical formalisms. For instance, the resonant structures are usually described using the $R - matrix$ formalism in which the shape of each resonance is given by its energy and spin as well as several widths Γ_i related to each open reaction channel. In this way a large number of data points is reduced to just a few parameters for each resonance. Furthermore, one or more resonances with negative energies can be used for describing the $1/v$ region. The $R - matrix$ formalism can be further extended to the URR which can be represented as a large number of overlapping resonances described by a reduced set of average *resonance parameters*. As the energy increases, the cross sections are described using complex models, such as the Optical Potential, based on nuclear structure theory and the information from the cross sections at lower energies.

2.1.1 The Compound Nucleus: neutron resonances

The resonant structures observed in neutron induced reactions can be understood from the point of view of the Compound Nucleus theory, which was first discussed by Niels Bohr in 1939 in his work *The mechanism of nuclear fission* [29]. The basic idea of this theory is that neutron induced reactions take place in two steps.

In a first step, the neutron and the target nucleus form a Compound Nucleus with a high excitation energy that can be approximated by $E^* = S_n + \frac{A}{A+1}E_n$, where A and Z are the mass and charge numbers of the target nucleus, S_n is the neutron separation energy of the Compound Nucleus, and E_n is the energy of the incident neutron. The excitation energy is rearranged among all nucleons and, at some specific neutron energies, gives rise to a complex configuration corresponding to a quasi-stationary level or *resonance* defined by its half-life τ , or the associated width $\Gamma \sim 1/\tau$, as well as its energy, spin and parity. In a second step, the excitation energy is released by any of the open channels, such as emission of radiation (n,γ) , the emission of a neutron with equal or lower energy (n,n') , the fission of the nucleus (n,f) , etc.

Figure 2.2 shows a diagram of the complete neutron capture process, where the resonances in the cross section appear as quasi-stationary levels of the Compound Nucleus. The typical order of magnitude of the neutron separation energy S_n and the average level spacing D at different excitation energies are also indicated in the figure.

In the theory of the Compound Nucleus, the total neutron cross section of a given isotope is

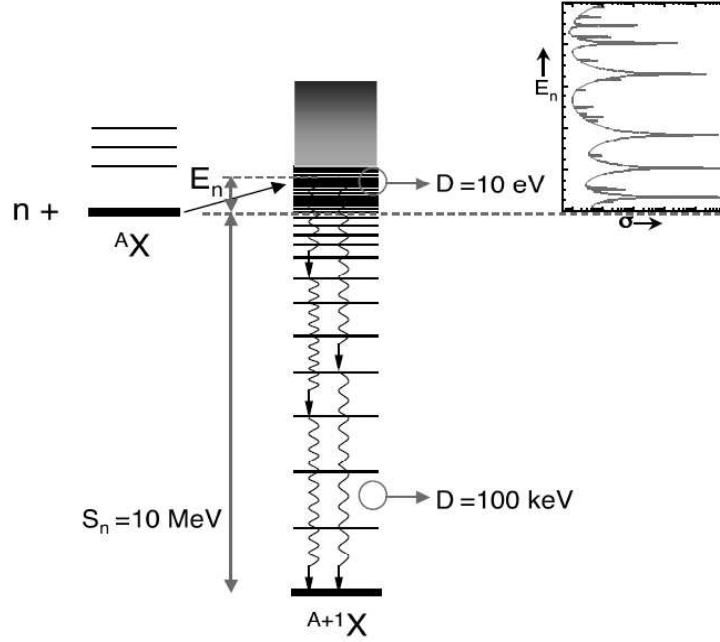


Figure 2.2: Schematic view of the formation of the Compound Nucleus (CN) by neutron capture and its decay to the ground state by emission of γ -rays. The resonances in the cross section appear as quasi-stationary levels of the CN. Typical values of the neutron separation energy (S_n) and the average level spacing (D) at different excitation energies are given.

related to the probability of absorption of the incident neutron to form a Compound Nucleus. The resonances observed in the cross section for the formation of the Compound Nucleus have in good approximation a Breit-Wigner shape [30] determined by the resonance energy E_0 and a set of partial widths related to the decay probability of the Compound Nucleus by the different mechanisms: capture Γ_γ , fission Γ_f , scattering Γ_n , etc., respectively. The sum of all widths Γ_{total} is related to the life-time of the quasi-stationary level of the Compound Nucleus state by the Heisenberg uncertainty principle $\tau = \hbar / \Gamma_{total}$. Since the observed values of Γ_{total} are of the order of electronvolts, the expected half-life is $\sim 10^{-15}$ s, several orders of magnitude larger than the typical time needed by a neutron to cross a nucleus without interaction. The physical foundation of this theory is given by the R – matrix theory.

2.1.2 The R – matrix formalism

The R – matrix formalism was introduced by Wigner and Eisenbud [31, 32] after the World War II in order to give a better physical foundation to the theory proposed by Breit and Wigner for the capture of slow neutrons [30] a few years before. An extensive and detailed overview of the theory was given by Lane and Thomas [33] in 1958 and recently by Fröner [34], whose notation is assumed in this work.

The basic idea of the R – *matrix* formalism is to split the neutron-nucleus system into two geometrical regions. When the neutron is far away from the nucleus, the interaction between them is negligible due to the short range of the nuclear forces; hence, they can be treated as two independent systems with known wave functions. At shorter distances, smaller than the so-called channel radius a_c , all the nucleons interact with each other to form a unique system, the Compound Nucleus. This channel radius is usually expressed as a function of the mass of the nucleus ($a_c \propto A^{1/3}$). The key point of the R – *matrix* formalism is that, although the wave function of the Compound Nucleus is not known, it can be expanded as a linear combination of spherical wave functions that satisfy the boundary conditions at the channel radius a_c imposed by the external wave functions. In this way, each wave function in the internal region contains the properties of a well defined quasi-stationary level λ of the Compound Nucleus, that is a resonance at a given energy E_λ with spin and parity J_λ^Π , and partial widths $\gamma_{\lambda,c'}$ corresponding to the decay probability of each decay mode c' .

In the general theory of nuclear reactions, the entrance and exit channels are defined by the nature, spin (i and I) and energy of the interacting particles. Hence, a channel c is characterized by the quantum numbers $c = \{\alpha, l, s, J\}$, where:

- α : nature, mass, spin and excitation state of the interacting particles;
- ℓ : relative angular momentum of the two particles;
- $\mathbf{s}$: spin of the channel $\mathbf{s} = \mathbf{i} + \mathbf{I}$ with $|I - i| \leq s \leq |I + i|$;
- $\mathbf{J}$: angular momentum of the system $\mathbf{J} = \ell + \mathbf{s}$ with $|\ell - s| \leq J \leq |\ell + s|$.

In the particular case of the incoming neutral particles, Blatt and Biedenharn solved in 1952 [35] the non relativistic Schrödinger equation with the boundary condition "stationary ingoing plane wave + stationary outgoing spherical wave", which integrated over angles gives the expression

$$\sigma_{cc'} = \pi \bar{\lambda}_c^2 g_c |\delta_{cc'} - U_{cc'}|^2 \quad (2.3)$$

where $U_{cc'}$ are the elements of the collision matrix, and therefore $|U_{cc'}|^2$ is the probability of a transition from channel c to channel c' , and the kinematic factor $\pi \bar{\lambda}_c^2$ relates probability and cross section. The spin factor g_c is the probability of getting the correct angular momentum J from the spins of the collision partners:

$$g_J = \frac{2J + 1}{(2i + 1)(2I + 1)} \quad (2.4)$$

where i , I and J are the spins of the incident particle, target nucleus and Compound Nucleus, respectively.

Resonances are introduced by the R-matrix formalism that allows expressing $\mathbf{U}$ in terms of the channel matrix $\mathbf{R}$ (for further details see Lane and Thomas [33]) and, furthermore, in terms of the level matrix $\mathbf{A}$:

$$U_{cc'} = e^{-(\varphi_c + \varphi_{c'})} (\delta_{cc'} + i \sum_{\lambda\mu} \Gamma_{\lambda c}^{1/2} A_{\lambda\mu} \Gamma_{\mu c'}^{1/2}), \quad (2.5)$$

$$\Gamma_{\lambda c} \equiv 2P_c \gamma_{\lambda c}^2 \quad (2.6)$$

$$(\mathbf{A}_{\lambda\mu}^{-1}) = (E_\lambda - E) \delta_{\lambda\mu} - \sum_c \gamma_{\lambda c} L_c^0 \gamma_{\mu c} \quad (2.7)$$

where Roman and Greek subscripts refer to reaction channels and compound levels, respectively.

The resonance parameters are level energies E_λ and the probability amplitudes $\gamma_{\lambda c}$ of decay (or formation) of compound nuclear states λ via exit (or entrance) channels c . The signs of $\gamma_{\lambda c}$ are practically random except near the ground state. The centrifugal barrier penetrabilities P_c that relate the partial widths $\Gamma_{\lambda c}$ with the probability amplitudes $\gamma_{\lambda c}$ are a function of the type of particles in the channel, of the orbital angular momentum ℓ , and of the neutron energy E . In the case of γ and fission channels they are very slowly varying with energy and hence are usually considered as constants. On the other hand, the penetrability factor of the neutron channel depends on the neutron energy in such a way that, for s -wave resonances

$$P_{\ell=0} = a_c \kappa_c = a_c \sqrt{\frac{2M_\alpha E}{\hbar^2}}, \quad \text{where } \kappa_c \equiv \text{wave number} \quad (2.8)$$

and the relation becomes

$$\Gamma_{\lambda n}(E) = 2a_c \sqrt{\frac{2M_\alpha E}{\hbar^2}} \gamma_{\lambda n}^2 \propto \sqrt{E} \gamma_{\lambda n}^2 \quad (2.9)$$

showing a direct proportionality with $\sqrt{E}$.

For incident neutrons, the hard-sphere phases φ_ℓ and the logarithmic derivatives L_ℓ in Equation 2.5 can be expressed as

$$L_0 = ia_c \kappa_c, \quad L_\ell = -\ell - \frac{(a_c^2 \kappa_c^2)}{L_{\ell-1} - \ell} \quad (2.10)$$

$$\varphi_0 = a_c \kappa_c, \quad \varphi_\ell = \varphi_{\ell-1} + \arg(\ell - L_{\ell-1}) \quad (2.11)$$

2.1.3 Approximations to the *R-matrix* formalism

The resonance parameters E_λ and $\gamma_{\lambda c}$ depend on the unknown nuclear interaction and therefore are used as adjustable parameters in the comparison between calculated cross sections and experimental data. However, in the calculation of the total and partial cross sections the inversion of the level matrix $\mathbf{A}$ is in general impossible without formulating some assumptions. For instance it is possible to consider only one level (Single Level Breit-Wigner approximation)

$$(A^{-1})_{\lambda\mu} = (A^{-1}) = E_0 - E - \sum_c L_c^0 \gamma_c^2 \equiv E_0 + \Delta - E - \frac{i}{2} \Gamma, \quad (2.12)$$

where the level shift Δ_λ and the total width Γ_λ are real; or many non correlated levels (Multi Level Breit-Wigner approximation)

$$(A^{-1})_{\lambda\mu} = (E_\lambda - E - \sum_c L_c^0 \gamma_{\lambda c}^2) \delta_{\lambda\mu} \equiv (E_\lambda + \Delta_\lambda - E - \frac{i}{2} \Gamma_\lambda) \delta_{\lambda\mu}, \quad (2.13)$$

or to neglect only the correlation between the photon channels (Reich-Moore approximation)

$$(A^{-1})_{\lambda\mu} = (E_\lambda + \Delta_\lambda - E - \frac{i}{2} \Gamma_{\lambda\gamma}) \delta_{\lambda\mu} - \sum_{c \neq \gamma} \gamma_{\lambda c} L_c^0 \gamma_{\mu c}. \quad (2.14)$$

Other approximations are those of Wigner-Eisenbud [32], Kapur-Peierls [36] and Adler-Adler [37].

In the next section follows the derivation of the complete cross section expressions in terms of the resonance parameters for the most simple but maybe more illustrative Single Level Breit-Wigner (SLBW) approximation.

2.1.3.1 The Single Level Breit-Wigner approximation

In the Single Level Breit-Wigner (SLBW) approximation only one level is considered and therefore the level matrix can be expressed from Equation 2.12 as:

$$(A^{-1})_{\lambda\mu} = (A^{-1}) = E_0 - E - \sum_c L_c^0 \gamma_c^2 \equiv E_0 + \Delta - E - \frac{i}{2} \Gamma, \quad (2.15)$$

where the level shift is the difference between the level and resonance energies $\Delta = E_0 - E_r$. The collision matrix of Equation 2.5 can be then expressed as

$$U_{cc'} = e^{-i(\varphi_c + \varphi_{c'})} \left(\delta_{cc'} + \frac{i\sqrt{\Gamma_c \Gamma_{c'}}}{(E_0 - E) + \Delta - \frac{i}{2} \Gamma} \right). \quad (2.16)$$

For incident neutrons and choosing the boundary conditions in such a way that the level energy is the same as the resonance energy ($\Delta=0$) and defining the effective radius of the Compound Nucleus as $R = a_c$

$$U_{nc'} = e^{-2i\kappa R} \left(\delta_{nc'} + \frac{i\sqrt{\Gamma_n \Gamma_{c'}}}{(E_0 - E) - \frac{i}{2} \Gamma} \right). \quad (2.17)$$

In the case of a s -wave resonance with spin J and noting that $\kappa R \ll 1$, the resulting total cross section is given by

$$\sigma_n^J = 4\pi g_J R_J^2 + 4\pi \bar{\lambda}^2 g_J \left(\frac{\Gamma_n \Gamma}{\Gamma^2 + 4(E_0 - E)^2} + 4\kappa R_J \frac{\Gamma_n (E_0 - E)}{\Gamma^2 + 4(E_0 - E)^2} \right), \quad (2.18)$$

where the first term is the potential scattering proportional to the effective radius of the square of the Compound Nucleus R_J^2 ; the second term is a resonant term symmetric in energy; and the third is an interference term asymmetric in energy around the resonance energy E_0 .

The partial cross sections are given by

$$\sigma_{nx}^J = 4\pi\bar{\lambda}^2 g_J \frac{\Gamma_n \Gamma_x}{\Gamma^2 + 4(E_0 - E)^2}, \quad x \neq n, \quad (2.19)$$

which is symmetric in energy, and $\sigma_{nn} = \sigma_n - \sum_x \sigma_{nx}$ with $x \neq n$.

2.1.4 Statistical properties of the Resonance Parameters

The study of the statistical properties of the resonance parameters describing the cross section in the RRR are key for: (i) testing the consistency between all of them because they are usually linked by some physical constraints, and (ii) to collect the information from isolated resonances for its use in the description of the cross sections at higher neutron energies.

In the case of heavy nuclei with a large number of nuclear levels below the neutron separation energy S_n , the quasi-stationary levels above S_n show the characteristic features of the statistical model. The application of this feature to the field of resonances in nuclear reactions is referred to as the Gaussian Orthogonal Ensemble (GOE) and was first introduced by Wigner [38] and further developed by Dyson [39] and Porter [40].

The next sections summarize some of the statistical properties that should be observed in the study of the complete set of resonance parameters describing the RRR.

2.1.4.1 Average resonance spacing and probability distribution

The distribution of observed resonances as a function of neutron energy is directly related to the level density of the Compound Nucleus at the neutron separation energy. Indeed, the level density ρ^J can be calculated from the number of observed resonances N^J of a given spin J in an energy interval ΔE_n as

$$\rho^J = \frac{N^J}{\Delta E_n} = \frac{1}{\langle D^J \rangle}, \quad (2.20)$$

where D^J is the average level spacing for the spin J . The latter can be calculated from the cumulative distribution of observed resonances $N^J(E_n)$:

$$N^J(E_n) = a + \langle D^J \rangle E_n \quad (2.21)$$

There are several theoretical models aimed at the prediction of nuclear level densities [41] as a function of the mass number that usually include shell and pairing effects. For instance, the

spin dependence of the level density can be studied from the observed resonances and compared with level density formulas such as the Fermi Gas [42] or the Gilbert-Cameron [43]. The latter predicts a dependence that introduces the spin cut-off parameter σ :

$$\rho^J \propto e^{-\frac{J^2}{2\sigma^2}} - e^{-\frac{(J+1)^2}{2\sigma^2}} \quad (2.22)$$

The spacing between consecutive resonances for the same total angular momentum and parity should also exhibit a random behaviour that can be tested experimentally. For a set of N consecutive resonances with energies E_i , the level spacing is defined as $D_i = E_i - E_{i-1}$ and its probability distribution function should follow a distribution close to the Wigner law [44]:

$$p(x)dx = \frac{\pi x}{2} \exp\left(-\frac{\pi x^2}{4}\right) dx \quad \text{with } x = \frac{D_k}{\langle D \rangle}. \quad (2.23)$$

The Wigner law, which corresponds to the limiting case for a chaotic system, was the first mathematical prediction of the level spacing to provide excellent agreement with the experimental results. An interesting feature is that the distribution goes to zero for small separations between levels, which is known as the repulsion effect. The average value of the distribution is $\langle x \rangle = 1$ and its variance $\sigma^2 = (4/\pi - 1)$. Therefore the relative uncertainty of the average level spacing calculated from N resonances is:

$$\frac{\Delta \langle D \rangle}{\langle D \rangle} = \sqrt{\frac{1}{N} \left(\frac{4}{\pi} - 1 \right)}. \quad (2.24)$$

2.1.4.2 Resonance widths distribution

The resonance widths Γ_λ of each quasi-stationary level λ are defined (see Equation 2.6) by the penetrability factor P_α and the amplitude probabilities $\gamma_{\lambda\alpha}$ as

$$\Gamma_{\lambda c} = \sum_{\alpha=1}^{\nu} 2P_\alpha \gamma_{\lambda\alpha}^2 \quad (2.25)$$

where ν is the number of possible de-excitation modes c , and $\gamma_{\lambda\alpha}$ is known to follow a Gaussian distribution.

Assuming that P_α is nearly constant in the energy range under study and that $\langle \gamma_{\lambda\alpha} \rangle$ is constant for each level λ decaying via α , the distribution of widths $\Gamma_{\lambda c}$ is given by a χ^2 distribution with ν degrees of freedom (see Ref. [45])

$$p(x, \nu)dx = \frac{\frac{\nu}{2}}{\Gamma(\frac{\nu}{2})} \left(\frac{\nu}{2}x\right)^{\frac{\nu}{2}-1} e^{-\frac{\nu}{2}x} dx \quad \text{with } x = \frac{\gamma_{\lambda c}^2}{\langle \gamma_{\lambda c}^2 \rangle} = \frac{\Gamma_{\lambda c}}{\langle \Gamma_{\lambda c} \rangle} \quad (2.26)$$

with mean value 1 and variance $\frac{2}{\nu}$.

The assumption made in the previous paragraph stands for all exit channels except for $c = n$ (neutron scattering). However, it is possible to define a reduced neutron width $\Gamma_{\lambda n}^0$ independent of energy as

$$\Gamma_{\lambda n}^0 = \Gamma_{\lambda n}(E_\lambda) \sqrt{1/E} \propto \gamma_{\lambda n}^2, \quad (2.27)$$

in such a way that the reduced neutron widths is directly proportional to the square of the amplitude probability $\gamma_{\lambda c}$, similarly to the case of all other partial widths.

Since the neutron scattering reactions have only one decay channel ($\nu = 1$), the equation above turns into the well known Porter-Thomas distribution of the reduced neutron width $\Gamma_{\lambda n}^\ell$ [45]

$$p(x)dx = \frac{1}{\sqrt{2\pi c}} e^{-\frac{x}{2}} dx, \quad (2.28)$$

On the contrary, the radiative width $\Gamma_{\lambda\gamma}$ for heavy nuclei is expected to be the sum of many partial widths $\Gamma_{\lambda\gamma i}$ corresponding to the decay to each of the many nuclear levels existing below S_n . In the limit of infinite partial widths the χ^2 distribution turns into a Dirac delta function, which implies that all radiation widths have a constant value

$$\Gamma_{\lambda\gamma} = \Gamma_{\mu\gamma} = \sum_i \Gamma_{\lambda\gamma i} = \langle \Gamma_\gamma \rangle. \quad (2.29)$$

This constant behaviour of the radiative width for heavy nuclei can be also understood from the fact the number of primary transition from a level λ or from a higher energy level μ are practically the same.

The neutron induced fission can be considered as a few channels process. Hence its width distribution should follow a χ^2 with a few degrees of freedom. Indeed, the value of ν is fitted from experimental data and usually found between 2 and 3.

2.1.4.3 Strength function

The neutron strength function S_l is, together with the average level spacing $\langle D \rangle$ and radiative width $\langle \Gamma_\gamma \rangle$, one of the main ingredients of the optical model calculations for high energy neutron cross sections [46]. For a given angular momentum ℓ , the strength function is calculated as

$$S_l = \frac{\langle g_l \Gamma_{\lambda n}^\ell \rangle}{\langle D_l \rangle} = \frac{1}{\Delta E} \sum_J \sum_\lambda g_J \Gamma_{\lambda n}^{\ell, J}, \quad (2.30)$$

where ΔE is the energy range considered and $\Gamma_{\lambda n}^{\ell, J}$ is the reduced neutron width for a resonance with spin J and orbital momentum ℓ . The uncertainty of the strength function when it is calculated from a set of N resonances in the range ΔE is given by

$$\frac{\Delta S_l}{S_l} = \sqrt{\left(\frac{4}{\pi} + 1\right) \frac{1}{N}}. \quad (2.31)$$

2.2 Measurement of neutron cross sections

There exist several experimental methods to measure the total and partial neutron cross sections. All of them require a neutron source and a sample as isotopically pure as possible. Cross sections can be measured at specific energies using monoenergetic neutron beams, or in the form of point-wise cross sections or average values using a continuous energy neutron beam. The most complete measurements are those of the point-wise cross section of all open reaction channels in the widest possible energy range, which is only possible by means of the *time-of-flight* method.

2.2.1 The *time-of-flight* method

The *time-of-flight* (TOF) method is based on the direct relation that exists between the energy of a neutron and the time (time-of-flight) that it needs to travel a given distance.

In this type of measurements, the neutrons are produced by a pulsed neutron source at time t_0 after which the neutrons travel towards the sample placed at a distance L , which can range from a few centimetres to hundreds of meters. The reactions of interest take place and are detected at a time t_{det} that is directly related to the energy of the incident neutron by the relativistic formula

$$E_n = E_{tot} - mc^2 = \frac{mc^2}{\sqrt{1 - v^2/c^2}} \quad (2.32)$$

where c is the speed of light. The first term of the series expansion gives the classical expression for the neutron kinetic energy

$$E_n(eV) = \frac{1}{2}m_n v^2 = \left(\frac{72.2983 \cdot L(m)}{t_{det}(\mu s) - t_0(\mu s)} \right)^2. \quad (2.33)$$

Hence, the resolution of a *time-of-flight* facility is determined by the length of the flight path and its uncertainty, the time resolution of the detection system, and the Resolution Function related to the spread in the *time-of-flight* related to the different distances λ followed by the neutrons of the same energy before escaping the neutron production target. The latter is characteristic of the production mechanism and the geometry of the production target. For the specific case of the n.TOF facility it is discussed in Section 3.1.3.

2.2.2 Experimental facilities

The time-of-flight facilities can be classified by the mechanism used for neutron production and the length of the flight path. The first is responsible of the energy distribution of the neutrons in the beam, the integrated and instantaneous intensity of the neutron beam, the

in-beam background, and the Resolution Function of the facility. The second determines the resolving power of the experiment.

Neutrons can be produced by a large variety of mechanisms: spontaneous fission, α decay induced reactions (Am/Be), proton ${}^7\text{Li}(p, n)$ or deuterium $D(T, n)$ induced reactions, photofission, spallation, etc. Most facilities worldwide are based on the use of electron or proton accelerators inducing photofission or spallation reactions, respectively. These facilities produce an initially fast neutron spectrum that is generally degraded in energy by placing a moderator between the production target and the neutron beam line.

The electron-based facilities produce neutrons via photon-induced reactions in a heavy target using the photons produced by Bremsstrahlung emission of the high energy electrons impinging on the target itself. Examples of this type of white pulsed neutron sources are GELINA [47] of the EC-JRC-IRMM at GEEL in Belgium, using 140 MeV electrons incident on an uranium target; ORELA [48] at the Oak Ridge National Laboratory in USA, using 180 MeV electrons on a tantalum target; the RPI facility [49, 50] at Troy in USA; and the KURRI facility [51] at Kyoto in Japan.

The proton-based facilities produce neutrons as secondary particles from reactions in the region up to 100 MeV or by spallation process at higher energies. Examples are the n.TOF facility [27, 52] at CERN (Switzerland), the LANSCE facility [53] at Los Alamos National Laboratory (USA) and the KEK [54] facility in Kyoto (Japan).

	n.TOF	LANSCE	ORELA	GELINA	nELBE
Accelerated particles	p^+	p^+	e^-	e^-	e^-
Production mechanism	spallation	spallation	(γ, f)	(γ, f)	(γ, n)
Pulse length (ns)	7	125	4	1	0.4
Frequency (Hz)	0.4	20	500	800	$5 \cdot 10^5$
Flight Path (m)	185	20	40	20	4
E_{min} (eV)	0.1	1	10	10	$5 \cdot 10^4$
E_{max} (eV)	10^8	10^8	$5 \cdot 10^6$	$4 \cdot 10^6$	$\cdot 10^7$
$\Delta E/E$ @ 1 MeV	0.5%	10%	$< 1\%$	$< 2\%$	1%
$\Phi_n(n/cm^2/s/decade)$	10^5	$3 \cdot 10^5$	10^4	$4 \cdot 10^4$	$3 \cdot 10^6$
$\Phi_n(n/cm^2/pulse/decade)$	$2.5 \cdot 10^5$	$1.5 \cdot 10^4$	20	50	6

Table 2.1: *Main characteristics of some of the mentioned time-of-flight facilities.*

Table 2.1 summarizes the main characteristics [55] of some of the mentioned time-of-flight facilities, i.e. the range of flight-paths, pulse frequencies and neutron fluxes. For those facilities having more than one flight-path, the most frequently used flight-path has been chosen for the summary. It is observed that the facilities based on electron accelerators reach higher frequencies while the spallation driven facilities reach much higher instantaneous neutron flux. The latter feature is of major importance when measuring radioactive targets because a high instantaneous neutron flux maximizes the ratio of neutron induced reactions to the background caused by the

activity of the samples. The combination of the highest instantaneous neutron flux with the longest available flight path indicates that the n-TOF facility is the best suited facility for measuring partial cross sections of radioactive samples.

2.2.3 Measuring techniques

2.2.3.1 Total cross section measurements

The total cross section σ_t is measured by placing a neutron detector at the end of the neutron beam line and counting the number of neutrons (C) in two situations: with a sample of surface density n intercepting the beam C_{in} , and without any sample along the beam C_{out} .

This technique, usually referred as *sample-in/sample-out*, directly gives the dimensionless neutron transmission as a function of neutron energy, which is related to the total cross section by

$$T(E_n) = N \frac{C_{in}(E_n) - B_{in}(E_n)}{C_{out}(E_n) - B_{out}(E_n)} = e^{-n\sigma_t(E_n)}, \quad (2.34)$$

where $B_{in/out}(E_n)$ refers to the background contribution to the number of detected neutrons, and N is a normalization factor that takes into account the different number of neutrons allocated to *in* and *out* measurements. In this way the detector efficiency cancels out and there is a low uncertainty in the normalization, only related to the monitoring of the number of incident neutrons in each measurement.

2.2.3.2 Partial cross section measurements

Partial cross sections as a function of neutron energy are measured indirectly by detection of the secondary particles emitted in each process, for instance γ -rays, neutrons or fission fragments following (n,γ) , (n,n') and (n,f) reactions, respectively. The observable quantity in these measurements is the counting rate C_x which is related to the reaction yield by

$$Y_x(E_n) = \frac{C_x(E_n) - B(E_n)}{\varepsilon_{n,x}\Phi_n(E_n)}, \quad (2.35)$$

where B accounts for the background counts, $\varepsilon_{n,x}$ is the efficiency of the detector for the reactions under study (n,x) , and Φ_n is the incident neutron flux. The systematic uncertainties in these type of measurement are given by the accuracy in the determination of the detection efficiency, the characterization of the background and the neutron fluence.

The theoretical value of the reaction yield, defined as the fraction of the beam that undergoes a certain reaction (n,x) , is a function of the surface density of the sample n , and the total and partial cross sections. The first approximation can be expressed as

$$Y_x(E_n) = \{1 - e^{-n\sigma_t(E_n)}\} \frac{\sigma_x(E_n)}{\sigma_t(E_n)} \quad (2.36)$$

where the first term is related to the probability that a neutron induced reaction takes place, and the second term gives the probability that such a reaction is the one under investigation.

For very thin samples where $e^{-n\sigma_t} \sim 0$, the yield becomes directly proportional to the target thickness and the reaction cross section $Y_x \simeq Y_0 \simeq \sigma_x n$. On the contrary, for thick samples showing large values of $n\sigma_t$ there is a non negligible probability that the incident neutrons are scattered one or more times within the sample before undergoing the reaction of interest. In such cases, the reaction yield becomes a sum of contributions from multiple-collision events where the neutron undergoes zero, one, two, etc. scattering collisions before it finally induces the reaction of interest,

$$Y_x = Y_0 + Y_1 + Y_2 + \dots \quad (2.37)$$

where the *zero* term is given by equation 2.36, and the higher order terms are increasingly complicated functional of the cross section which can be calculated by means of Monte Carlo simulations.

2.2.3.3 Broadening effects

There are two experimental effects that broaden the shape of the resonances and that must be taken into account in *time-of-flight* measurements:

1. The Thermal broadening, which is related to the thermal motion of the target nuclei. Due to the thermal motion of nuclei in the sample, a neutron approaching the nucleus with a given velocity in the laboratory system will have a spread in its velocity in the centre of mass system, which is known as the Doppler effect. In good approximation, for metallic samples the motion of the target can be described by the Free Gas model (FGM), resulting in a Gaussian broadening of the resonances with a standard deviation

$$\sigma_D = \sqrt{\frac{2\kappa_B T}{M/m}} \quad (2.38)$$

where M/m the ratio of the masses of the target nucleus and the incident particle. For crystalline lattices a more complicated description, the Crystal Lattice Model (CLM), is sometimes needed [56].

2. The Resolution broadening, which is caused by the fact that neutrons of a given energy do not reach the target at the same time but with a distribution that is known as the Resolution Function. This spread in time is directly related with the different path that each neutron follows inside the production target and the neutron moderator before it starts to travel along the flight path. The Resolution Function is characteristic of each facility and depends mainly on the neutron production mechanism and the size of the target. The specific case of the n_TOF facility is discussed in some detail in Section 3.1.3.

2.2.4 Neutron capture cross section measurements

There are a wide variety of techniques for measuring neutron capture cross sections, which can be determined either as an average over a certain energy range or point-wise as a function of neutron energy. Concerning the first type of measurements, the most common ones are based on the activation and the pile oscillation methods:

- The activation method [57] consists in the irradiation of a sample $^A Z$ with a known neutron flux and the subsequent spectroscopic, radiochemical or even mass spectroscopy analysis of the sample in order to calculate the number of reaction products $^{A+1} Z$.
- The pile oscillation method [58] consists in the insertion of an oscillating sample into a well characterized nuclear reactor. The cross section of the isotope is determined from the variations induced by the pile oscillations in the reactivity of the system.

The neutron capture cross section as a function of the neutron energy is determined in time-of-flight measurements by detecting the electromagnetic (EM) cascade following the capture reactions. The most common type of detectors used in (n, γ) measurements are:

1. Ge detectors. In spite of their low intrinsic efficiency and bad timing (if compared to scintillators), HPGe detectors have been used for (n, γ) measurements due to their excellent energy resolution that allows discriminating between the γ -rays emitted by capture reactions in the sample under study and background γ -rays [59, 60]. However, this technique requires knowing the decay pattern of the compound nucleus $^{A+1} Z$.
2. Moxon-Rae detectors. M. C. Moxon and E. R. Rae [61] developed in 1963 a new type of detector using the idea that a Geiger tube with a thick wall of low Z material has a γ -ray detection efficiency that increases almost linearly with the energy of the γ -ray. This property implies that the detection efficiency for detecting a capture event is almost independent of the γ -rays emitted in the cascade, and varies linearly with the binding energy of the Compound Nucleus under study. The problem of the long decay time of the Geiger tube was overcome by replacing the Geiger tube by a thin piece of plastic scintillator and a photomultiplier as the electron detector.
3. Total energy detectors. H. Maier-Leibnitz showed that for a wide range of radiation detectors one can generate an average response function of the detector proportional to energy after applying a weighting function which is only proportional to the pulse height. In this way it is possible to recover the total energy proportionality characteristic of the Moxon-Rae detector but have some freedom in the optimization of the detection system in terms of the efficiency, time resolution, sensitivity to scattered neutrons, etc. The pulse height weighting technique (PHWT) was first applied to total energy detectors in 1967 by R.L. Macklin et al. [62, 48] at ORELA [63]. Since then the PHWT has been improved [64, 65] and new total energy detectors have been used at many facilities [66, 67, 68].
4. Total absorption detectors. This technique is based on the detection of the complete EM cascade following a neutron capture reaction. The desirable features of a good total

absorption detector are [61] large solid angle coverage, high photopeak efficiency, good energy resolution, high segmentation, low neutron sensitivity, and fast time response. The efficiency of such a detector is independent of the γ -ray cascade and its response to capture cascades is, ideally, a peak in the pulse height spectrum at the neutron separation energy of the Compound Nucleus. The high photopeak efficiency and the segmentation of the detector provide additional means for the identification and subsequent reduction of the different sources of background: sample radioactivity, reactions produced by scattered neutrons, etc. These type of detectors are found, among others, at n_TOF (41 BaF₂ crystals) at CERN in Switzerland, at FZK (41 BaF₂ crystals [69]) in Germany, at KURRI (4 π Ge [70] and 16 BGO crystals [71]) in Japan, at RPI (16 NaI crystals [72]) in USA, and at LANL (162 BaF₂ crystals [73]).

Since the aim of this work is the measurement of the neutron capture cross section of ²³⁷Np and ²⁴⁰Pu, both radioactive and only available in small amounts, the total absorption technique has been identified as the best suited mainly because of its background reduction capabilities and high detection efficiency.

2.3 Analysis of neutron cross sections

The present sections contains a brief introduction to the analysis of cross section measurements in the resolved resonance region, with emphasis on the information that can be extracted from the observables of total and partial cross section measurements, i.e. transmission coefficients $T(E_n)$ and reaction yields $Y_x(E_n)$.

2.3.1 Resonance analysis

The resonance parameters of the $R - matrix$ formalism which describe each resonance can be extracted from the analysis of the observable quantities in total and/or partial (n, x) cross section measurements like the transmission coefficients $T(E_n)$ and reaction yields $Y_x(E_n)$.

In principle, it is possible to extract the resonance parameters from the analysis of the area of the resonances observed in $T(E_n)$ and $Y_x(E_n)$. In the case of transmission measurements the area of each resonance is given by

$$A_t = \int (1 - T(E_n)) dE_n = 2\pi^2 n \bar{\lambda}_0^2 g_J \Gamma_n, \quad (2.39)$$

where $\bar{\lambda}_0$ is the reduced wave length of the neutrons at the resonance energy. For a particular reaction yield and assuming the thin sample approximation, the area below each s -wave resonance is given by

$$A_x = \int \sigma_x(E_n) dE_n = n \int Y_x(E_n) dE_n = 2\pi^2 n \bar{\lambda}_0^2 g_J \frac{\Gamma_n \Gamma_x}{\Gamma}. \quad (2.40)$$

If the measurements have enough energy resolution and statistics, it is more accurate to calculate the parameters (E_0 , J , Γ_n , Γ_γ , ...) describing each resonance by fitting the shape of the experimental transmission and reactions yields. Such fits require the calculation of a theoretical transmission and/or reaction yields from the resonance parameters, including a set of experimental effects such as the resolution function of the facility, the Doppler broadening of the resonances, the self-shielding and the multiple scattering, etc. These experimental corrections to the theoretical observables are usually calculated with auxiliary codes such as SAMMY [74] or REFIT [75], which are further used to adjust the resonance parameters to the experimental data by means of Bayesian or Least-Squares fitting procedures, respectively.

In order to reduce the uncertainties of the fitted values and their correlations it is desirable to analyze at the same time the observables corresponding to all open reaction channels (transmission, capture, fission, ...) measured at different temperatures (for an accurate estimation of the Doppler broadening) and with samples of different thickness (for an accurate estimation of the self-shielding and multiple scattering effects).

2.3.2 Analysis of several data sets

The analysis of several data sets can be performed sequentially or simultaneously, each option having its advantages and disadvantages. The first option is used in SAMMY and the second is used in the REFIT code.

In any case the analysis of several data sets proceeds as follows (see [76, 34, 77] for further details):

1. **Identification of incompatibilities.** A preliminary analysis of each data set and the comparison of the results can help to identify incompatibilities such as wrong energy calibrations, a different number of resonances indicating the existence of impurities in the samples, etc.
2. **Resonance analysis of small energy regions.** Small energy regions must be studied individually analyzing together the transmission data and the partial yields in order to determine the partial widths Γ_n and Γ_x and to reduce the correlation between them.
5. **Final resonance analysis.** The results of the previous analysis must be used as a starting point for the analysis of the complete energy range including all the experimental data, resulting in a complete and consistent set of resonance parameters and of the associated covariance matrix.

2.3.3 The SAMMY code

The SAMMY code [78, 74] was developed at Oak Ridge National Laboratory by Dr. Nancy Larson and has been widely used during the last decades for the analysis and evaluation of

neutron cross sections in the thermal, resolved and unresolved resonance regions. Since there is a very detailed and complete manual [74] available at the SAMMY website [78], only the most important features are presented here.

In the Resolved Resonance Region (RRR), SAMMY uses the multichannel multilevel R – $matrix$ formalism and calculates the experimental corrections (Doppler broadening, self-shielding, etc.) for fitting the calculated transmission and/or yields to the experimental data. The theoretical yields and transmission are calculated via the Reich-Moore approximation to R - $matrix$ theory, in which each channel is characterized by the spin of the two particles, their relative angular momentum, the spin of the resonance and its parity. All the parameters used for the theoretical calculation, mainly the resonance energy and width but also other parameters such as the temperature and composition of the sample, can be fitted or kept fixed.

In the Unresolved Resonance Region (URR), SAMMY incorporates Fröhner’s code FITACS [79] with only a few modifications [74]. This code uses Hauser-Feshbach theory with width fluctuations, in which the adjustable parameters are the strength functions, distant-level parameters, average radiation widths and average fission widths for each spin group. These quantities can be determined from the resonance analysis and used as a starting value from the analysis of the URR. In this energy range the quantities to be fitted are not the transmission and reaction yields but the total and reaction cross sections, respectively.

The fitting procedure used in SAMMY is the *Bayes’ method*. It incorporates all uncertainties in the measured data, including both statistical and systematic. While the statistical uncertainties contribute to the diagonal elements of the data covariance matrix, the systematic uncertainties involved in the data reduction process (detector efficiency, background, normalization, isotopic abundances in the sample, etc) are included in the fitting process in the non-diagonal part of the data covariance matrix. The final result of an analysis with SAMMY, in both the resolved and unresolved resonance regions, is a set of resonance parameters with their uncertainty and the associated covariance matrix.

Part II

MEASUREMENTS AND DATA REDUCTION

Chapter 3

The experimental set-up

The neutron capture cross section measurements presented in this work were performed at the n_TOF facility using the segmented BaF₂ Total Absorption Calorimeter. The aim of this chapter is to describe the n_TOF facility (neutron fluence, beam profile, energy resolution, etc.), the Total Absorption Calorimeter, the Data Acquisition System and the samples used in the measurements.

3.1 The n_TOF facility at CERN

The construction of the n_TOF facility [27] was proposed by Rubbia et al. [80] in 1998 and built at CERN in 2000-2001. The facility provides the scientific community of nuclear technology and astrophysics with a powerful tool for the measurement of neutron cross sections in an energy range from thermal energies up to 250 MeV.

At n_TOF, neutrons are produced by spallation reactions in a lead block. Figure 3.1 offers a general view of the n_TOF facility with details of the lead spallation target and the experimental area. The CERN Proton Synchrotron (PS) delivers 6 ns wide pulses of protons at 20 GeV/c with an intensity of $7 \cdot 10^{12}$ or $4 \cdot 10^{12}$ protons per pulse, corresponding to dedicated or parasitic pulses, respectively. The proton beam hits the lead spallation target of $80 \times 80 \times 60$ cm³ and generates by spallation about 600 neutrons per proton with a repetition rate of 0.4 Hz. This very low is a great advantage for measuring radioactive samples because the signal (neutron induced reactions in the sample) to background (activity of the sample) ratio is highly favourable.

The lead block is immersed in water that serves both as coolant and as moderator that transforms the initially fast neutron spectrum into a white spectrum down to thermal energies. The neutrons generated in the lead block travel towards the experimental area in an evacuated beam line that is shifted by an angle of 10° with respect to the incident proton direction in order to reduce the amount of charged particles and γ -rays which are produced mainly in the

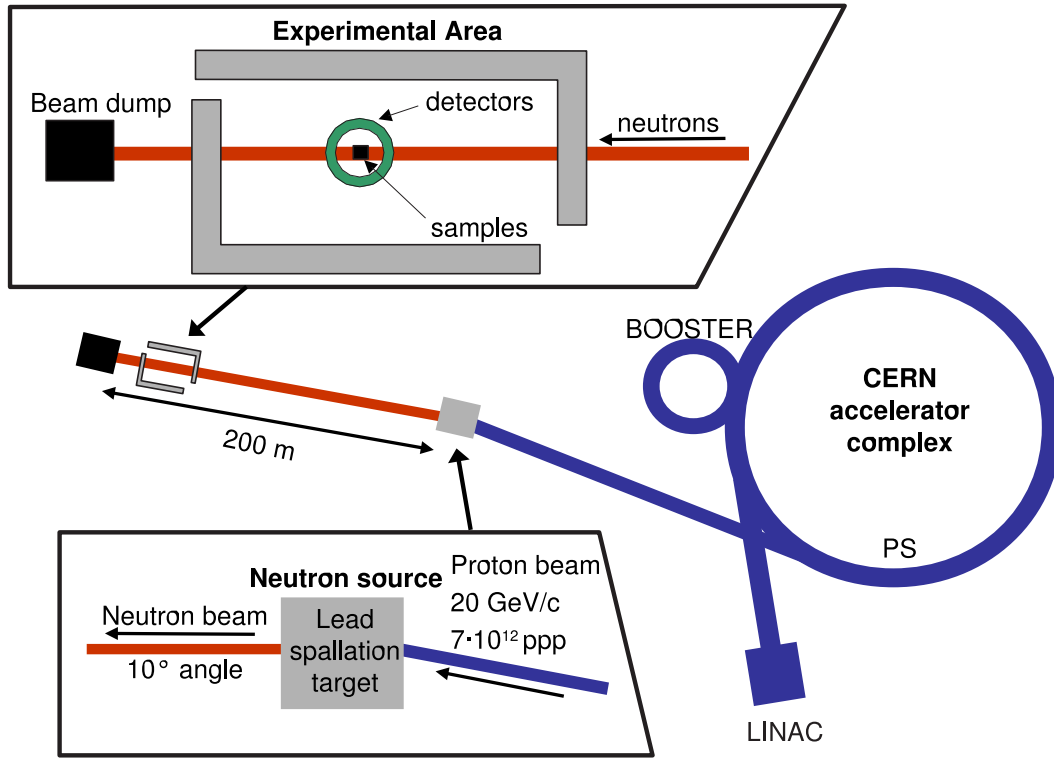


Figure 3.1: General view of the *n*-TOF facility with details of the lead spallation block and of the experimental area.

forward direction. Furthermore, a set of magnets and collimators are placed along the beam line, see Figure 3.2, for reducing the number of charged particles in the beam and for shaping the neutron beam, respectively.

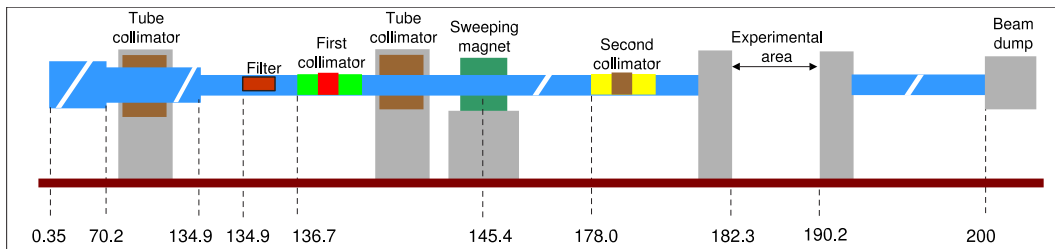


Figure 3.2: *n*-TOF beam line up to the end of the tunnel (200 m).

The experimental area (EAR-1), where the samples and detection systems are placed, is a 7.5 m long room starting at 182.5 m downstream of the spallation target. The beam line extends 10 meters beyond the end of the experimental area and ends in a beam dump consisting of a 49x49x47 cm³ polyethylene block containing three BF₃ monitors.

3.1.1 Neutron fluence

The energy shape of the neutron fluence at the measuring station was determined using two calibrated fission chambers from PTB (Physikalisch-Technische Bundesanstalt) [81] and other auxiliary detection systems such as a silicon detector viewing a ^6Li foil, C_6D_6 gamma-ray detectors and Parallel Plate Avalanche Counters (PPAC) [27].

The PTB detection system consisted of two identical parallel plate ionization chambers with deposits of ^{238}U (197.78 ± 0.60 mg) and ^{235}U (201.56 ± 0.60 mg), equivalent to 436 and 444 $\mu\text{g}/\text{cm}^2$, respectively. Using such thin fission targets, the thin target approximation can be assumed: the measured counting rate $R_{n,f}(E_n)$ should be directly proportional to the incident neutron intensity $\Phi_n(E_n)$ and to the fission cross section $\sigma_f(E_n)$ of ^{235}U , which is a standard. Hence, the neutron intensity can be calculated from the counting rate as

$$\Phi_n(E_n) = \frac{R_{n,f}(E_n)}{n \cdot \sigma_f(E_n)} \quad (3.1)$$

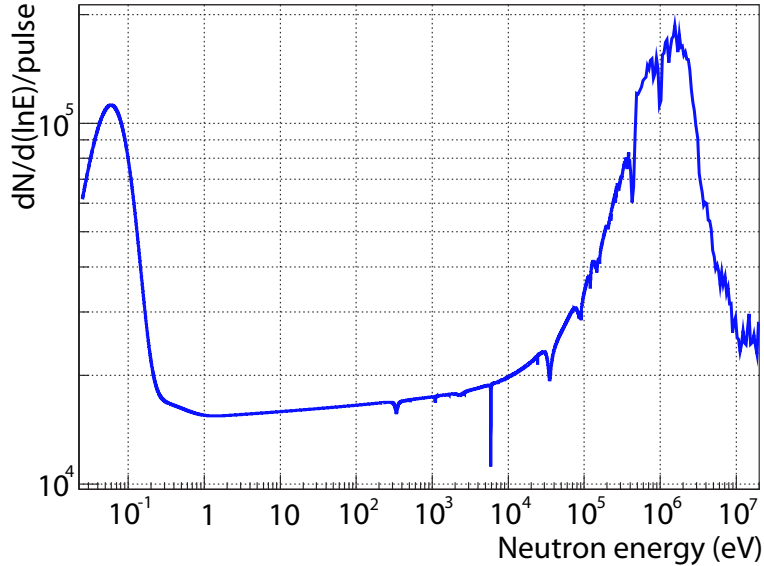


Figure 3.3: *Neutron fluence at the measuring station evaluated from the PTB measurements.*

Figure 3.3 shows the n_TOF neutron intensity per pulse in isolethargic units resulting from the measurement with the PTB fission chamber. Three different energy regions can be identified concerning the shape of the neutron fluence. The high energy region ($\sim \text{MeV}$) shows the typical neutron evaporation spectrum, the intermediate energy region (1 eV to 10 keV) corresponds to partially moderated neutrons and follows an isolethargic distribution, and the low energy region (< 0.1 eV) corresponds to completely thermalized neutrons. Table 3.1 summarizes the number of neutrons per decade between 1 eV and 100 MeV.

The neutron fluence per pulse of the n_TOF facility is the largest among the time-of-flight facilities around the world. This feature is of great advantage for the measurement of radioactive

Neutron energy	Neutrons/pulse
< 1 eV	$3.1 \cdot 10^5$
1 - 10 eV	$4.5 \cdot 10^4$
10 - 100 eV	$4.7 \cdot 10^4$
100 - 1000 eV	$5.1 \cdot 10^4$
1 eV - 1 keV	$1.4 \cdot 10^5$
1 - 10 keV	$5.4 \cdot 10^4$
10 - 100 keV	$7.1 \cdot 10^4$
100 - 1000 keV	$2.3 \cdot 10^5$
1 keV - 1 MeV	$3.5 \cdot 10^5$
1 - 10 MeV	$2.4 \cdot 10^5$
10 - 100 MeV	$7.2 \cdot 10^4$
1 - 100 MeV	$3.1 \cdot 10^5$
Total (1 - 10^8 eV)	$8.0 \cdot 10^5$

Table 3.1: *Number of neutrons per pulse at different neutron energy ranges.*

samples because it minimizes the background to noise ratio. Table 2.1 summarizes some of the main characteristics of the n_TOF facility and other time-of-flight facilities.

3.1.2 Neutron beam profile

Two collimators are installed along the beam line (see Figure 3.2) to shape the neutron beam at the sample position. The first one, referred to as *source screening collimator*, consists of 1 m of iron and 1 m of concrete with an inner diameter of 11.5 cm and is located at 136.7 m. The second collimator, referred to as *beam shaping collimator*, is located at 178 m and consists of 50 cm of 5% borated polyethylene, 125 cm of iron and 75 cm of 5% borated polyethylene with an inner diameter of 1.8 cm.

The resulting spatial distribution of the neutrons in the beam was measured with a pixelized Micromegas detector [82]. The measurements were performed with the strip electrodes of the Micromegas rotated at different angles (0° , 30° and 90°) in order to determine the two dimensional beam profile, which is shown in Figure 3.4 in the energy range between 10 eV and 100 eV. The neutron beam was found to have an approximate Gaussian shape with a width of $\sigma=0.77$ cm, which varies slowly with neutron energy. The detailed beam profile and an analytical function for its description are given in Ref. [82].

In addition to the measurement with the Micromegas, a complete Monte Carlo simulation of the production and moderation of the neutrons and their transport through the beam line was performed using the intranuclear cascade code FLUKA [83] and the Energy Amplifier Monte Carlo code EAMC [84]. The results from the simulation were validated by comparison with the measurements [82]. Since the sample position may vary within 0.5 mm (see Section 6.2.3.3)

this simulation is used to estimate the uncertainty in the number of neutrons intersecting the samples in different measurements.

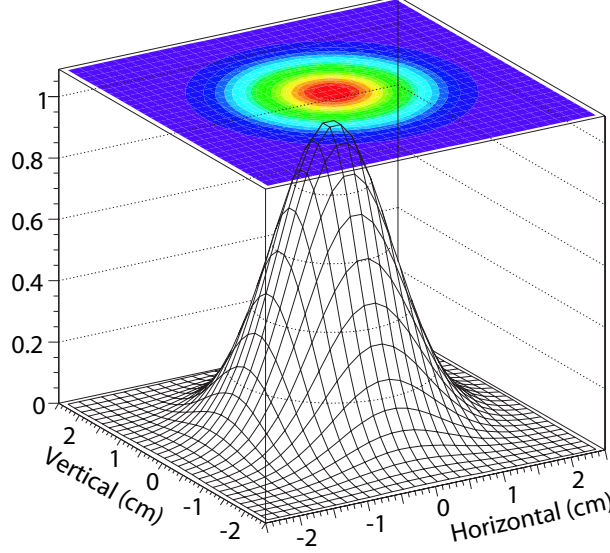


Figure 3.4: *Spatial profile of the n_TOF neutron beam at the measuring station.*

3.1.3 Resolution Function

The neutrons of a given energy reach the sample in the experimental area at different times due to the different equivalent distance that each neutron travels inside the lead block and the moderator before entering the beam line. The result is that the relation between the neutron energy and the time-of-flight is not univocal, but instead is given by a distribution that is known as the Resolution Function (RF).

The n_TOF RF has been calculated by a combination of simulations and measurements. The simulations were done [85] combining the codes CAMOT [86], FLUKA [83] and MCNPX [87]). The results of the simulations were validated by measuring narrow p - and d -wave capture resonances on a ^{nat}Fe sample [88, 89] that are very sensitive to the shape of the RF.

The RF is better understood in terms of a spread in the equivalent distance travelled by each neutron from its production to the sample position. Figure 3.5 shows the RF of the n_TOF facility expressed in equivalent distance as a function of neutron energy, where it is observed that the spread in the equivalent distance increases with neutron energy. The n_TOF RF is described analytically by a chi-square distribution with six degrees of freedom plus two exponential terms [90] that has been implemented in the SAMMY code for the analysis of resonances.

The Resolution Function introduces a broadening in the shape of the resonances because

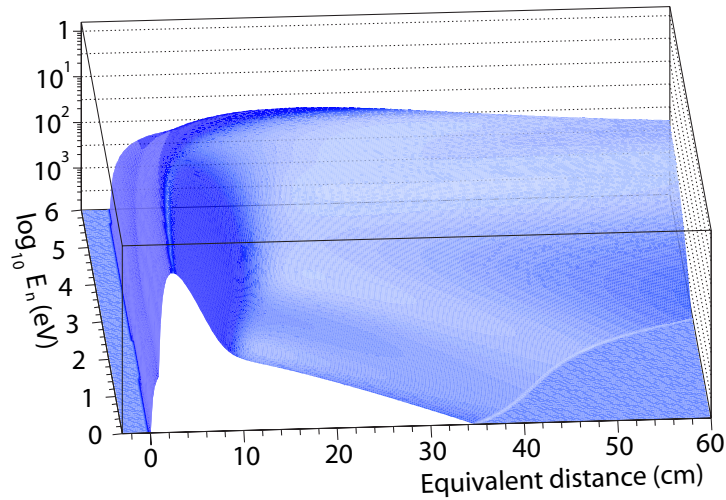


Figure 3.5: *Resolution Function of the n_TOF facility, expressed as the distribution of the equivalent distance travelled by each neutron as a function of its energy.*

they are measured as a function of the time-of-flight. There is an additional broadening effect caused by thermal motion of the nuclei in the sample that is known as the Doppler broadening. The relative magnitude of these two effects depends on neutron energy and is illustrated in Figure 3.6, where the Doppler broadening is determined using the Free Gas Model for a nucleus with mass $A=232$. The effect of the Resolution Function is negligible at low neutron energies ($E_n < 100$ eV) but dominates above a few keV.

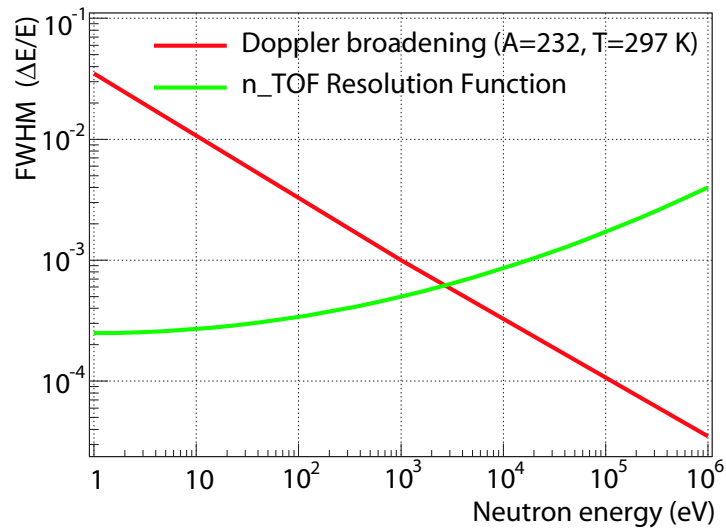


Figure 3.6: *Resonance broadening caused by the Resolution Function and the Doppler effect as a function of neutron energy.*

3.1.4 Neutron beam monitors

The intensity of the neutron beam is monitored constantly during the measurements. The continuous monitoring provides the means to perform relative measurements with respect to a reference sample, which is important for the characterization of the different types of background (see Section 4.2.2) or for normalization purposes (see Section 5.2.2).

The intensity of the neutron beam is monitored at the entrance of the experimental area using the Silicon Flux Monitor *SiMon*. The SiMon, which is shown in the left panel in Figure 3.7, was designed to interfere with the neutron beam as little as possible and consists of a thin Mylar foil with a ${}^6\text{LiF}$ deposit surrounded by four silicon detectors outside the neutron beam. The monitoring is based on the detection of the products of the ${}^6\text{Li}(n,\alpha){}^3\text{H}$ reaction, which has a large, smooth and well known cross section. A detailed description of the *SiMon* system and its performance can be found in Ref. [91].

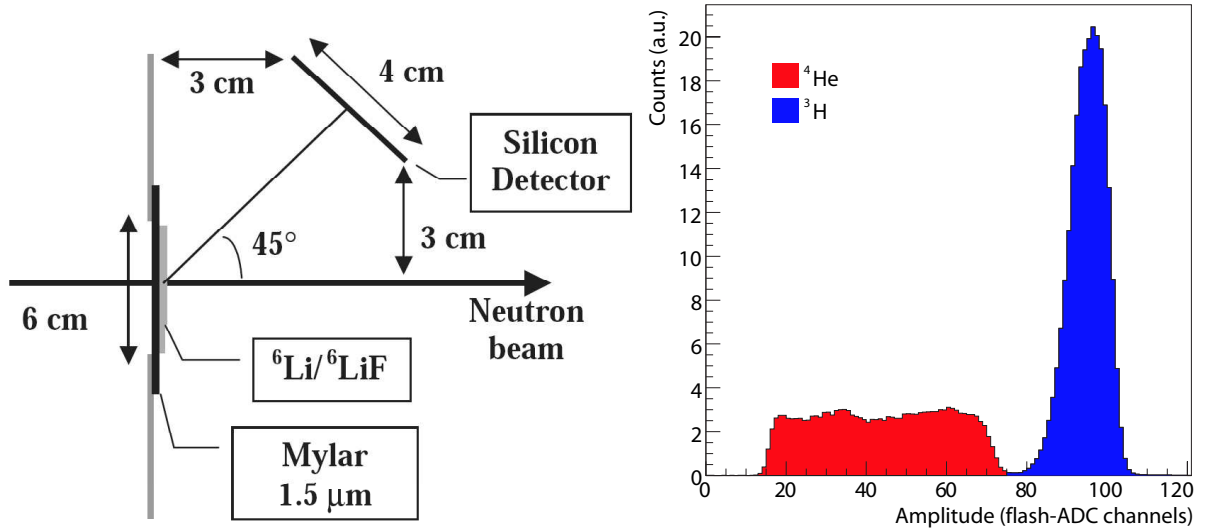


Figure 3.7: Schematic view of the *SiMon* set-up (left) and the corresponding pulse height spectrum (right), where the alpha and triton peaks are observed at low and high amplitudes, respectively.

The right panel in Figure 3.7 shows the pulse height spectrum of one of the silicon detectors in which the structures corresponding to alpha and triton particles are observed at low and high amplitudes, respectively. The lower mass of the tritons allow them to escape completely from the lithium deposit, which is not the case for the alpha particles. For this reason, only the triton peak was used to determine the intensity of the neutron beam.

3.2 Neutron capture experimental set-up

3.2.1 The n_TOF Total Absorption Calorimeter

The n_TOF Total Absorption Calorimeter (TAC) is an improved version of the BaF₂ calorimeter at FZK [69] and has been designed in order to fulfil all the requirements of an ideal total absorption detector: large solid angle coverage, high γ -ray total absorption efficiency, good energy resolution, high segmentation, low neutron sensitivity, and fast time response. A picture of the assembly is shown in Figure 3.8.

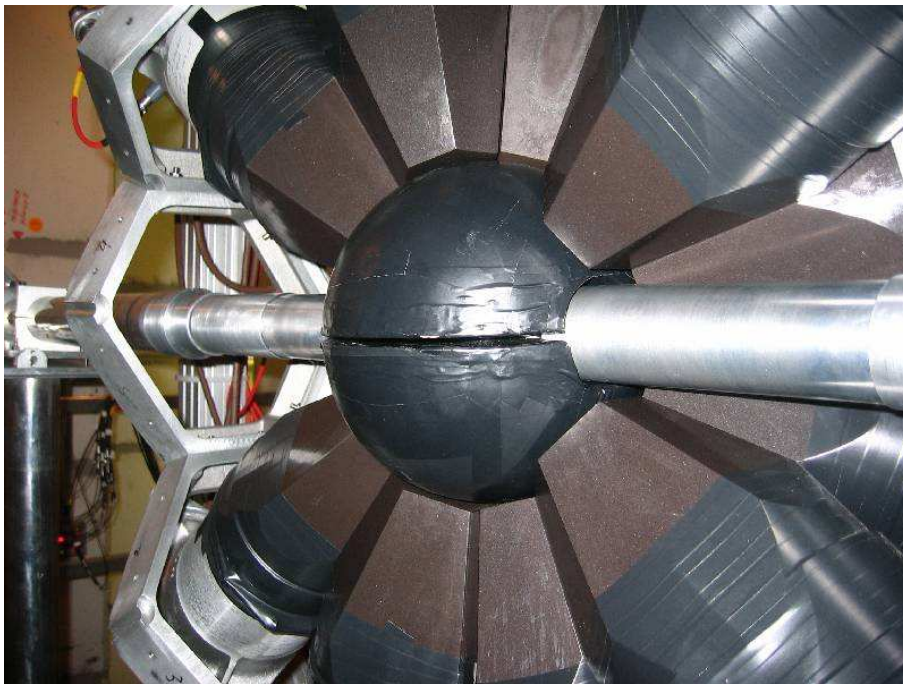


Figure 3.8: *Picture of the n_TOF TAC assembly. The structure can be opened in two parts to replace the samples and the neutron absorber.*

The TAC consists of 40 BaF₂ crystals, 12 pentagonal and 28 hexagonal, that form a sphere with an inner radius of 10.5 cm covering a large solid angle (95% of 4π) with respect to the sample position at the centre of the TAC. Two holes in opposite positions are left in order to allow the entrance and exit of the beam line through the detector. Both types of crystals, pentagonal and hexagonal, are shaped from BaF₂ cylinders 14 cm in diameter and 15 cm in thickness, with raw and final weights of 12 kg and 7.5 kg, respectively. The surface of the crystals is polished and the reflector consists of two layers of Teflon foil 0.1 mm in thickness followed by 0.1 mm of polished aluminium.

The scintillation light of BaF₂ has two components, both emitted in the UV range. The fast

component with a characteristic time $\tau_{fast}=0.6$ ns is emitted at 220 nm, while the slow component with $\tau_{slow}=620$ ns is emitted at 315 nm. The fast scintillation component is responsible for the good timing properties of the BaF₂ detectors, while the slow component is the cause of pile-up events that appear at high counting rates.

The 12 cm diameter Photonis XP4508 photomultiplier was chosen due to its resolution and spectral range. The plateau region of the cathode is found between 200 nm and 550 nm, and is peaked at 420 nm. The PMs are operated at a voltage between 1200 and 1600 V with voltage dividers that were especially designed at ITN-Lisbon for minimizing the recovery time of the PM after saturation.

The BaF₂ modules (crystal, PM and voltage divider) are attached to an aluminium honey-comb structure that holds the 40 modules forming a hollow sphere of inner and outer diameters of 10.5 cm and 25.5 cm, respectively. The aluminium structure, shown in Figure 3.9, can be opened into two hemispheres for accessing the inside of the TAC and placing the samples.



Figure 3.9: *Picture of the aluminium honey-comb structure that holds the complete assembly.*

One of the very useful features of the TAC is that its high detection and total absorption efficiencies combined with its good energy resolution allow the discrimination between the reactions of interest and the different sources of background from the Q -value of each type of reactions. Such a capability is further improved based on the large segmentation (40 crystals) that allows the identification of different reactions by the number of emitted γ -rays.

3.2.2 The neutron sensitivity

One of the main sources of background that appear in the measurement of neutron capture reactions is related to the detection of neutrons scattered by the sample or any other material along the beam line. The probability of detecting those scattered neutrons is usually referred to as the neutron sensitivity of the detector. In the case of the TAC set-up, this has been minimized by the combination of a neutron moderator/absorber surrounding the sample and the use of crystal capsules with a high content in ^{10}B . The idea behind this design is that neutrons scattered at the sample position are moderated before reaching the crystals and then absorbed either inside the actual moderator or in the capsules of the crystals by means of $^{10}\text{B}(n,\alpha)^7\text{Li}$ reactions.

Monte Carlo simulations performed with GEANT3 [92, 93] and GEANT4 [94, 95, 73] have confirmed that the best material for the neutron moderator/absorber is ^6LiH , which in addition has the advantage of a very low γ -ray attenuation due to its low effective atomic number Z . However, the safety rules at CERN exclude the use of ^6LiH due to its high flammability and toxicity. Therefore, $\text{C}_{12}\text{H}_{20}\text{O}_4(^6\text{Li})_2$, an inert and inflammable compound, was used instead. For mechanical stability, the compound was encapsulated in a spherical 0.5 mm thick aluminium shell with inner and outer radii of 5 and 10 cm, respectively, which is placed inside the TAC as shown in Figure 3.8.

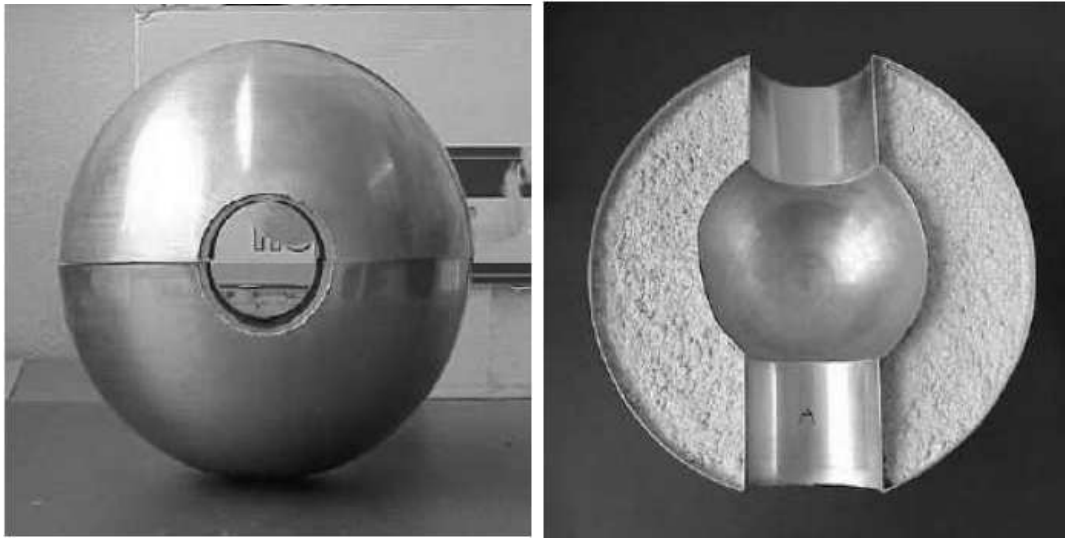


Figure 3.10: *Left: Aluminium capsule of the neutron absorber filled with the ^6Li salt. Right: Complete neutron absorber assembly.*

The crystal capsules were manufactured at Legnaro (INFN) and consist of a carbon fibre resin enriched with ^{10}B (70 g of ^{10}B and 77 g of resin in each capsule). The simulations with GEANT4 carried out during the design stage [95] predicted a factor of three in the reduction of the neutron sensitivity on the TAC set-up at neutron energies of the order of 1 keV.

3.2.3 The samples

The measurement of the neutron capture cross section of a given isotope implies not only the measurement of the sample of interest but also a set of auxiliary measurements that are needed for energy calibration, normalization, background characterization, etc. In this section the two types of samples are described.

The actinide samples were manufactured at IPPE Obninsk [96] in the form of oxide powder deposited in a disk shaped aluminium backing 1 cm in diameter and then sandwiched between 0.15 mm thick aluminum foils. The actinide samples are encapsulated in a 0.2 mm thick titanium canning so that the whole assembly satisfies the *ISO-2919 norm* required by the CERN's radioprotection services. For easy handling, the samples are mounted between two Kapton foils of 25 μm in a carbon fibre ring. A schematic view of the sample assembly is shown in Figure 3.11. The main characteristics of the two actinide samples measured in this work are summarized in Table 3.2.

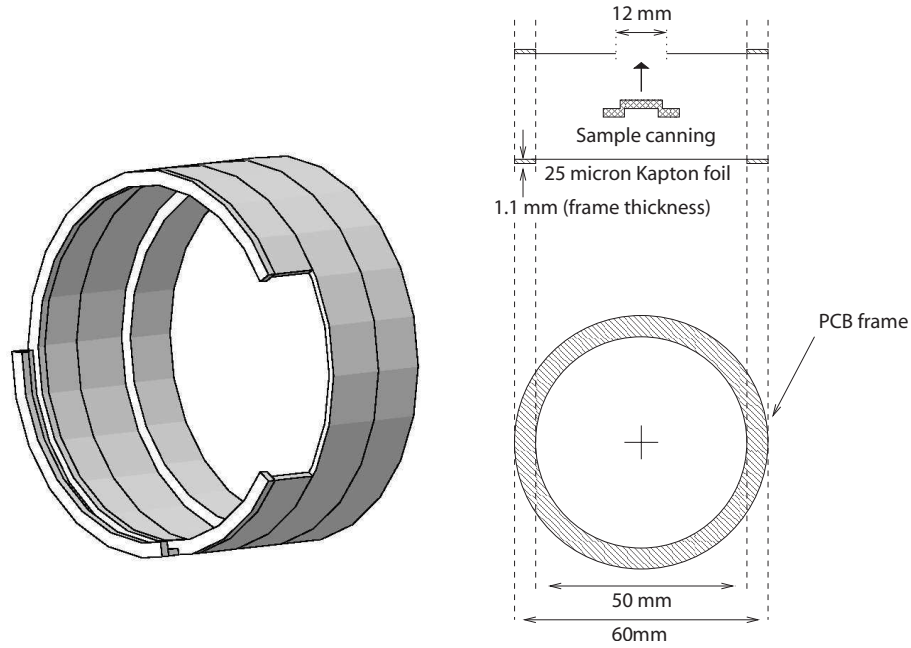


Figure 3.11: *Left: geometry of the sample holder placed at the centre of the TAC. Right: drawings of the sample assembly including the titanium canning and the Kapton frame.*

Several disk shaped samples, also with 1 cm diameter, were used in auxiliary measurements: (i) a ^{197}Au sample of 185.4 mg was used as a reference and for the validation of the experimental technique, (ii) a *dummy-sample* was used for characterizing the effect of the titanium capsule and the aluminium backings, (iii) a graphite sample of 70 mg was used for characterizing the detector response to scattered neutrons, and (iv) a dummy Kapton frame, referred to as sample-out, was used for the characterization of the TAC response due to the presence of the beam.

	^{237}Np	^{240}Pu
Total mass (mg)	49.1	58.0
Isotopic mass (mg)	43.3	51.2
Isotopic purity	99.2%	88%
Impurity	^{233}Pa (0.8%)	^{239}Pu (11.1%)
Impurity	-	Others (0.9%)
Activity (MBq)	1.3	422
Half life (years)	2.14×10^6	6538
Thickness (at/barn)	1.401×10^{-4}	1.636×10^{-4}

Table 3.2: *Main characteristics of the actinide samples*

3.3 Data acquisition system

One advantage of n_TOF with respect to other facilities is the use of a fully digital Data Acquisition System, which is presented in detail in Ref. [97]. The advantage of such a system is that it allows the analysis offline of the recorded pulses by dedicated *pulse shape analysis* (PSA) algorithms optimized for each detector type.

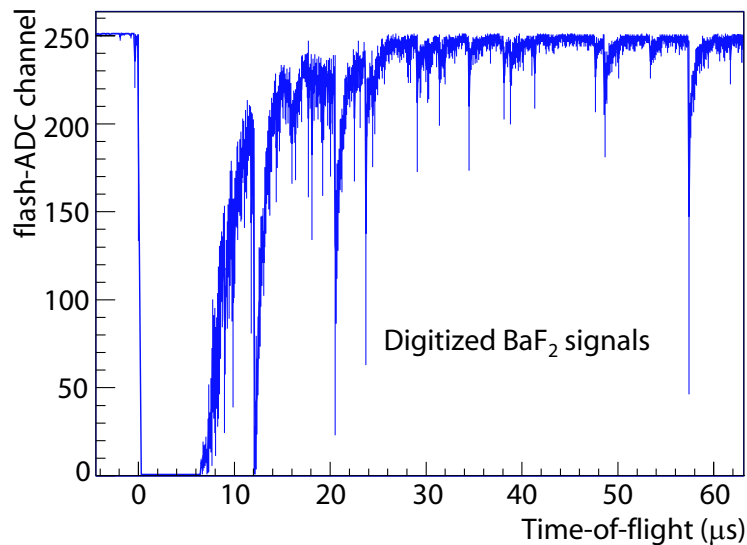


Figure 3.12: *Example of the saturation of a BaF_2 detector by the γ -flash.*

The read-out of the BaF_2 modules is carried out by a total of forty channels of high performance digitizers, Acqiris-DC270 [98], with 8 bit resolution and 8 MByte memory working at 500 MSamples/s. This system corresponds to 16 ms long data buffers containing digitized signals of the BaF_2 modules for neutron energies between 0.3 eV and 20 GeV. A reduction in the sampling rate down to 100 MSamples/s allows measuring neutron cross sections in the $1/v$ energy region

down to 0.028 eV.

A fast zero suppression algorithm selects only pieces of data containing true detector signals with amplitudes above a threshold for each flash-ADC, reducing in this way the amount of data to be stored. The raw flash-ADC data files are sent via Gigabit Ethernet to a temporary disk pool located close to the measuring station. Once the files are closed, they are transferred via Gigabit Ethernet to a second disk pool from which they are finally migrated to tape. Such an acquisition system requires large storage capabilities which at n_TOF are provided by the *CERN Advanced STORage manager* (*CASTOR*) [99].

The first particles arriving at the experimental area are γ -rays and relativistic charged particles produced during the spallation process. The intensity of these particles, referred to as the γ -flash, is so high that the detectors saturate and remain *blind* during a certain period of time. This is shown in the example of Figure 3.12 [100]. The time needed for the recovery of each detector determines the upper limit of the neutron energy range that can be analyzed, which in the case of the BaF₂ crystals is a few hundred keV.

3.3.1 Pulse Shape Analysis

The relevant parameters of each recorded BaF₂ signal are the type of incident particle that produced it, its time-of-flight, and its amplitude, from which the corresponding signal energy is calculated. A dedicated Pulse Shape Analysis routine (PSA) is used to extract all these parameters which, after the appropriate formatting, are saved as Data Summary Tapes (DSTs) and stored on tapes and disk for fast access.

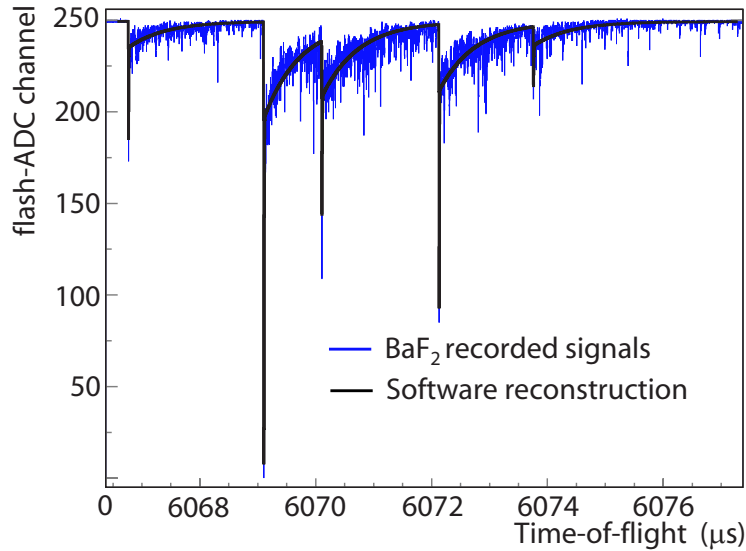


Figure 3.13: *Digitized BaF₂ signals (grey) and the corresponding reconstruction after the pulse shape analysis (black).*

The desirable features of the PSA are:

- Identification of the BaF₂ signals in the recorded data buffers, which show large fluctuations related with the statistics of the scintillation process and the fast sampling (500 MSamples/s).
- Accurate determination of the time and amplitude of the signal.
- Identification and analysis of the pile-up events that appear at high counting rates due to the existence of the slow scintillation component of BaF₂.
- Discrimination between the signals produced by γ -rays and those produced by the α activity of the crystals, in particular from the decay of the ²²⁶Ra and its progeny existing in the crystals.

The PSA implemented in the Data Acquisition Systems uses a sophisticated algorithm based in a two-component fit: a Maxwellian function for fitting the fast component and an exponential function for fitting the slow component. Full details are given in Ref. [101]. Figure 3.13 shows an example of the fit to a series of pulses from the ¹⁹⁷Au(n, γ) measurement at 4.9 eV, where the black line is a fit of the recorded pulses (shown in grey). The individual signals are well identified above the fluctuations even in the case of pile-up events.

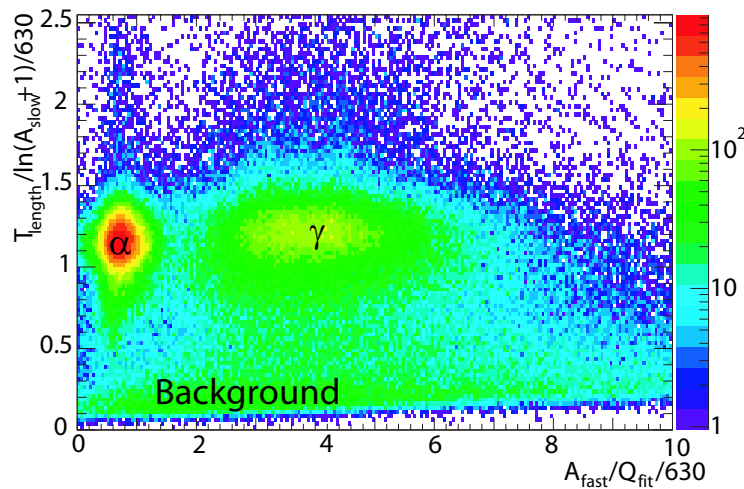


Figure 3.14: Scatter plot where the coordinate and ordinate axes are related to the contribution of the fast and slow components to the total light output of many pulses, allowing the discrimination between the two types of particles.

The discrimination between γ -rays and α -particles is achieved based on the direct relationship that exists between the type of ionizing particle and the relative contribution of the slow and fast scintillation component to the total light output [102]. In particular, the signals produced by α particles show a significantly lower fast component than those produced by γ -rays. The

degree of discrimination between the two types of ionizing particles is illustrated in Figure 3.14, which shows a scatter plot with the coordinate and ordinate axis related to the contribution of the fast and slow components to the total light output. The background caused by spikes in the digitizers is observed at the bottom of the figure, corresponding to a negligible contribution of the slow component.

Chapter 4

Performance of the TAC

An important fraction of this work has been dedicated to characterize the performance of the Total Absorption Calorimeter (TAC).

This chapter describes the measurements for the determination of the energy and time resolution of the detector, the characterization of the different sources of background, the investigation of the detector response to neutron capture and scattering events, and the type of analysis conditions that optimize the signal to background ratio in neutron capture measurements.

4.1 The response of the TAC to standard γ -ray sources

4.1.1 Energy calibration

During the capture measurements all modules were calibrated at least once per week. A parabolic calibration was necessary in order to take the non linearity of some of the modules into account. This is shown in Figure 4.2 where the coloured curves correspond to different polynomial fits.

The variation of the gain of the detectors was monitored daily from the position of the peaks of the α lines produced in the decay of the ^{226}Ra and its progeny, present in the BaF_2 crystals. The right panel in Figure 4.2 shows the different α lines observed in a single crystal. The most energetic α line, marked in red line and corresponding to ^{214}Po with $Q_\alpha=7.8$ MeV, was used for the gain shift correction of the energy calibration by assuming that the variation of its position determines the linear variation of the calibration coefficients.

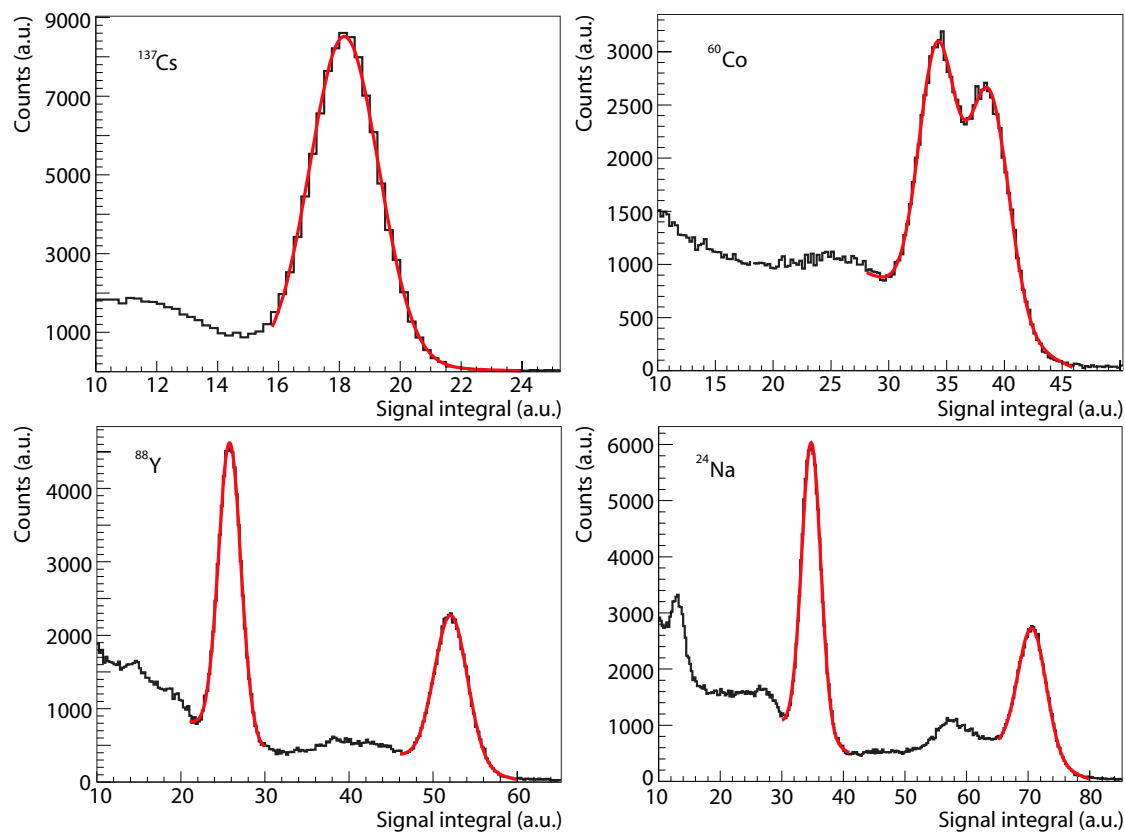


Figure 4.1: Single crystal (#11) signal integral spectrum of the γ -ray sources used at n-TOF: ^{137}Cs , ^{60}Co , ^{88}Y and ^{24}Na

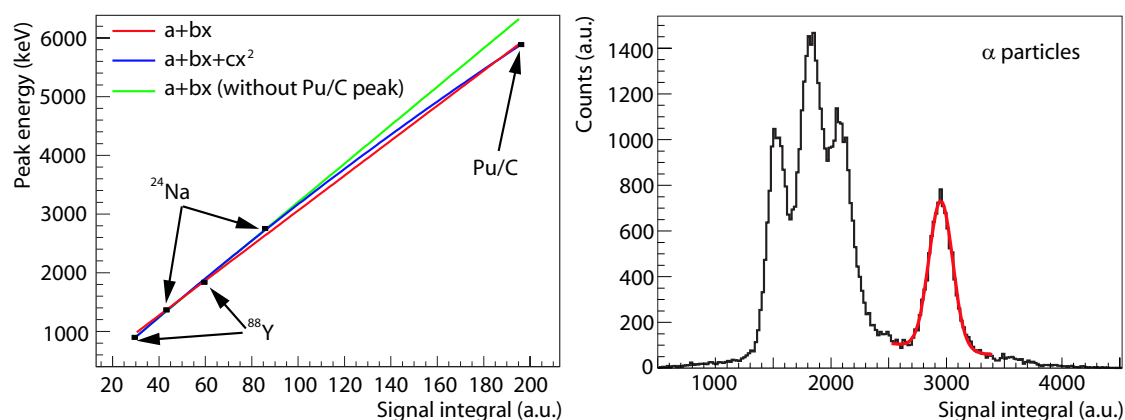


Figure 4.2: Energy calibration of an individual crystal using the ^{88}Y , ^{24}Na and Pu/C sources. The coloured curves correspond to different fitting functions with and without the Pu/C peak.

4.1.2 Event reconstruction

The signals recorded in the individual crystals are added up by requiring a time coincidence. In this way, the complete list of signals from the individual modules is reduced to a list of physical events characterized by a crystal multiplicity m_{cr} , calculated as the number of crystals involved in the detection of the event, a time-of-flight TOF , corresponding to the first signal within the event, and a total energy E_{sum} , calculated as the sum of the energy of all the signals within the event.

It was necessary to synchronize all flash-ADC modules for the coincidence analysis because the precision of their internal clocks have an accuracy of only 2 ppm [98]. Such an uncertainty in the clocks results in time deviations between detectors of the order of a few tens of ns. The time response of each flash-ADC channel was calibrated with an external *pulse generator*.

The optimal width of the coincidence window was found at $\Delta T=11$ ns and set to this value. Figure 4.3 illustrates how the number of counts in the total absorption peak of a ^{60}Co source remains stable for widths larger than 11 ns and becomes smaller for shorter time windows.

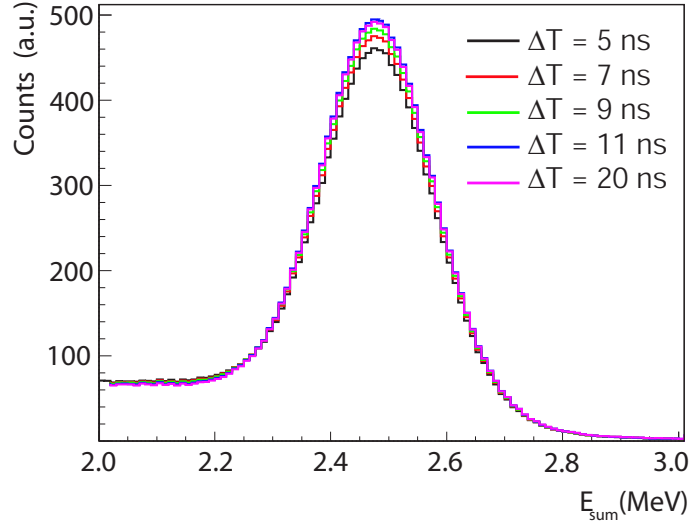


Figure 4.3: *Sum peak of the ^{60}Co source for different values of the time coincidence window.*

4.1.3 Energy resolution

The energy response of each individual BaF_2 module has been determined for the γ -rays in the decays of ^{137}Cs , ^{60}Co , ^{88}Y and ^{24}Na . Figure 4.4 shows the energy resolution of each module at the 662 keV line of ^{137}Cs . The values are spread widely around a mean energy resolution of 14.3%.

The energy resolution of the TAC operating in coincidence is displayed in Figure 4.4. The

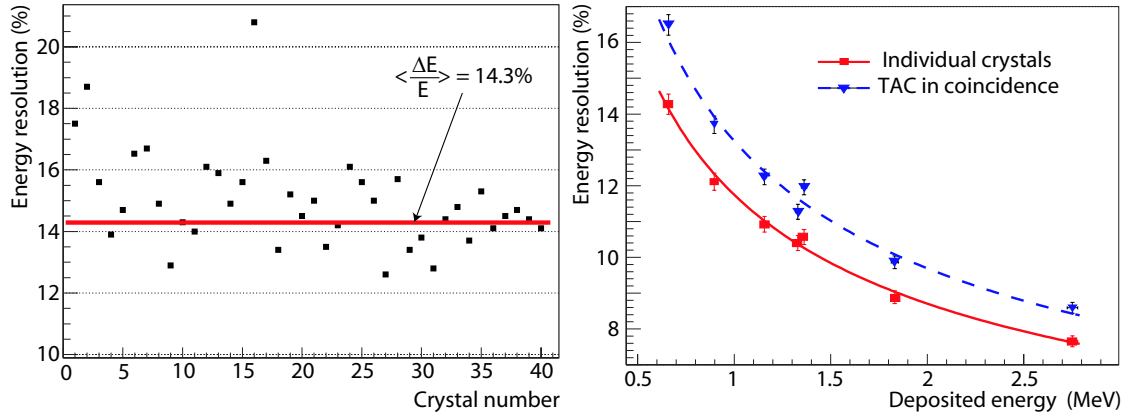


Figure 4.4: Energy resolution of the individual crystals at the energy of the ^{137}Cs peak: 662 keV.

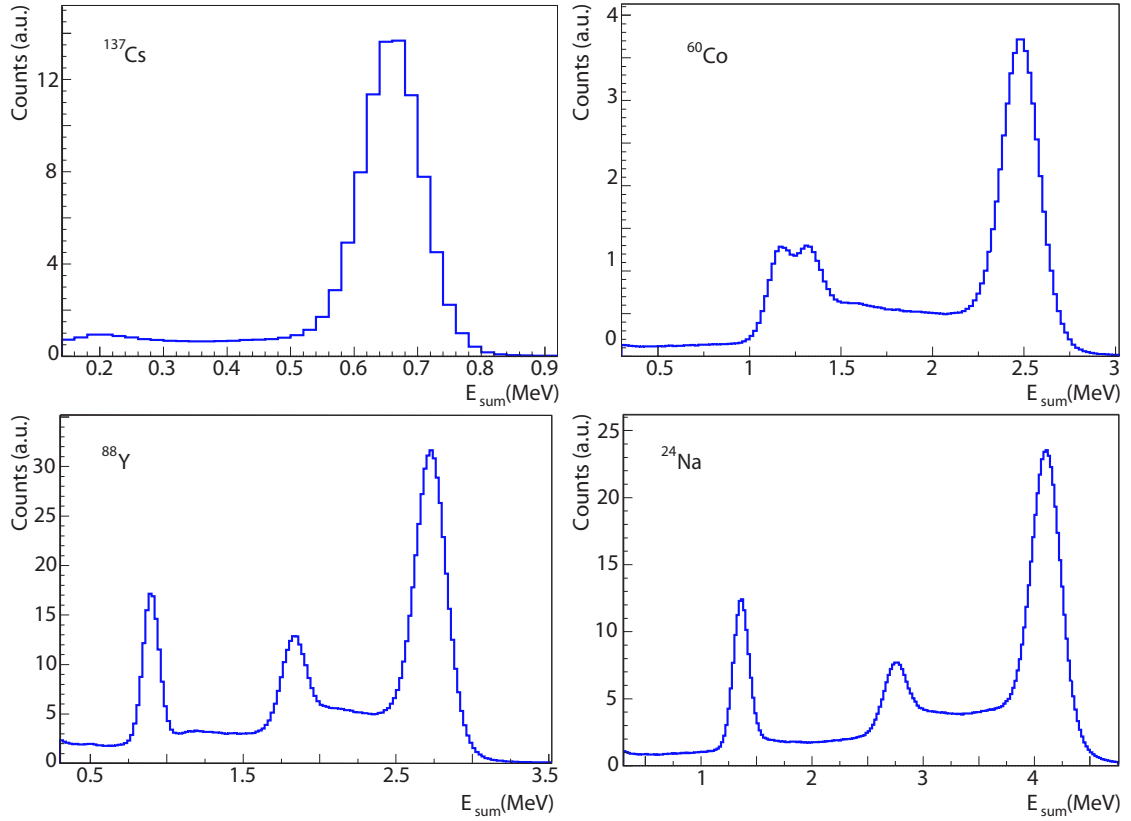


Figure 4.5: TAC response to standard γ -ray calibration sources.

squares correspond to the measurement of the available calibration sources and the solid line corresponds to a fit of the expected behaviour. The energy resolution of the complete TAC is larger than that of the individual crystals, see Figure 4.4, due to the spread in resolution of the individual crystals and to the uncertainty in the energy calibration.

Last, the complete response of the TAC to the β -decays of ^{137}Cs , ^{60}Co , ^{88}Y and ^{24}Na is shown in Figure 4.5. As expected for a high efficiency calorimeter, the photopeaks corresponding to single γ -rays are much weaker than the total absorption peak corresponding to the detection in coincidence of the two γ -rays in the case of ^{60}Co , ^{88}Y and ^{24}Na .

4.1.4 Signal pile-up and dead-time

BaF_2 is an excellent scintillator for timing applications due to its fast scintillation component $\tau_{fast} = 0.6$ ns. However, this scintillation component contributes only 20% to the total light emission, and the rest is emitted with a much slower decay time $\tau_{slow} = 620$ ns. The slow decay time results in a non-negligible probability of pile-up events which might be not possible to identify during the Pulse Shape Analysis [101]. Hence, even though a digital acquisition system eliminates the dead-time due to the acquisition system itself, some signals are missed and a dead-time appears when measuring at high counting rates.

The dead-time losses of the TAC have been characterized from the deviations of the measured and theoretical distributions of time intervals between consecutive signals detected in a given crystal. The expected distribution is given by [103]:

$$I(t)dt = re^{-rt}dt. \quad (4.1)$$

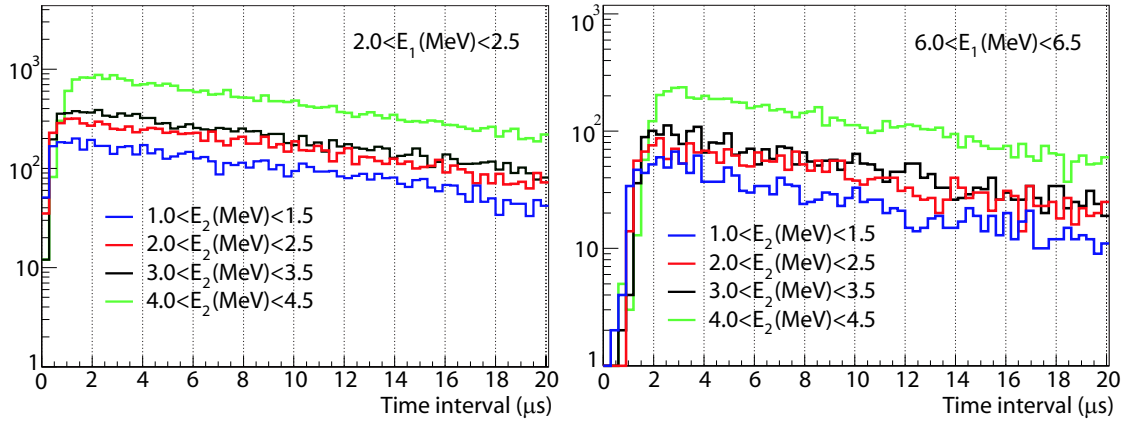


Figure 4.6: Time interval distributions for several E_2 ranges for $2 < E_1(\text{MeV}) < 2.5$ (left) and $6 < E_1(\text{MeV}) < 6.5$ (right).

It was found that this exponential behaviour is lost for time intervals below a certain limit identified as the dead-time. Furthermore, a careful investigation revealed that there is a dependency of the dead-time on the amplitudes of the pile-up signals. This is illustrated in Figure 4.6 which shows the time interval distribution for some specific energy ranges of the first E_1 and the second E_2 signals in the pile-up. For a fixed E_1 between 2 MeV and 2.5 MeV, the dead-time

is variable but always lower than $2 \mu s$, while for the case of E_1 between 6 MeV and 6.5 MeV it is always larger than $2 \mu s$.

The left panel in Figure 4.7 shows the dead-time values for all combinations of energies E_1 and E_2 . The dead-times range up to $3 \mu s$ and have an average value of $1 \mu s$. The dead-time increases directly proportional to E_1 and inversely proportional to E_2 . This behaviour indicates that it is more difficult to identify a small signal in the tail of a large one than in the opposite case. These two situations are illustrated in the right panel in Figure 4.7 for ideal exponential signals with $\tau=620$ ns and a time separation of $1 \mu s$.

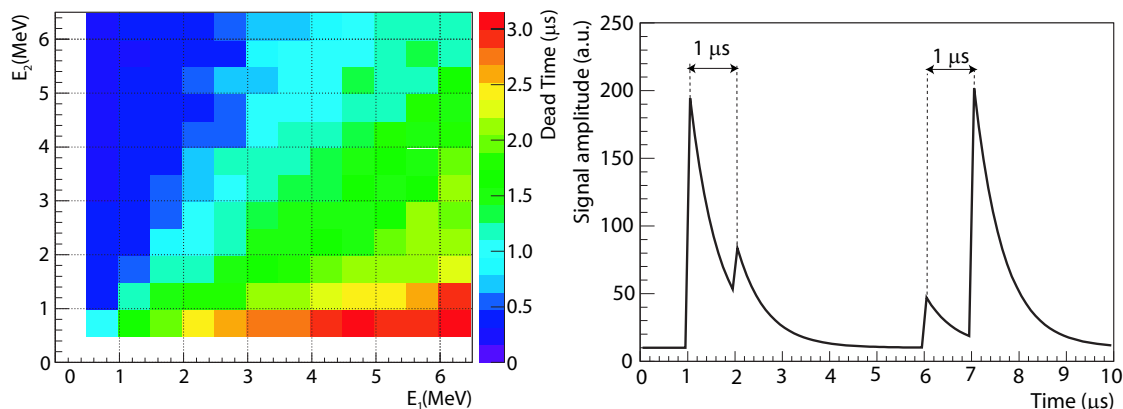


Figure 4.7: *Dead time as a function of the energy of the first (E_1) and second (E_2) signals.*

The dead-time has important consequences because the signal losses become significant for counting rates of the order of $1 \text{ count}/\mu$. For instance, in the reference measurement of ^{197}Au there are many resonances with even higher counting rates, as it is shown in the two examples in Figure 4.8 corresponding to different intervals of the counting rate spectrum as a function of the time-of-flight. Concerning the effect of the dead-time losses, two situations can be distinguished:

- The first, illustrated in the left panel, corresponds to long time-of-flight intervals in which the time bins are wide compared to the dead-time ($\sim \mu s$) and therefore the dead-time losses caused by signals in a given bin do not affect the counting rate in the neighbouring bins.
- The second, illustrated in the right panel, corresponds to short time-of-flight in which the time bins is comparable to the dead-time and hence the counting rate in a given bin is responsible for dead-time losses in the bins at later times.

These two situations must be treated properly, as it is done in this work by the use of a dead-time correction model based on Monte Carlo simulations that is presented in detail in Section 5.1.2.

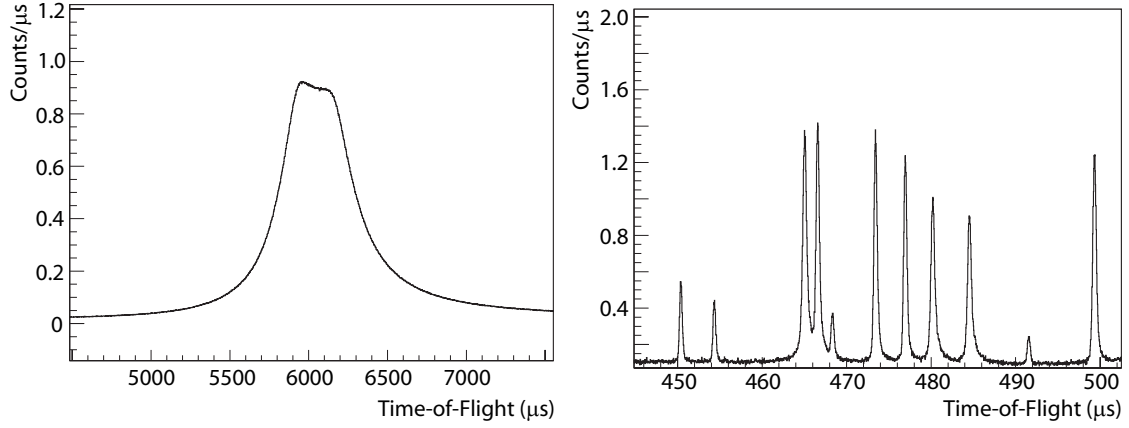


Figure 4.8: Counting rate as a function of the time-of-flight calculated for the $^{197}\text{Au}(n,\gamma)$ measurement using the ENDF/B-VII evaluated cross section.

4.2 The response of the TAC to real (n, γ) measurements

4.2.1 Stability of the TAC and the neutron beam monitor

Since the neutron intensity of all the measurements has been calculated from counts in the neutron beam monitor *SiMon* compared to the counts in the *SiMon* for the ^{197}Au reference measurement, the proportionality between counting rates of the TAC and the *SiMon* has been verified for every proton pulse delivered to the spallation target.

Figure 4.9 shows the ratio of counts registered in the TAC with respect to the *SiMon* as function of the Run number for the three main measurements of the experimental campaign. A Run contains a large number of proton pulses (~ 2000) delivered sequentially in a given time interval, usually no more than two hours. The TAC and *SiMon* counting rates correspond only to events in the neutron energy range between 1 eV and 10 keV. The figure shows also the ratio of counting rates of the TAC and the *SiMon* with respect to the intensity of the proton beam provided by the PS.

It can be concluded that the proportionality between TAC and *SiMon* is better than 0.5% for all the measurements and that the ratio between the counting rate of the TAC and the proton intensity remains constant within 0.7%. These results prove also that all the detectors were stable along the measurements and that the *SiMon* system is a very accurate neutron beam monitor for normalization purposes.

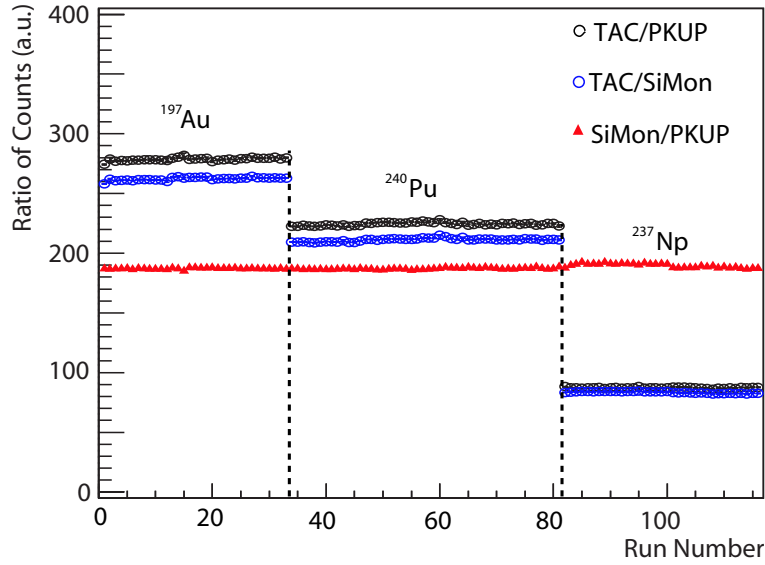


Figure 4.9: Proportionality along the ^{197}Au , ^{237}Np and ^{240}Pu measurement between the counting rates of the different detection systems: TAC, SiMon and WCM.

4.2.2 Characterization of the background

There are different sources of background that appear in (n,γ) measurement with the TAC, each producing a characteristic response in terms of energy and multiplicity. They have been classified by their time structure with respect to the neutron beam:

- a. The backgrounds that are not correlated with the neutron beam are mainly caused by the γ activity of the radioactive samples with energies up to a few hundred keV, the decay chain of the ^{226}Ra contamination in the BaF_2 crystals with α and γ energies up to a few MeV [69] and the room background in the experimental area, mainly caused by the decay of ^{40}K . These types of background have been characterized by dedicated beam-off measurements.
- b. The backgrounds that are directly produced by the beam are summarized as follows:
 1. *In-beam γ -rays* of 2.2 MeV from $^1\text{H}(n,\gamma)$ reactions produced in the moderator surrounding the spallation target and scattered along the beam line. Details of its distribution in time-of-flight are given in Ref. [27].
 2. *γ -ray cascades* from (n,γ) reactions taking place in any of the materials exposed to the neutron beam, mainly the vacuum windows (aluminium and Kapton) and the sample assembly (aluminium and titanium).
 3. *Neutrons scattered along the beam line* captured in the neutron absorber, the crystal capsules, or the Ba and F isotopes of the crystals.

The beam related backgrounds have been characterized in dedicated measurements: (i)

Sample-out for determining the background not related to the sample, (ii) *Dummy-sample* for determining the background due to capture and scattering reactions in the sample assembly (Ti, Al, Kapton), and (iii) *Graphite sample* for the investigation of the detector response to scattered neutrons.

4.2.2.1 The reference ^{197}Au sample

The different background contributions of a 185.4 mg ^{197}Au sample are illustrated in Figures 4.10 and 4.11 together with those of the sample-out and beam-off measurements. All distributions are normalized to the neutron beam intensity given by the neutron monitor *SiMon*.

Figure 4.10 shows the distributions of the energy deposited in the TAC (E_{sum}) in two neutron energy intervals: 1 to 10 eV (left) and 1 to 10 keV (right). The main difference between the two spectra is the relative magnitude of the overall background with respect to the capture reactions in ^{197}Au that are observed up to ~ 7.0 MeV, just above the neutron separation energy of the compound nucleus $S_n(^{198}\text{Au}) = 6.54$ MeV.

Three components can be distinguished in the spectra of the beam-off measurement corresponding to the decay of Ra contaminant and its progeny with Q values of 2.2 MeV, 3.2 MeV and 5 MeV [69]. On the other hand, the 1.46 MeV peak corresponds to the decay of ^{40}K present in the concrete walls.

The sample-out spectrum shows a peak at 478 keV from the decay of $^7\text{Li}^*$ following $^{10}\text{B}(n, \alpha)$ reactions in the crystal capsules, a small structure at 2.2 MeV from $^1\text{H}(n, \gamma)$ reactions in the neutron absorber, and a high energy contribution from scattered neutrons that are captured in $^{nat}\text{Ba}(n, \gamma)$ reactions and produce γ -ray cascades with energies from 4.7 to 9.1 MeV.

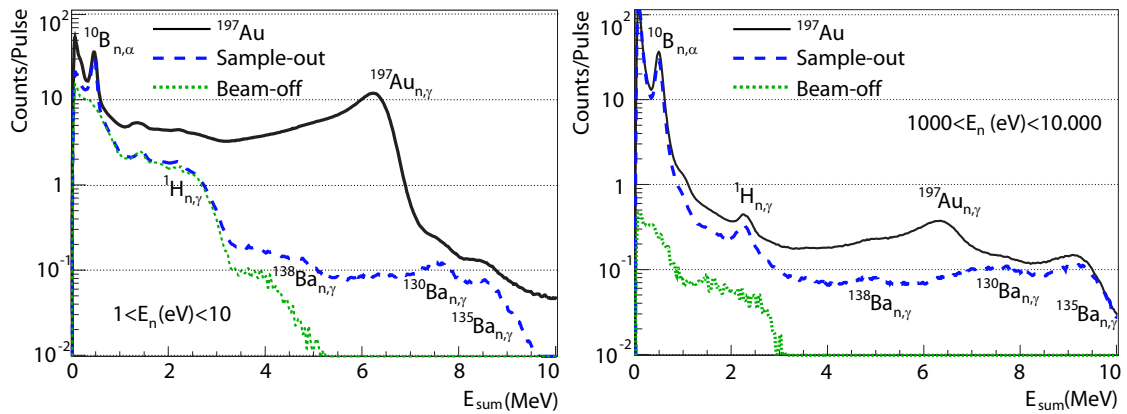


Figure 4.10: Deposited energy distributions recorded during the ^{197}Au and the dedicated background measurements in two different neutron energy intervals: 1 to 10 eV (left) and 1 to 10 keV (right).

The counting rate recorded as a function of neutron energy in the ^{197}Au sample and the dedicated background measurement is displayed in Figure 4.11. Only the events with energies between 2.5 MeV and 7 MeV, where the capture to background ratio is more favourable, were selected.

The resonant structures of $^{197}\text{Au}(n,\gamma)$ are observed well above the overall background up to several keV. The dominant background is caused by the incident beam in the form of neutron induced reactions outside the sample that are detected in the TAC. Up to a few keV, this background is only relevant in the regions between resonances. The activity of the different materials in the experimental area contributes to the overall background only at low neutron energies and is, at least, one order of magnitude below the capture signal in the complete energy range.

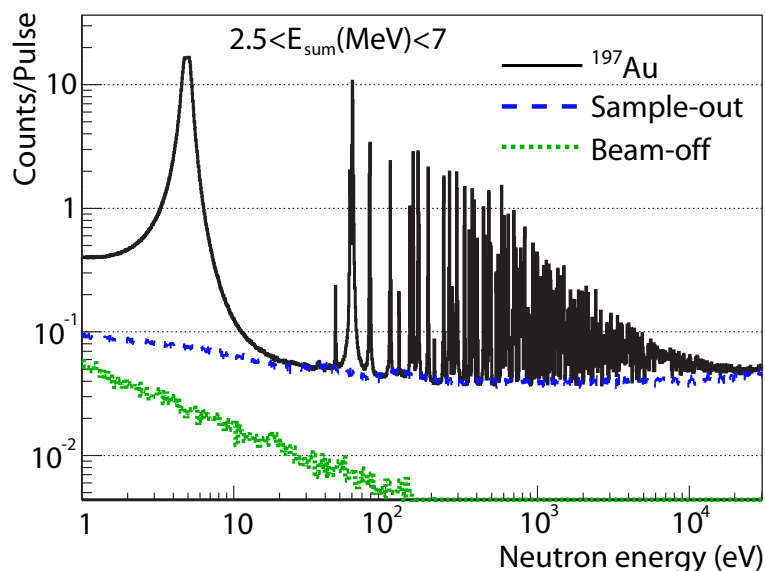


Figure 4.11: Measured counting rate distributions corresponding to the ^{197}Au and the dedicated background measurements for deposited energies between 2.5 and 7 MeV, where the capture to background ratio is more favourable.

4.2.2.2 The radioactive and encapsulated samples

The main difficulties associated to the measurement of radioactive samples are:

1. The samples have small masses because of the low availability of these isotopes but also in order to reduce the activity of the samples. This implies that the room and beam induced background may dominate the measurement.
2. The activity of the samples induces a very intense low energy background. This is not an

issue when using the total absorption technique because it is possible to select a minimum threshold in deposited energy just above the energy of the activity, usually a few hundred keV.

3. At n-TOF, the radioactive samples need to satisfy the ISO-2019 norm and therefore are encapsulated in a massive aluminium and titanium assembly. This assembly interacts with the neutron beam and produces a background that can even dominate the contribution from capture reactions in the sample.

All these features are illustrated in Figures 4.12 and 4.13 which correspond to the measurements of a 43.3 mg ^{237}Np sample. In the first figure the contribution associated to capture reactions in the ^{237}Np sample is observed up to 6 MeV, just above the neutron separation energy of the compound nucleus $S_n(^{238}\text{Np}) = 5.49\text{ MeV}$. The activity of the sample is observed only at low deposited energies, while the background produced in the sample assembly is observed at all deposited energies. The latter is characterized by two total absorption peaks at 6.4 MeV and 8.1 MeV that correspond to $^{50}\text{Ti}(n,\gamma)$ and $^{48}\text{Ti}(n,\gamma)$ reactions.

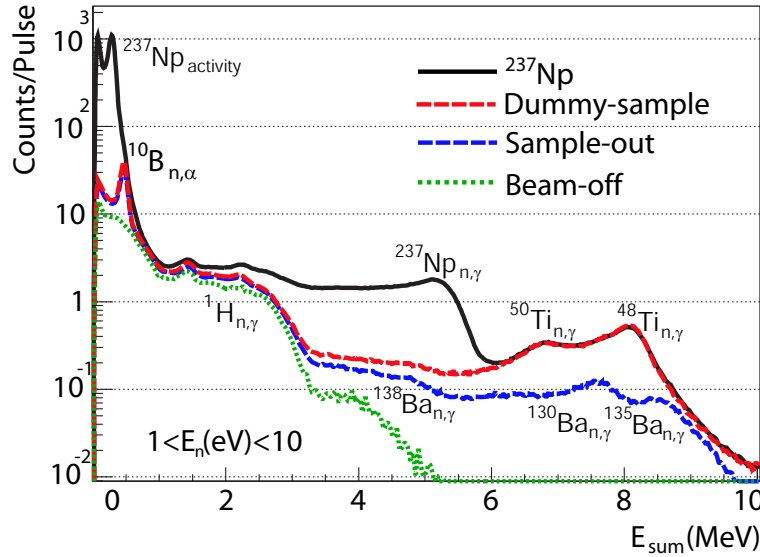


Figure 4.12: Measured deposited energy distributions corresponding to the ^{237}Np and the dedicated background measurements in the neutron energy interval between 1 eV and 10 eV.

The magnitude of the different background contributions with respect to the capture reactions in the sample can be observed in Figure 4.13. Only the events with energies between 2.5 MeV and 7 MeV, where the capture to background ratio is more favourable, were selected. The background due to the sample assembly dominates above a few keV, setting the high energy limit of the analysis. However, below 2 keV, the capture to background ratio is highly favourable, even for a radioactive sample with a very small mass. Such a high capture to background ratio, which is mainly due to the use of the total absorption technique and the selection of events in a reduced range of deposited energies, provides a very reliable counting rate.

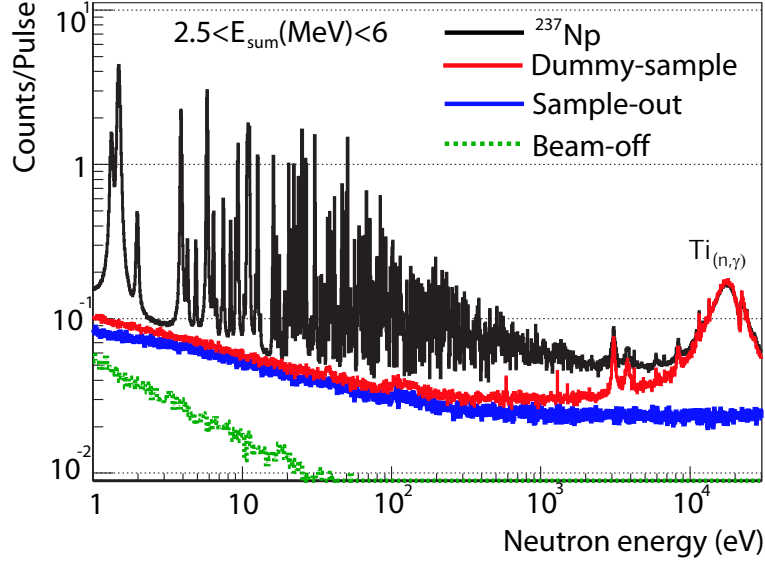


Figure 4.13: Measured counting rate distributions corresponding to the ^{237}Np and the dedicated background measurements for deposited energies between 2.5 and 6 MeV, where the capture to background ratio is more favourable.

4.2.3 Event selection conditions on E_{sum} and m_{cr}

The use of a segmented TAC with high total absorption efficiency allows one to reduce the experimental background by selecting events that satisfy some specific conditions in terms of crystal multiplicity m_{cr} and deposited energy E_{sum} .

The analysis of the data from ^{197}Au and ^{237}Np samples (figures 4.10 and 4.12) already illustrated that the capture events are mainly distributed in a restricted interval of deposited energies between a few MeV and the neutron separation energy of the compound nucleus. Furthermore, the background contributions have different distributions in terms of crystal multiplicity. For instance, the activity of the sample or of the 478 keV line from $^{10}\text{B}(n,\alpha)$ reactions produce mainly events with $m_{cr}=1$. For this reason, selecting events with a restricted value of m_{cr} improves the capture to background ratio.

This is illustrated in Figure 4.14 where the deposited energy spectrum of ^{197}Au is displayed for different conditions in m_{cr} . The condition $m_{cr} > 2$ eliminates the major part of the background without reducing the number of capture events significantly.

The optimum constraints in E_{sum} and m_{cr} must be chosen according to the characteristics of each specific measurements, i.e. sample activity, capture to scattering ratio, and energy and multiplicity of the capture cascade.

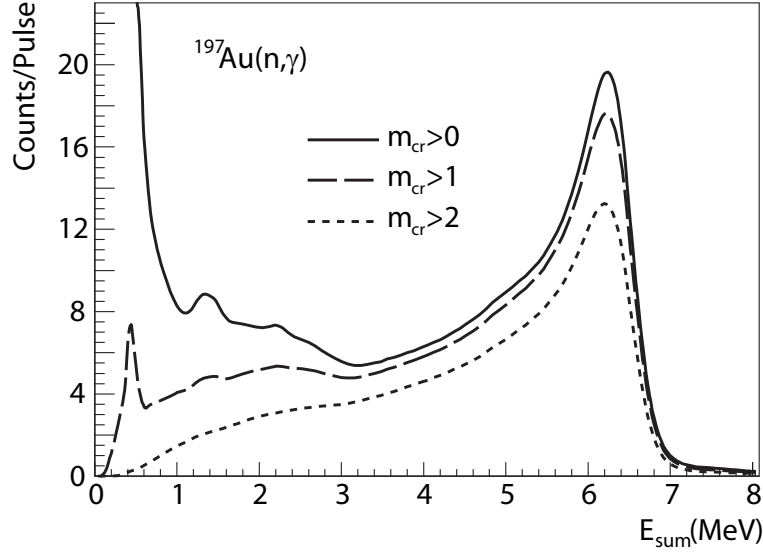


Figure 4.14: Deposited energy distribution of the $^{197}\text{Au}(n,\gamma)$ measurement under different constraints in crystal multiplicity. The data corresponds to neutron energies from 1 to 10 eV.

4.2.4 The neutron sensitivity of the TAC

The neutron sensitivity $\varepsilon_{n,n}$ of a detection system is defined as its efficiency for detecting scattered neutrons. Low values of $\varepsilon_{n,n}$ are necessary in neutron capture measurements in order to keep the background related to scattered neutrons as low as possible. Such a background is of special concern when the neutrons follow a resonant scattering similar to the capture reactions under investigation, inducing systematic errors in the determination of the resonance parameters, as it is demonstrated in Refs. [68, 104, 105].

In general, the detection systems intended for neutron capture measurements are designed in such a way that their $\varepsilon_{n,n}$ is minimized [65, 95] by using materials with low capture cross sections and by placing a neutron shielding between the sample and the detector. In the case of the TAC, such a reduction has been achieved by using a neutron moderator/absorber placed in the inner hole of the TAC with a large content of ^1H and ^6Li . In addition, the BaF_2 crystals were enclosed in ^{10}B enriched capsules in order to absorb the neutrons escaping the moderator/absorber.

The background associated to neutrons scattered by the sample can be accounted for by, at least, three different methods:

1. Analytically, from the theoretical probability of neutron scattering in the sample and taking into account the kinematics of the scattering reaction, the additional flight path between sample and detector, and the neutron sensitivity as function of neutron energy, which is usually calculated by Monte Carlo simulations. Such an analytical approach is implemented in the REFIT code [75].

2. A similar calculation was proposed by Allen et al. [106] using Monte Carlo techniques instead of an analytical approach for the description of the neutron scattering reactions and the subsequent detection of neutrons.
3. The total absorption technique makes it possible to measure the background due to scattered neutrons as a function of neutron energy from the comparison of the deposited energy distribution under study with that of a pure scatterer, such as a graphite sample ($\sigma_n > 10^4 \sigma_\gamma$) [107].

We have used the third method in order to rely only on experimental data and not on less accurate neutron transport simulation codes and multiple scattering models. Hence, the neutron scattering background in a capture measurement is determined by comparison of the spectra measured with the sample under investigation with those of a graphite sample.

The determination of the neutron scattering background is illustrated in Figure 4.15. The left panel shows the TAC response to the measurement of a 70 mg graphite sample for neutron energies between 1 keV and 100 keV, where the different structures correspond to neutron capture reactions in the different isotopes in the detector assembly. The right panel shows the ^{197}Au energy spectra under the condition $m_{cr} > 2$ as a solid line, and two shaded areas corresponding to the pure (n, γ) and (n,n) distributions, the latter corresponding to the graphite spectrum scaled to reproduce the high deposited energy region ($E_{sum} > 8.5$ MeV) of the ^{197}Au spectrum. The method is applied in the complete neutron energy interval bin to bin in such a way that the resonant (n,n) background is determined point-to-point as a function of the neutron energy.

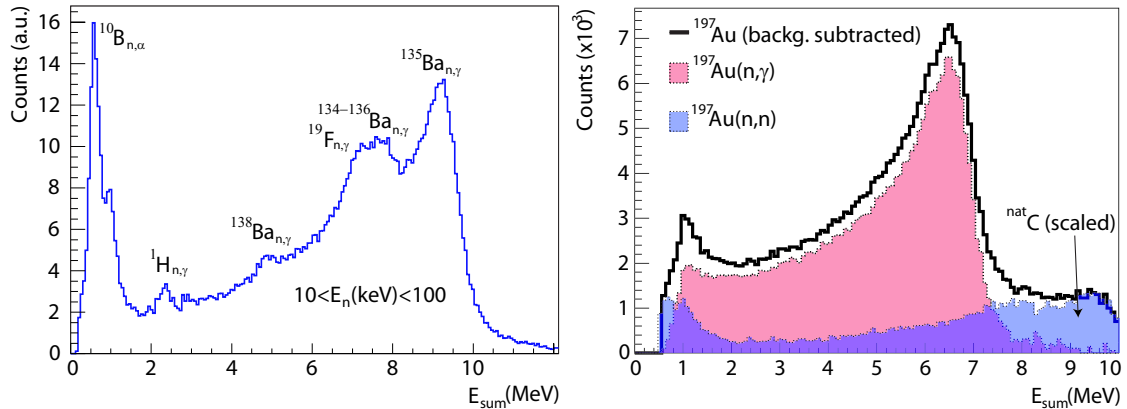


Figure 4.15: Deposited energy distribution recorded during the measurement of a 70 mg graphite sample for neutron energies from 1 to 100 keV.

The measurement of the graphite sample is also used to calculate the neutron sensitivity of the detector as a function of neutron energy by comparing the measured and theoretical (n,n) reaction yields. In the case of neutron scattering, the theoretical and observed yields are given

by

$$Y_{n,n}^{th}(E_n) = (1 - e^{-n\sigma_t(E_n)}) \frac{\sigma_n(E_n)}{\sigma_t(E_n)} \quad (4.2)$$

and

$$Y_{n,n}^{obs} = \frac{C - B}{\Phi_n}, \quad (4.3)$$

where the total (σ_t) and scattering (σ_n) neutron cross sections can be retrieved from any evaluated cross section data base.

The neutron sensitivity $\varepsilon_{n,n}$ is then given as the ratio between the theoretical and observed yields. The values obtained with the cross sections of the JEFF-3.1 evaluation [108] are displayed as a function of neutron energy in Figure 4.16. It was found that $\varepsilon_{n,n}$ varies from 0.2% in the low neutron energy region up to 1% above ~100 eV. The sudden increase of $\varepsilon_{n,n}$ above 30 eV is related to resonances in the different Ba and F capture cross sections, which are also responsible for the structures around 40 eV and 100 eV.

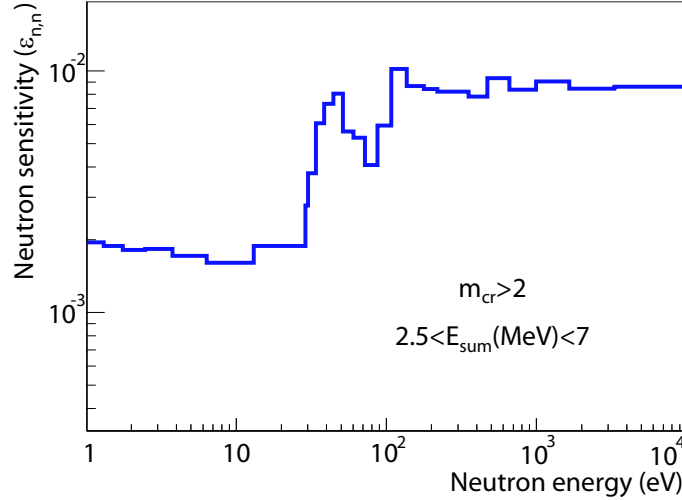


Figure 4.16: Neutron sensitivity of the TAC set-up measured with a 70 mg graphite sample using the JEFF-3.1. evaluated cross section.

Since the neutron sensitivity is approximately 100 times lower than the efficiency of the TAC for detecting capture cascades, the background due to scattered neutrons in the sample under study becomes significant ($>1\%$) only for those resonances with $\Gamma_n \geq \Gamma_\gamma$.

Chapter 5

Calculation of the experimental capture yield

The observable quantity in neutron capture cross section measurements is the capture yield, which is defined as the fraction of neutrons impinging on the sample that undergo a capture reaction (see Section 2.2.3). Experimentally, the capture yield $Y_{n,\gamma}(E_n)$ is obtained as a function of neutron energy E_n from the total $C(E_n)$ and background $B(E_n)$ counting rates, the efficiency of the detector ε , the intensity of incident neutrons recorded in the neutron monitor $\phi_n(E_n)$, and the normalization factor N that relates the neutron intensity in the monitor with that of the neutrons impinging in the sample:

$$Y_{n,\gamma} = \frac{C - B}{\varepsilon \cdot N \cdot \phi_n}, \quad (5.1)$$

where all but N are neutron energy dependent quantities.

The detection of capture events, as well as the contribution of the different types of background and the intensity of the neutron beam have been already discussed in Section 4.2. In this chapter, the determination of the detection efficiency is described, followed by the validation for the $^{197}\text{Au}(n,\gamma)$ reference measurement.

5.1 Determination of the detection efficiency

The detection efficiency of the TAC depends on the investigated isotope, on the conditions selected for crystal multiplicity and deposited energy selected, and on the counting rate. An accurate calculation of the detection efficiency taking into account all the mentioned dependencies can be achieved best by means of Monte Carlo (MC) simulations. The idea is to reproduce the measured distributions for all combinations of deposited energy and crystal multiplicity in order to achieve a realistic and accurate determination of the detection efficiency. A reliable

simulation code should include the geometry of the detector, all the physical processes that may occur in the experiment and a realistic event generator.

5.1.1 Monte Carlo simulation of the TAC

The simulation tool GEANT4 [94] with the Standard Electromagnetic Package [109] has been chosen for simulating the TAC. It allows one to implement complex geometries, its physics models have been widely validated, and provides the powerful tracking capabilities needed for the event reconstruction.

The TAC geometry [110] has been modelled from the CAD drawings of the engineering design with high fidelity, as it can be appreciated in Figure 5.1. The simulated geometry includes the 40 BaF₂ crystals with their corresponding ¹⁰B enriched carbon fibre capsules, the photomultiplier tubes with their aluminium housings, the aluminium beam pipes with the sample holder, the ⁶Li enriched neutron moderator/absorber and the aluminium honey-comb structure that holds the complete assembly.

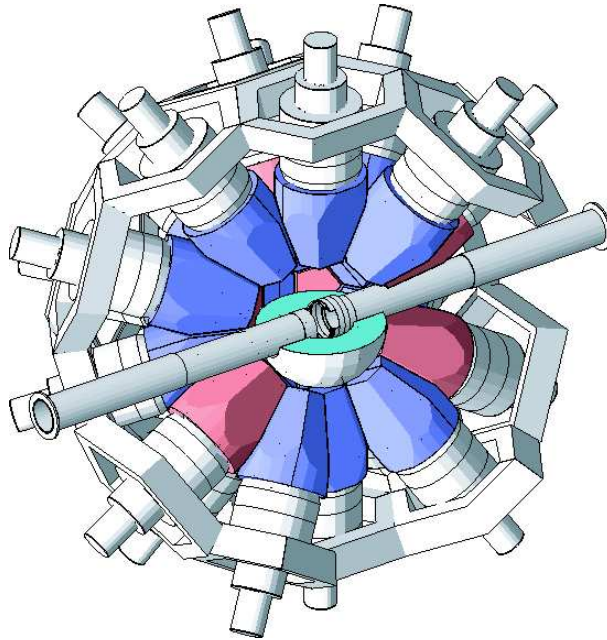


Figure 5.1: *View of half of the Total Absorption Calorimeter as it is implemented in the code GEANT4 used for the Monte Carlo simulations.*

The simulation begins with the generation of a number of *initial events* at the centre of the TAC. Each *initial event* is generated at a specific time-of-flight and contains the complete information of the EM cascade following the neutron capture reaction: energy and emission angle of each γ -ray and/or conversion electron of the cascade. The MC code simulates the transport of all particles through the detector until they are completely absorbed or transported

outside the TAC. When an interaction takes place in the BaF₂ crystals, the deposited energy, time-of-flight and crystal identification number of the interaction are recorded for the subsequent event reconstruction.

The event reconstruction algorithm of the simulated data is similar to that used in the analysis of the experimental data, see Section 4.1.2. The energy deposited inside a crystal, within a time interval of 20 ns with respect to the first interaction in that crystal, is considered to correspond to one single *hit*. An event is then defined as a set of *hits* with deposited energy above the selected threshold (100 keV) and occurring inside a coincidence window of 20 ns started by the first interaction in any of the crystals. The energy resolution of the TAC is added by performing a convolution of the deposited energy of each event with a Gaussian distribution with $\Delta E/E = a + b/\sqrt{E}$, where a and b are obtained from the fit to experimental data.

In this way, one obtains a list of events, which are characterized by their crystal multiplicity (m_{cr}), energy deposited in each crystal (E_{cr}^i with $i=1, m_{cr}$), total deposited energy (E_{sum}), time-of-flight (TOF) and the associated neutron energy (E_n).

5.1.2 Dead-time correction model

One of the innovative features of this work is the model for correcting dead-time losses [111, 112] (see Section 4.1.4) related to the slow component of the BaF₂ crystals. The existence of a dead-time in each crystal may produce the loss of one or more of the γ -rays of the capture cascade, affecting the m_{cr} and E_{sum} distributions and hence the detection efficiency. The most accurate method for calculating the dead-time losses is to include the measured dead-time $DT(E_1, E_2)$, which depends on the energy of the two signals involved in the pile-up signals E_1 and E_2 , in the event reconstruction of the simulated data. The experimental dead-time in the individual crystals $DT(E_1, E_2)$ is discussed in Section 4.1.4.

The loss of individual hits due to dead-time effects is calculated crystal by crystal prior to the event reconstruction of the simulated data. First, the *hits* registered in each crystal are ordered in increasing time-of-flight. Then the dead-time is calculated for each *hit* with respect to those registered at later times and, if $DT(E_1, E_2)$ is larger than the time difference between the *hits* ($TOF_1 - TOF_2$), the later hit is flagged as *masked by a previous signal* and is not included in the event reconstruction. In this way, the original set of simulated *hits* is transformed into a new set in exactly the same order that they occur in the measurements. Figure 5.2 illustrates a schematic situation, both in the measurement and the simulation, where only four out of seven consecutive signals are detected, while the rest are *masked by a previous signal* due to the dead-time in the crystal.

Since the TAC covers a large solid angle and has a high efficiency for detecting individual γ -rays, the total number of detected events is not strongly affected by the loss of a single *hit*. However, the characteristic E_{sum} and m_{cr} distributions may change significantly due to the loss of single *hits*, hence affecting the value of the detection efficiency when certain event selection

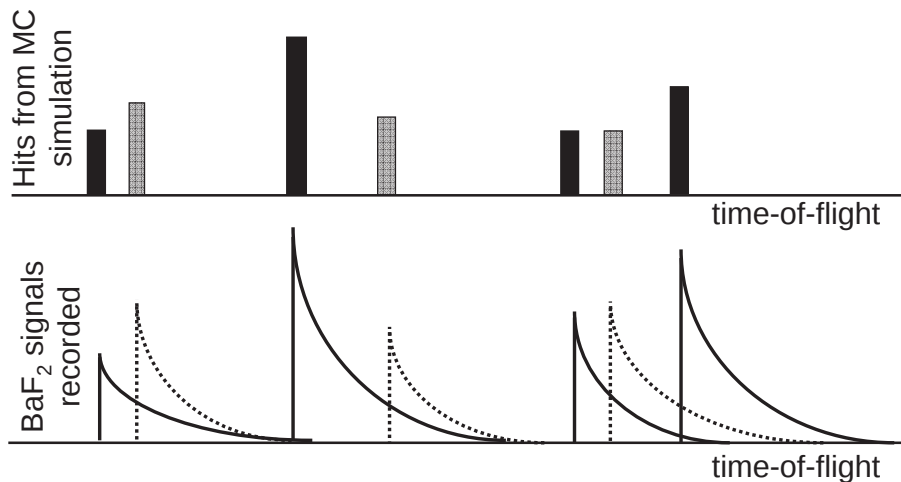


Figure 5.2: Schematic illustration of the dead-time losses in the simulation (top) and the experimental data (bottom). The signals in grey (top) and dashed (bottom) are masked by the previous signals and therefore not included in the event reconstruction.

conditions are applied on E_{sum} and m_{cr} .

5.1.3 β -decay event generator and validation with experimental data

Two types of event generators have been developed for simulating β -decay and neutron capture reactions, respectively. In this way, the Monte Carlo data have been compared to real measurements of γ -ray sources and (n,γ) reactions. This section is devoted to the β -decay event generator while the next is devoted to the neutron capture cascade generator.

The standard γ -ray sources that have been measured emit single (^{137}Cs) or two-step (^{24}Na , ^{60}Co and ^{88}Y) cascades. The aim of simulating the TAC response to these measurements was to validate the detector geometry and the physics models implemented in the simulation.

The exact β -decay scheme of all sources [113] was implemented in the event generator, thus including the information on the β feeding to the different levels of the daughter nucleus and the probability and multipolarity of the transitions between levels. In the case of two-step cascades the multipolarity of consecutive transitions was used for the calculation of the angular correlation between the emitted γ -rays. The time stamp of each β -decay event is calculated following the time interval distribution expected for constant counting rate processes:

$$I(t)dt = ne^{-rt}dt, \quad (5.2)$$

where r is the counting rate recorded during the measurements.

The geometry of the detector implemented in the simulation was fine tuned by comparison with experimental data:

- The distance of each crystal to the centre R_i varies slightly between the different crystals and an effective radius R_{TAC} had to be adjusted. A value of $R_{TAC}=10.67$ cm was found to provide an excellent simultaneous agreement between the simulations and the experimental data for all the β -decay measurements, as it is illustrated in Figure 5.3.
- A most important and uncertain parameter of the detector geometry is the density of the neutron absorber, which depends on the number of bubbles that appear during the solidification process of the ${}^6\text{Li}$ enriched salt [93]. The simulations with a value of $\rho=0.77$ g/cm³ were found to reproduce all β -decay measurements.

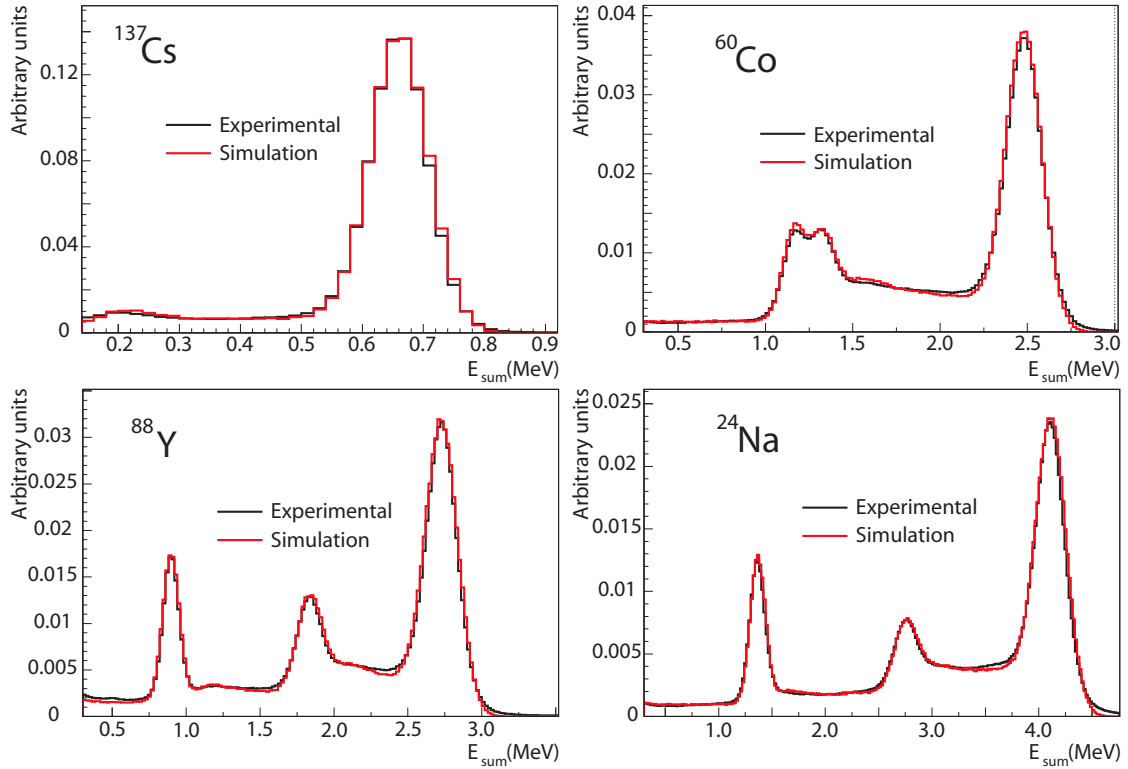


Figure 5.3: *Experimental (black) and simulated (red) TAC response to pure γ -ray sources.*

5.1.4 (n, γ) event generator and validation with experimental data

The generation of realistic capture cascades following neutron capture requires the complete nuclear level scheme below the neutron separation energy: energy, spin and parity of all levels and the transition probabilities between them. Such information is completely known only for a reduced number of isotopes, usually for the lighter nuclei. The general case is that the level scheme is known only up to certain excitation energy and the experimental information is scarce above. Hence, the generation of capture cascades has to be based on a model that combines the experimental information at low excitation energies, with theory based assumptions and statistical models at high excitation energies.

The capture events are generated following the experimental counting rate distribution as a function of time-of-flight. In this way, the dead-time losses can be accounted for during the event reconstruction of the simulated data, as discussed in Section 5.1.5.3.

5.1.4.1 The cascade generator

The model used in this work for the generation of realistic capture cascades was previously reported by Taín et al. [64] and applied specifically to the TAC measurements in Ref. [114]. The model considers two different excitation energy ranges:

- The lower energy range ($E < E_{cut}$) corresponds to the region of known levels (energy, spin and parity) with known transition probabilities. This information is retrieved from the Evaluated Nuclear Structure Data Files (ENSDF) [115] and allows one to calculate the low energy part of the branching ratio matrix. The electron conversion process is modelled for the K -, L - and M -shells from the tabulated values of fluorescence yields and internal conversion coefficients available in the literature.
- At higher energies ($E > E_{cut}$) the levels and transitions probabilities between them are calculated by means of a statistical model. The individual levels are created by sampling the level density distribution given by the Back Shifted Fermi Gas (BSFG) model [116], and the transition probabilities between levels are calculated only for E1, M1 and E2 transitions from the associated Photon Strength Functions (PSF) [117].

Starting from the capture level, the algorithm for generating a cascade includes the following features:

1. The branching ratio matrix for E1, M1 and E2 transitions is calculated from the known capture level to all levels below using the parameterized PSF.
2. The transition to a new level is sorted randomly according to the branching ratio matrix.
3. If the new level is in the statistical energy range, a new branching ratio matrix is computed. Otherwise the experimental branching ratio matrix is used.
4. The loop returns to step 2 until the ground (or a metastable) state is reached.

The correctness of the model parameters is validated by the comparison of the simulation and the experimental data. The experimental information for low energy ($E_\gamma < \sim 1$ MeV) and high energy transitions (> 10 MeV) comes from decay and photoabsorption experiments, respectively. In the case of intermediate energy transitions, governing the de-excitation of the compound nucleus from the capture state, the information is very scarce and only the Oslo method and the double step cascade method provide some experimental information [118]. For this reason, the PSF available in the literature are only a good starting point for the simulation of

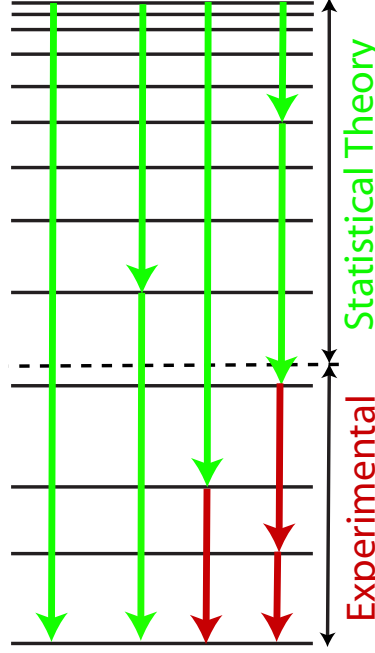


Figure 5.4: Nuclear level scheme used in the cascade generator model. The low energy range is known experimentally, while the upper energy range is described from nuclear level density statistical models and parameterized EM transition probabilities.

capture cascades but need to be adjusted in order to reproduce the experimental data. The (n,γ) measurement with the TAC have been shown to be a sensitive way for performing systematic PSF studies [114, 119].

5.1.4.2 Validation with the $^{197}\text{Au}(n,\gamma)$ measurement

The neutron capture generator for the specific case of the $^{197}\text{Au}(n,\gamma)$ measurement uses as input: (i) at low energies and up to 1.4 MeV, the nuclear levels and transition probabilities found in the ENSDF data base [115]; (ii) at higher excitation energies, the Back Shifted Fermi Gas model for the calculation of the level density with the parameters recommended in the RIPL-2 database [41], and the PSF for the calculation of the transition probabilities with the parameterization of Kopecky et al. [117]. This *initial PSF* (see Table 5.1) is described by two Generalized Lorentzian distributions for E1 transitions, and a standard Lorentzian distribution for M1 and E2 transitions. A first simulation was performed with the *initial PSF* and assuming a constant counting rate of 0.9 counts/ μs , which corresponds to the experimental counting rate (see Figure 5.7) registered at the flat top part of the 4.9 eV saturated resonance.

Figure 5.5 shows the distributions of deposited energy and crystal multiplicity resulting from the simulation (dotted blue line) and the experiment (black line). The clear disagreement between the simulation (*initial PSF*) and the experimental data observed in Figure 5.5 is also

	E1		M1	E2
E (MeV)	13.72	5.8	7.05	10.81
Γ (MeV)	4.61	1.5	4.00	3.73
σ_0 (mb)	541.0	6.0	1.12	5.03

 Table 5.1: Parameters of the Initial PSF for ^{198}Au .

found when looking at the distributions of energy deposited in the individual crystals for specific values of the crystal multiplicity, which are shown in Figure 5.6.

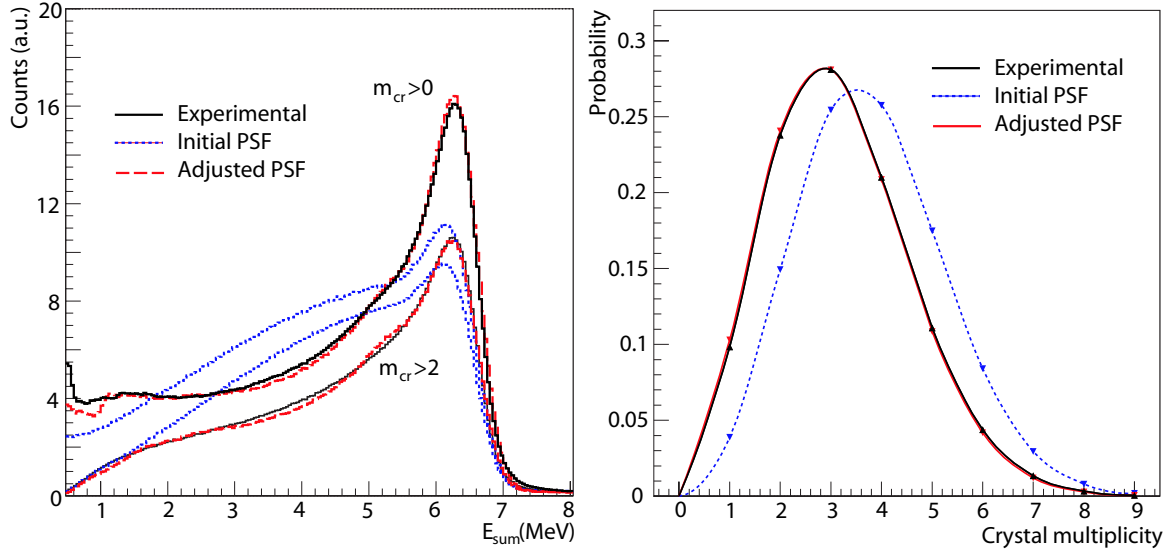


Figure 5.5: TAC response to $^{197}\text{Au}(n,\gamma)$: deposited energy (left) and crystal multiplicity (right). Black: experimental. Blue: simulated using the initial PSF. Red: Simulated using the adjusted PSF.

The results suggest that the theory (*initial PSF*) overestimates the multiplicity of the capture cascades and underestimates the intensity of primary high energy transitions. Using these observations to fine tune the parameters of the PSF, a good reproduction of the experimental data was obtained after:

- increasing the intensity of E1 transitions around S_n in order to decrease m_{cr} , because these are the only possible transitions from the capture state ($2+$) to directly populate the 2^- ground state and the lowest levels laying levels at 55 keV and 192 keV, both with a spin 1^- .
- introducing two small resonances (only one is introduced by Kopecky et al.) in the E1 transitions at 1.1 and 5.5 MeV,
- decreasing the energy and width of the Lorentzian describing the M1 transitions.

The parameters of the *adjusted PSF* are given in Table 5.2 and the corresponding simulation is represented by the dashed-red lines in Figures 5.5. It is observed that the simulated (*adjusted PSF*) and the experimental distributions are in excellent agreement.

	E1			M1	E2
E (MeV)	6.38	5.5	1.1	5.05	10.81
Γ (MeV)	0.042	1.5	0.8	1.5	3.73
σ_0 (mb)	500.	6.5	0.2	2.0	5.03

Table 5.2: Parameters of the ^{198}Au PSF adjusted below S_n to reproduce the present experimental data.

The energy spectra of the individual crystals for each value of m_{cr} are also very sensitive to the characteristics of the capture cascade and are complementary to the overall E_{sum} and m_{cr} distributions. For this reason, the reproduction of the experimental data by the simulation for all combinations of deposited energy and multiplicity (see Figure 5.6), gives additional confidence in the assumption that the detection efficiency calculated from the simulation is an accurate estimate for the true value.

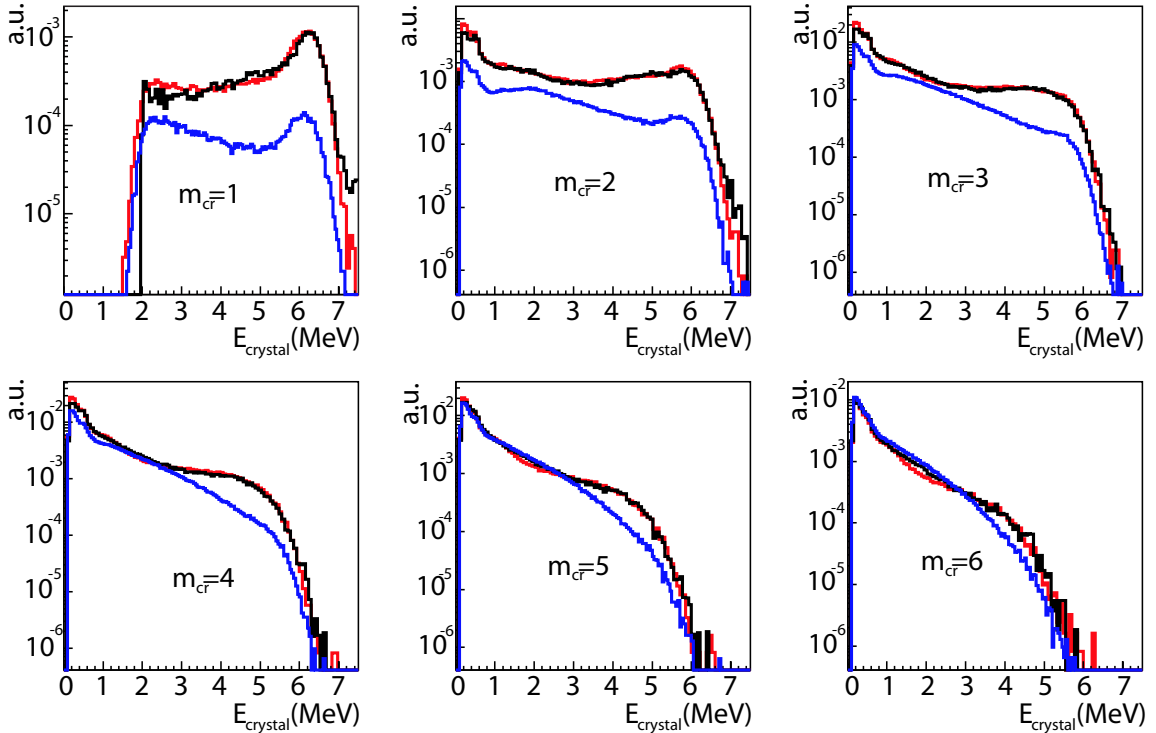


Figure 5.6: $E_{crystal}$ for $^{197}\text{Au}(n,\gamma)$ events with $E_{sum} > 2$ MeV for each m_{cr} . Black: measured. Blue: simulated with the initial PSF. Red: simulated with the adjusted PSF.

5.1.5 The detection efficiency of the TAC: the case of ^{197}Au

The detection efficiency of the TAC for any conditions on E_{sum} and m_{cr} is directly calculated from the MC simulation as the ratio of detected events, satisfying the selected conditions, to the number of initial events. Since the primary events in the simulations are distributed in time-of-flight according to the experimental counting rate distribution, the resulting detection efficiency is also a function of time-of-flight, i.e. neutron energy.

The ratio between the characteristic time variations of the counting rate and the dead-time of the crystals ($DT < 4 \mu\text{s}$) determines if the counting rate can be approximated as constant or not. An illustrative example is given in Figure 5.7, which shows the counting rate recorded during the ^{197}Au measurement: in the long time-of-flight region (right) the resonant structures occur within milliseconds ($\gg DT$) while in the short time-of-flight region (left) the resonances occur within a few μs ($\sim DT$). The difference in the effect of the dead-time is that in the first case, the counting rate in one time-of-flight bin does not affect neighbouring bins, while in the second case the counting rate in one bin produces dead-time losses in the neighbouring bins at later times.

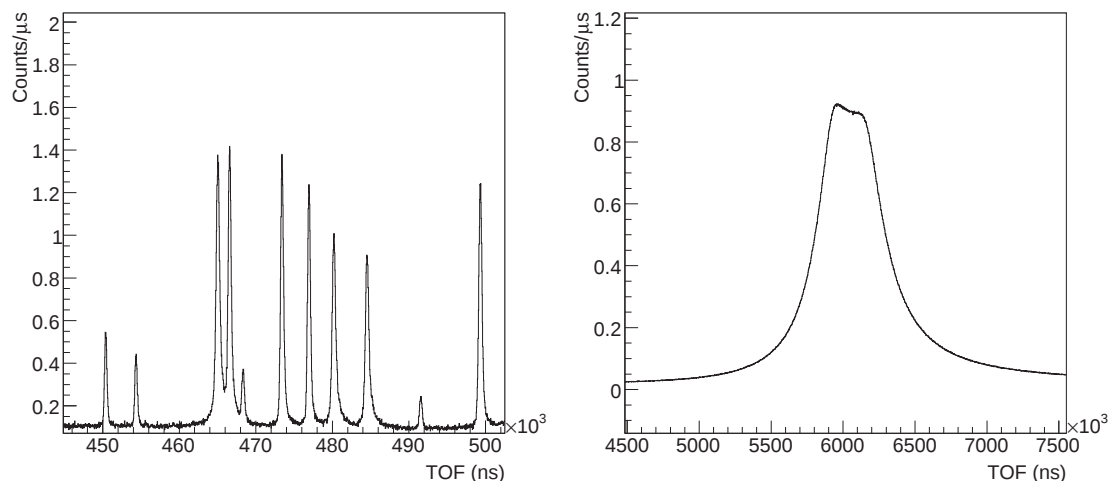


Figure 5.7: Counting rate recorded during the $^{197}\text{Au}(n,\gamma)$ measurement and used for the distribution in time-of-flight of the capture events in the simulations.

In the following discussion the detection efficiency is calculated for three cases:

1. monoenergetic γ -rays at different counting rates,
2. the $^{197}\text{Au}(n,\gamma)$ measurement at constant counting rates,
3. the $^{197}\text{Au}(n,\gamma)$ measurement using the experimental counting rate distribution.

5.1.5.1 Detection efficiency for monoenergetic γ -rays

The detection efficiency as well as the total absorption efficiency for monoenergetic γ -rays are illustrated in Figure 5.8. The values correspond to the efficiencies calculated with and without the neutron absorber.

The results show that the use of the neutron absorber affects only the total absorption efficiency while the detection efficiency remains unchanged. The values of the detection efficiency, larger than 80% for all energies, indicate that the detector is an excellent total absorption calorimeter for neutron capture measurements: the detection efficiency for a cascade of any energy with at least two γ -rays is higher than 95%.

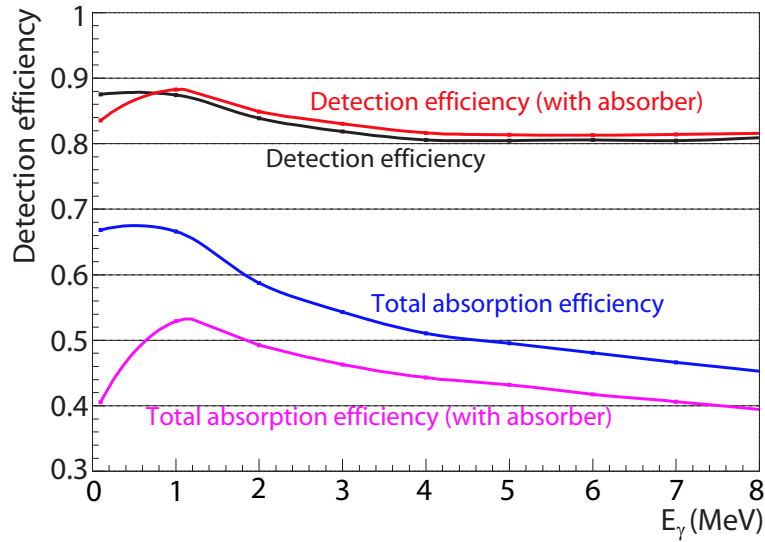


Figure 5.8: Detection and total absorption efficiencies of the TAC to monoenergetic γ -rays, with and without using the neutron absorber.

5.1.5.2 Detection efficiency at a constant counting rate

The response of the TAC to $^{197}\text{Au}(n,\gamma)$ reactions at a constant counting rate has been simulated. The resulting E_{sum} distributions are shown in Figure 5.9, where the two panels correspond to the detected events with (right) and without (left) analysis conditions. With increasing counting rate, the dead-time effect give rise to increasing losses in the total number of detected events. This affects mainly the sum energy peak.

The detection efficiency has been calculated for counting rates from 0 to 2 counts/ μs for several combinations of conditions on E_{sum} and m_{cr} . The values listed in Table 5.3 show that the detection efficiency at low counting rates is very close to 100%.

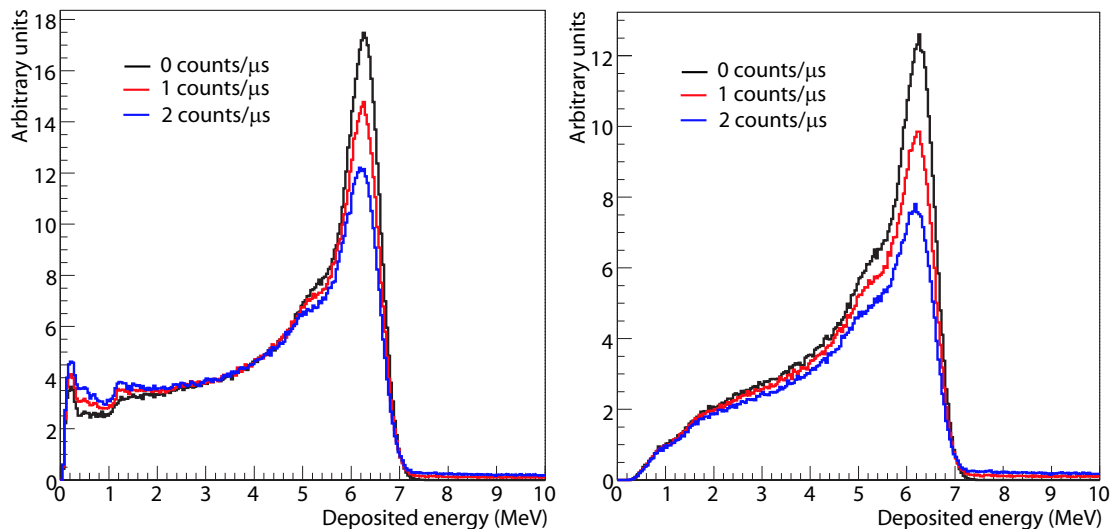


Figure 5.9: Simulated deposited energy distributions of $^{197}\text{Au}(n,\gamma)$ with (right) and without analysis conditions (left) for different counting rates.

Event selection		ϵ	Dead-time losses (%)			
m_{cr}	$E_{sum}(\text{MeV})$		$0.5 \mu\text{s}^{-1}$	$1 \mu\text{s}^{-1}$	$1.5 \mu\text{s}^{-1}$	$2 \mu\text{s}^{-1}$
> 0	> 0.1	.974	1.6	3.2	4.8	6.3
> 0	> 1.0	.912	2.2	4.4	6.6	8.6
> 0	> 2.5	.789	3.1	6.1	9.0	12
> 1	> 1.0	.843	3.1	6.2	9.3	12
> 1	> 2.5	.738	3.9	7.6	11	16
> 2	> 1.0	.656	5.4	10	16	21
> 2	> 2.5	.591	5.9	11	17	22

Table 5.3: Detection efficiency of the TAC calculated for different event selection conditions. The dead-time losses are given as a function of the counting rate and of the selection conditions.

The dead-time losses are not only depending on the counting rate but are also affected by the conditions chosen for the event selection. Although an accurate model has been implemented for calculating the dead-time losses, the results in Table 5.3 indicate that it is desirable to keep the counting rate of the measurements below $1 \mu\text{s}^{-1}$ in order to avoid large corrections.

5.1.5.3 Detection efficiency at a variable counting rate

When the simulated capture events are distributed in time according to the measured density of events, the resulting efficiency is a point-wise function of neutron energy. Dividing the experimental time-of-flight spectra with this efficiency yields corrected spectra where dead-time losses are properly corrected.

Figure 5.10 illustrates the effect of these corrections for three energy regions in the TOF spectrum of ^{197}Au . While the original data are indicated by the black curve, the properly corrected spectra are given in red. The two additional lines correspond to different approximations:

- the green lines correspond to the recorded distribution corrected by the detection efficiency without including dead-time losses,
- the blue lines correspond to the recorded distribution corrected under the assumption of a constant counting rate, which means that each time-of-flight bin is affected only by its own counting rate.

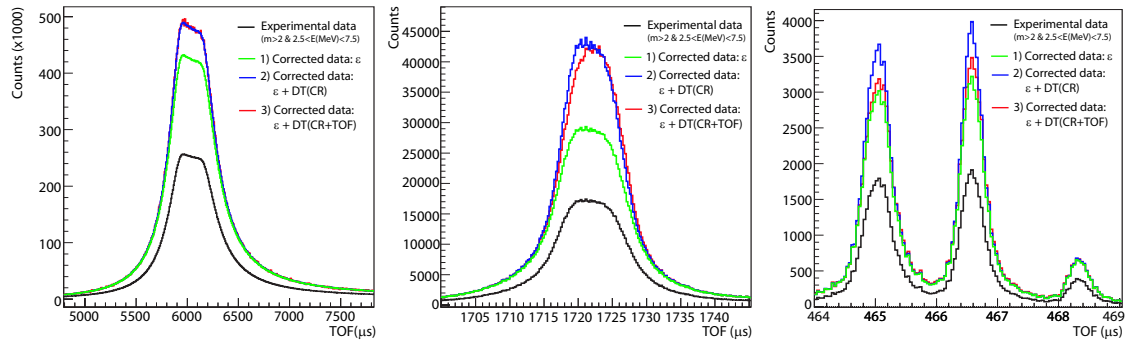


Figure 5.10: Measured counting rate corresponding to the $^{197}\text{Au}(n,\gamma)$ measurement corrected by the detection efficiency calculated under different assumptions. The full details are given in the text.

The conclusions that can be reached from the analysis of the three time-of-flight intervals shown in Figure 5.10 are:

- At long flight times (low E_n), the width of the resonances corresponds to a few hundred μs ($\gg \text{DT}$), hence the constant counting rate approximation is valid.
- At short flight times (high E_n), looking at the resonance with the highest counting rate ($\sim 2.5 \mu\text{s}^{-1}$ at 60 eV) and a width comparable to the dead-time, the dead-time losses introduce an asymmetry in the shape of the resonance due to the effect of each bin in later bins. Such effect is corrected properly only if the realistic distribution of events according to the experimental counting rate is used.
- At very short flight times (very high E_n), the widths of the resonances are smaller than the dead-time and the constant counting rate approximation clearly overestimates the correction.

Finally, it can be concluded that the resulting yields suffer from systematic uncertainties as large as 30% if the dead-time is not included in the calculation of the detection efficiency. These uncertainties depend on the characteristics in terms of the TOF structure and of the and multiplicity of the capture cascades.

5.1.6 Uncertainty in the detection efficiency

There are two main sources of uncertainty in the calculation of the detection efficiency by Monte Carlo simulations:

- The first is related to the fact that the strategy followed for the search of the appropriate PSF does not allow to determine a unique parameterization but several that yield equivalent results. In order to estimate the uncertainty in ε due to the choice of the PSF parameters, the TAC response has been simulated for several PSFs that reproduce the experimental data in an approximated way. The detection efficiencies for these PSFs have been calculated. The standard deviation of the results showed that the maximum uncertainty in ε is 2.5%.
- The second is related to the dead-time correction model. The uncertainty caused by the dead-time correction model has been estimated by performing different calculations with: (i) different dead-time models (paralizable and non-paralizable), (ii) different counting rate distributions for the primary events (experimental and calculated from the evaluated cross section), and (iii) using different values of the dead-time $DT(E_i, E_j)$ in each crystal with variations up to $\pm 15\%$. The results show that if the counting rate is kept below 1.5 counts/ μ s, the uncertainty in the correction is lower than 4%, which corresponds to a maximum uncertainty in ε of the order of 1%.

Event selection		Partial uncertainty		ε
m_{cr}	$E_{sum}(MeV)$	PSF	Dead-time	
>0	>0.1	0.003	0.001	0.932 $\pm$ 0.004
>2	>2.5	0.013	0.005	0.533 $\pm$ 0.014

Table 5.4: *Detection efficiency and its associated uncertainty for the measurement of ^{197}Au at the 4.9 eV saturated resonance with and without applying the event selection conditions.*

The values of the detection efficiency of the TAC for the measurement of $^{197}\text{Au}(n, \gamma)$ at the 4.9 eV saturated resonance and the corresponding uncertainty associated to the MC simulation and dead-time correction model are summarized in Table 5.4. If the counting rate is kept below 1 μs^{-1} , the total uncertainty in the detection efficiency is below 3%, even for the most restrictive conditions ($m_{cr} > 2$ and $E_{sum} > 2.5$ MeV).

5.2 Experimental capture yield of ^{197}Au

The experimental capture yield of ^{197}Au was calculated as

$$Y_{n,\gamma}(E_n) = \frac{C(E_n) - B(E_n)}{\varepsilon(E_n, m_{cr}, E_{sum}) \cdot N \cdot \phi_n(E_n)}, \quad (5.3)$$

where $C(E_n)$ and $B(E_n)$ are the total and background counting rates, ε is the efficiency of the detector calculated from the MC simulations, $\phi_n(E_n)$ is the neutron intensity recorded in the neutron monitor *SiMon*, and N is the normalization factor that relates the neutron intensity in the monitor to that of the neutrons impinging on the sample.

The choice of ^{197}Au for the validation of the measuring and analysis techniques is due to several reasons:

1. It has been previously used as a reference for neutron capture measurements at n_TOF and also other facilities.
2. It presents a strong resonance at 4.9 eV that can be analyzed in order to provide an accurate value of the normalization factor N .
3. The counting rate registered during this measurement was about two times higher than those of the measurements on the minor actinides which are the core of this work. In this way the analysis of ^{197}Au allows to validate the dead-time correction model.
4. The scattering to capture cross section ratio of this measurement is larger than in the measurements on the minor actinides. Therefore, the analysis of ^{197}Au allows one to validate the techniques developed for the calculation of the neutron scattering background.

5.2.1 Total and background counting rates

The $^{197}\text{Au}(n,\gamma)$ measurements have been performed using a disk of pure gold (>99.9) 1 cm in diameter and 185.4 mg in mass. The analysis conditions that have been chosen correspond to $2.5 < E_{sum}(\text{MeV}) < 7.0$ and $m_{cr} > 2$. Under these conditions the largest background contribution is due to the scattering of neutrons in the sample. It has been determined by comparing the deposited energy distribution registered in the measurement of gold with the one registered in the measurement of a graphite sample (see Section 4.2.4).

The neutron scattering background, which shows a resonant structure similar to that of the capture events, has been found to be 1.6% for the saturated resonance at 4.9 eV and as large as 6% in the case of the resonance at 60 eV. The overall background is characterized in a point-wise as a function of neutron energy and is included in the analysis of the capture yield. The experimental capture rate and the background contribution are shown in Figure 5.11 in the energy range between 1 eV and 100 eV.

5.2.2 Normalization to the saturated resonance at 4.9 eV

The Saturated Resonance Method [120, 68] allows one to calculate the normalization factor N that relates the neutron intensity recorded in the neutron monitor with the number of neutrons impinging on the sample. A resonance suitable for this method must satisfy the following

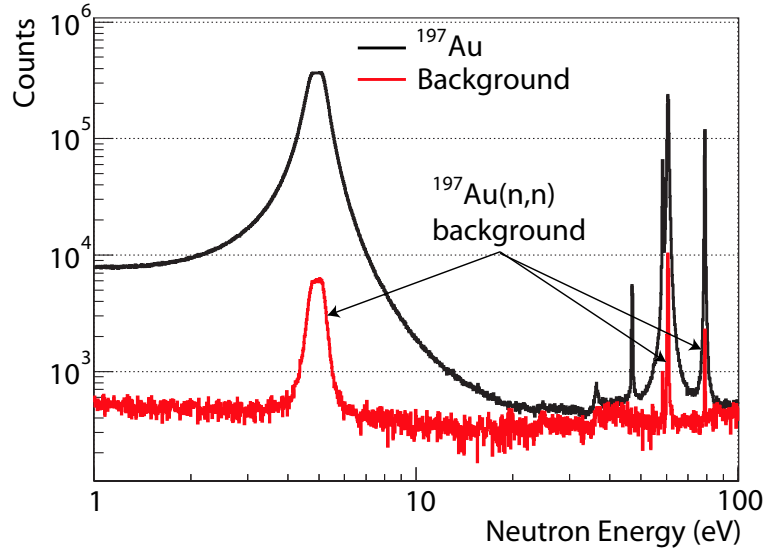


Figure 5.11: Measured counting rate as a function of E_n (red) and calculated background (blue) for ^{197}Au .

requirements: (i) it should have a large peak cross section so that the capture yield at the top of the resonance is saturated (all the incident neutrons interact with the sample), (ii) its radiative width should be much larger than its neutron width so that the uncertainty due to scattered neutrons and that of the calculation of the saturation value are minimized.

The resonance of ^{197}Au at 4.9 eV satisfies both conditions: $(1 - e^{-n\sigma}) > 0.9999$ and $\Gamma_\gamma \simeq 8 \cdot \Gamma_n$. Since the value at the saturated region of the resonance is well known, the value of the normalization N is obtained from the fit of the experimental yield to the expected value in the saturated region.

The fit was performed using the SAMMY code, which allows one to calculate the yield from the evaluated resonance parameters and the experimental conditions of the measurement, i.e. temperature, thickness of the sample, resolution function of the neutron beam, etc. The normalized experimental yield is displayed in Figure 5.12 together with the yield calculated from the resonance parameters of the JEFF-3.1 evaluation, showing that the agreement between both yields is excellent not only in the saturated region but also in the tails of the resonance.

The sources of uncertainty in the normalization factor are related to the detection efficiency of the TAC (3%) and the uncertainty in the theoretical yield related to the accuracy of its resonance parameters (1% [121]), resulting in a 3.2% uncertainty of the normalization factor N . Since the normalization factor depends on the neutron monitor and the position and size of the investigated sample, the value obtained for ^{197}Au measurement can be used for any other measurements given that the size and position of the sample are exactly the same.

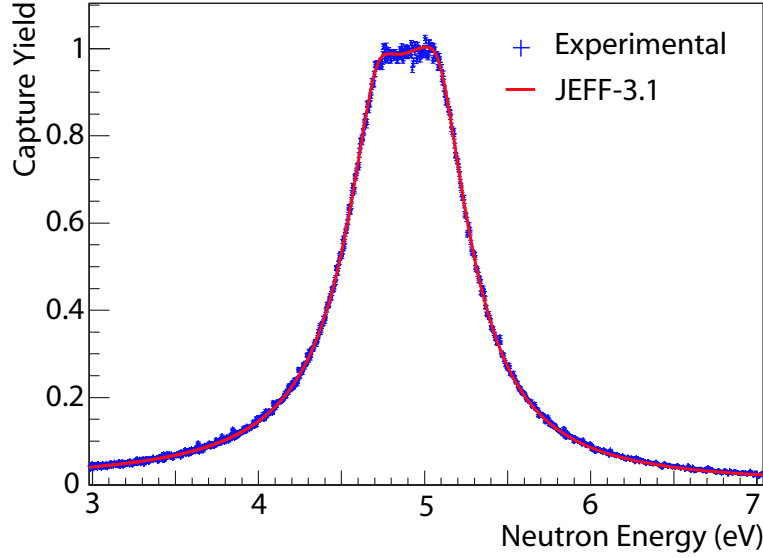


Figure 5.12: *Experimental capture yield of the 4.9 eV saturated resonance of ^{197}Au compared to the yield calculated from the resonance parameters of the JEFF-3.1 evaluation.*

5.2.3 Comparison to the evaluated capture yield

After applying all the necessary corrections the systematic uncertainty in the experimental yield is better than 3.5%. This value corresponds to the combined uncertainties of the energy dependence of the neutron intensity (2% [27]) and the normalization (3%), since the uncertainty of the detection efficiency has been already included in that of the normalization. The uncertainty related to the dead-time correction can increase the uncertainty to an overall 4% in the case of resonances with a high counting rate.

The experimental capture yield has been compared to the yield calculated from the evaluations in the complete energy range between 1 eV and 1 keV, see Refs. [122, 123]. Overall, the comparison indicates that the experimental and calculated yields are compatible within uncertainties. Indeed, the discrepancies that are observed are not correlated with the counting rate, neither with the size of the neutron scattering background or the neutron energy. Otherwise, this would have indicated a problem with the corrections applied in the data analysis.

The comparison for some resonances is illustrated in Figure 5.13. The set of 15 resonances shown in the figure has been chosen because it covers the wide range of situations that were identified in the comparison. The upper panels illustrate the overall agreement between the experimental and calculated yield, even for resonances with very different values of the counting rates and the scattering over capture ratio. The situation displayed in the left-bottom panel illustrates the improved energy resolution of the measurements at n.TOF over previous measurements. Last, the right-bottom panel illustrates the significant differences that are found for some resonances, even between the different evaluations, and how there is not any correlation

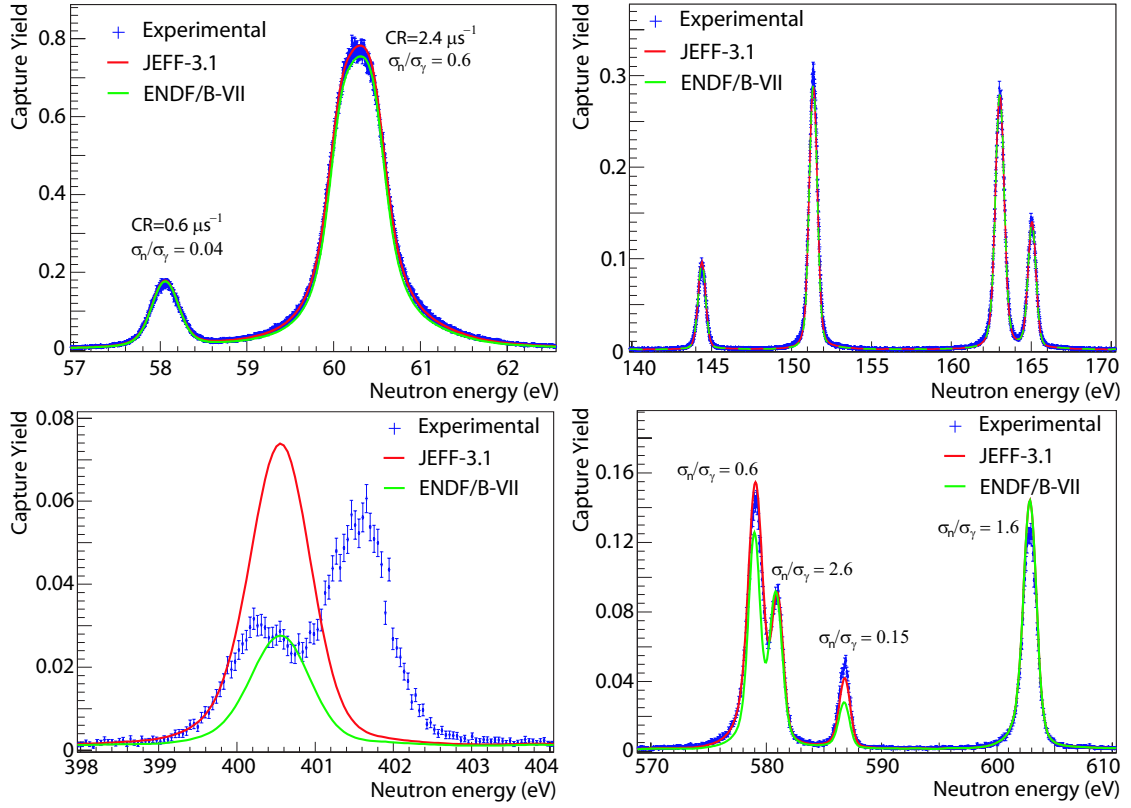


Figure 5.13: Comparison of the experimental and evaluated capture yield of ^{197}Au .

with the neutron sensitivity background or the counting rate.

This result is a final proof of the consistency of all the techniques that have been developed for the calculation of the neutron scattering background, the determination of the detection efficiency, the corrections related to dead-time losses, and the normalization to the 4.9 eV resonance.

Part III

CROSS SECTION MEASUREMENTS AND ANALYSIS

Chapter 6

Measurement and analysis of the $^{237}\text{Np}(n,\gamma)$ cross section

The first experiments aimed at measuring the neutron cross section of ^{237}Np date from the early fifties. Since then, the total and fission cross sections have been measured with reasonable accuracy, which is not the case concerning the capture reaction channel. The main reason for this is that many of the previous measurements used massive samples with a high activity that added significant difficulties to the measurements and their analysis. As a consequence, the most recent evaluated cross sections in the Resolved Resonance Region are based only on the analysis of transmission and fission data. In the Unresolved Resonance Region, the evaluated data are mostly referring to the work of Weston et al. from 1981 [124].

The available experimental capture data differ significantly from each other, even in the case of the most recent measurements using the total absorption technique. The result of a systematic study of the measurements and evaluations [22] are summarized in Table 6.1, showing that the uncertainty in the cross section of ^{237}Np is of the order of 15% to 40%.

Neutron energy	Uncertainty		
	$\sigma(n, f)$	$\sigma(n, n)$	$\sigma(n, \gamma)$
0.025 eV - 2 MeV	25%	5%	15%
2 MeV - 20 MeV	25%	5%	40%

Table 6.1: ^{237}Np cross section uncertainties.

The first goal of this work is to measure the capture cross section of ^{237}Np in the epithermal energy range and to provide the nuclear data community with a set of high quality experimental data, with a good control of the systematic uncertainties, suitable for its use in future evaluations for reactor calculations. The second goal is to analyze the experimental data and to obtain the corresponding cross section and the associated uncertainty in the resolved and unresolved

resonance regions. The analysis has been performed in two complementary ways: based on the n_TOF capture yield normalized to the 4.9 V resonance of ^{197}Au , and using the n_TOF capture yield in combination with the most reliable and recent transmission data.

6.1 Previous measurements and evaluations

There have been a significant number of experiments aimed at measuring the capture cross section of ^{237}Np in different energy ranges, which are briefly described in the following. In addition, some of the transmission and fission measurements that have been included in different evaluations are also presented.

6.1.1 Previous measurements

In 1957, Smith et al. [125] performed at a nuclear reactor the first measurement that gave access to the resonance parameters of ^{237}Np , but only in the interval between 0.02 eV and 2.8 eV. The thermal cross section was determined to be $\sigma_0 = 180 \pm 22$ barns and the first four resonances were analyzed with the Breit-Wigner formalism.

In 1972, Paya et al. [126] measured the total and fission cross sections of ^{237}Np between 0.5 eV and 45 keV at the 16.7 m and 53.7 m time-of-flight stations of the Saclay Linear Accelerator [127]. Three different samples were used. The resonance analysis, in the range from 0.5 to 250 eV, was reported later by Plattard et al. [128] using an analysis program based on the Breit-Wigner formalism for taking the interference between neighbouring resonances into account.

In 1976, Hoffman et al. [129] measured the total, scattering and capture cross sections of ^{237}Np from 20 eV to 250 keV using a metallic foil of $8.7 \cdot 10^{-4}$ at/barn and the neutron beam generated in an underground nuclear explosion. The γ -rays from neutron capture reactions were recorded with Moxon-Rae detectors. The authors claimed an uncertainty in the resulting capture cross section of 30%.

In 1978, Mewissen et al. [130] measured the transmission, capture and elastic scattering reactions between 8 eV and 204 eV using four different samples at the 30 m time-of-flight station of the GELINA facility [47]. The resonance analysis of the capture data consisted only in the study of the resonance integrals, while the resonance parameters were determined from the analysis of the transmission data.

In 1981, Weston et al. [124] measured the $^{237}\text{Np}(n,\gamma)$ cross section between 0.01 eV and 200 keV at the 20 m time-of-flight station of the ORELA facility [63]. The data were normalized to the cross section of Paya et al. The resonance integral and partial widths of all resonances below 100 eV were analyzed. This work has been the basis of most evaluations before 2002 and is still nowadays the reference measurement in the Unresolved Resonance Region.

In 1984, Auchampaugh et al. [131] performed transmission and fission measurements between 1 eV and 600 eV at ORELA in order to investigate the sub-threshold fission cross section of ^{237}Np . The combination of the long flight path (78 m), low sample temperature (77 K) and high sample mass (1.35 g) resulted in the highest energy resolution so far and enabled the identification of new resonances missed in previous experiments. The resolved resonances were analyzed between 1 and 600 eV using a fitting program with the Reich-Moore formalism, although the authors recall that the analysis above 160 eV is not as accurate as that achieved at lower energies. An important contribution of this work consisted in the determination of the spin of all observed resonances observed.

In 1999, Gressier et al. [132] performed the most recent total cross section measurement at the 26 m and 50 m time-of-flight stations of the GELINA facility [47]. The measurement was performed with seven samples of different thicknesses. The simultaneous resonance analysis of the three reliable transmission measurements was performed below 500 eV using the REFIT code [75]. The Crystal Lattice Model for the Doppler broadening was adopted for the analysis of the lowest lying resonances. The results were compatible within uncertainties with those of Paya et al, although not all the resonances reported by Auchampaugh could be observed. The experimental campaign included also three neutron capture measurements using C_6D_6 detectors, but, due to the high intrinsic activity of the samples the capture data were only used for the identification of small resonances that are more difficult to observe in transmission.

The last capture measurement using total energy detectors was performed in 2002 by Kobayashi et al. [133] at KURRI from 5 meV to 10 keV. The sample was surrounded by a 1 cm lead shield in order to reduce the background caused by its intrinsic activity. The results show large differences (up to 50% in some energy regions) with respect to the evaluated cross sections and previous experimental data.

In addition to the neutron capture measurement that is presented in this work, two recent measurements have combined the total absorption technique with digital electronics:

1. Esch et al. [134] used the DANCE detector to measure the capture cross section of ^{237}Np between 20 meV and 200 keV at the 20 m flight path of the LANSCE facility using a 0.44 mg sample. The capture yield was normalized to the resonance at 0.49 eV and resolved resonances were observed only up to 20 eV. A recent work by Noguere et al. [135] indicates that these data suffer from a systematic uncertainty of 7.5% in the normalization.
2. Scherbakov et al. [136] used a small total absorption detector consisting of 8.54 litres of BGO at the KURRI facility to measure the average capture cross section of ^{237}Np in a few energy intervals between 0.02 eV and 100 eV. The capture yield was normalized to the absorption cross section of ^{10}B .

To summarize, it can be stated that there are accurate transmission and fission measurements of ^{237}Np , but the existing neutron capture measurements do not have enough resolution (Scherbakov), suffer from systematic uncertainties (Hoffman, Mewissen, Gressier, Kobayashi and Esch) and/or lack sufficient statistics (Weston) for an accurate resonance analysis in the

complete resolved resonance region. In the unresolved resonance region, the data of Weston et al. are the basis of the evaluations but the most recent data (Kobayashi and Esch) disagree with the values of Weston beyond the uncertainties declared by the authors.

For all these reasons, the n-TOF Collaboration considered it necessary to perform a new measurement with high statistics, enough energy resolution and a good control of systematic uncertainties in the widest possible energy range, including both the resolved and unresolved resonance regions. This is the goal of the work presented in this chapter.

6.1.2 Evaluated cross sections

At the time of this work, the most recent Evaluated Nuclear Data Files for the ^{237}Np neutron cross sections are JENDL-3.3 (2002), JEFF-3.1 (2005) and ENDF/B-VII (2006).

The evaluated thermal capture cross section of ^{237}Np has been a matter of discussion in recent years [137]. The evaluated value of the thermal capture cross section before 2002 (JEFF-2.2, ENDF/B-VI.8 and JENDL-3.2) was $\sigma_\gamma^{th} = 181$ barns, which was changed to $\sigma_\gamma^{th} = 162$ barns in the later evaluations (JEFF-3.1, JENDL-3.3, ENDF/B-VII). However, the latest validations with integral experiments of the ^{237}Np thermal capture cross section [137] recommend increasing the value in the present evaluations by $12 \pm 2\%$, giving a value consistent with the JEFF-2.2 evaluation. The cross section values quoted in the JEFF evaluations and the corresponding resonance parameters with negative energies are summarized in Table 6.2.

	E_n (eV)	J	Γ_γ (meV)	Γ_n (meV)	σ_{tot} (barns)	σ_γ (barns)	σ_f (barns)
JEFF-2.2	-1.0	2	40.0	2.27	195.778	181.020	0.018
JEFF-3.1	-3.49	3	40.0	2.18	175.79	161.71	0.0204
	-0.56	2	40.0	0.45			

Table 6.2: Thermal cross section of ^{237}Np and associated negative resonances in the JEFF-2.1 and JEFF-3.1 evaluations.

The value of the capture cross section at thermal energies is important for the analysis of the lowest resonances lying on top of the $1/v$ tail starting at the thermal point. Therefore, choosing one or another value may affect the result of the analysis at low energies. In this work the uncertainty related to this choice has been determined.

In the most recent evaluations, the limit between the resolved and the unresolved resonance regions is found at 500 eV. The choice of this limit is arbitrary and does not correspond to the energy values at which the resonances start to overlap.

In the resolved resonance region all the present evaluations recommend the same set of

resonance parameters corresponding to the MLBW formalism. The neutron and radiative widths correspond to the analysis of transmission data by Gressier et al. [132] and Auchampaugh et al. [131]. The fission widths are a combination of data from the measurements of Auchampaugh et al. [131], Borzakov et al. [138] and Dermendjiev et al. [139]. In the Unresolved Resonance Region, that ranges up to 35 keV, the evaluated cross sections are essentially based on the values of Auchampaugh et al. [131] in the case of the total cross section, Weston et al. [124] for neutron capture, and Yamanaka et al. [140] and Iwasaki et al. [141] for the fission cross section.

6.2 The $^{237}\text{Np}(n,\gamma)$ measurement at n_TOF

Thanks to the excellent features of the experimental set-up at n_TOF (high instantaneous flux, high detection efficiency, powerful background reduction capabilities and well validated analysis techniques), the present results are the first capture data suited for resonance analysis in the complete resolved resonance region. The systematic uncertainties related to the measurements and to the analysis techniques have been reduced and well determined, thus providing a set of highly reliable data for the update of previous evaluations.

6.2.1 The ^{237}Np sample

The ^{237}Np sample was manufactured at IPPE Obninsk [96] from $^{237}\text{NpO}_2$ powder, which was deposited on a disk shaped aluminium backing 1 cm in diameter and then sandwiched between 0.15 mm thick aluminium layers. According to the requirements imposed by the radioprotection service at CERN, the ^{237}Np sample was further encapsulated in a 0.2 mm thick titanium container to satisfy the *ISO-2919 norm*. The sample was mounted between two 25 μm thick Kapton foils stretched over a carbon fibre ring, which served as a sample holder for the placement inside the TAC (see Figure 3.11). The physical description of the sample is given in Table 6.3.

$^{237}_{93}\text{Np}$	$A+1S_n$	5488.32 keV
	I^Π (GS)	+5/2
	Half-life	$2.1 \cdot 10^6$ years
Sample	Chemical form	$^{237}\text{NpO}_2$
	Activity	1.29 MBq
	Isotopic mass	43.3 ± 1.3 mg
	Thickness	$(1.40 \pm 0.04) \cdot 10^{-4}$ at/barn
	Diameter	1 cm
	Enrichment	99.2%
	Impurity (^{238}Pu)	0.8%

Table 6.3: *Main properties of the $^{237}\text{NpO}_2$ sample and the neutron capture measurement.*

The high purity of the sample was assessed at CERN by γ spectroscopy with germanium detectors, confirming that the daughter product ^{233}Pa was not yet in secular equilibrium with the α decay of ^{237}Np .

6.2.2 The $^{237}\text{Np}(n,\gamma)$ measurement

The measurements with the TAC provide a list of detected events, each characterized by the deposited energy (E_{sum}), crystal multiplicity (m_{cr}), time-of-flight (TOF), and the corresponding neutron energy (E_n).

The energy spectra measured with the ^{237}Np sample and in the dedicated background runs are displayed in Figure 6.1. The contribution due to the $^{237}\text{Np}(n,\gamma)$ reactions is observed up to the neutron separation energy of the compound nucleus $S_n(^{238}\text{Np})=5.49$ MeV. Most of the different background sources are common to any measurement with the TAC and were discussed in detail in Section 4.2.2. However, the background caused by the sample assembly is specific to the measurement of encapsulated samples. This background presents two characteristic total absorption peaks at 6.4 MeV and 8.1 MeV, which correspond to $^{50}\text{Ti}(n,\gamma)$ and $^{48}\text{Ti}(n,\gamma)$ reactions. The situation is different at larger neutron energies where neutron scattering becomes the dominant reaction in Ti, which contributes to the structures produced by neutron capture in barium.

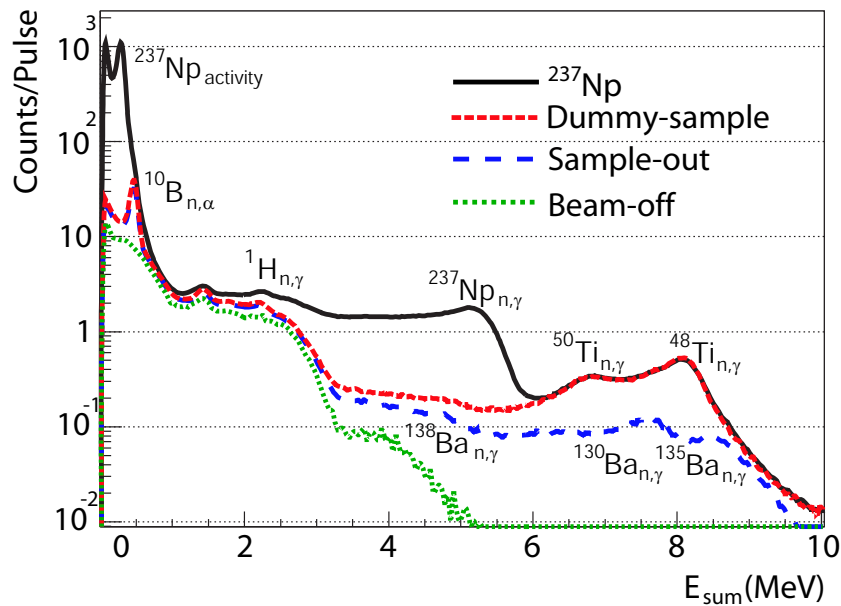


Figure 6.1: Measured deposited energy distributions corresponding to ^{237}Np and the dedicated background measurements in the neutron energy interval between 1 and 10 eV.

The counting rate distributions as function of neutron energy for ^{237}Np and the related back-

ground measurements are shown in Figure 6.2. The background in the beam-off and sample-out measurements is common to any measurement with the TAC and was discussed in Section 4.2.2. On the other hand, the background caused by the sample assembly dominates the measurement above a few keV where it shows sharp structures at neutron energies above a few keV that correspond to resonances in Ti. Consequently, the capture cross section could only be measured up to a few keV.

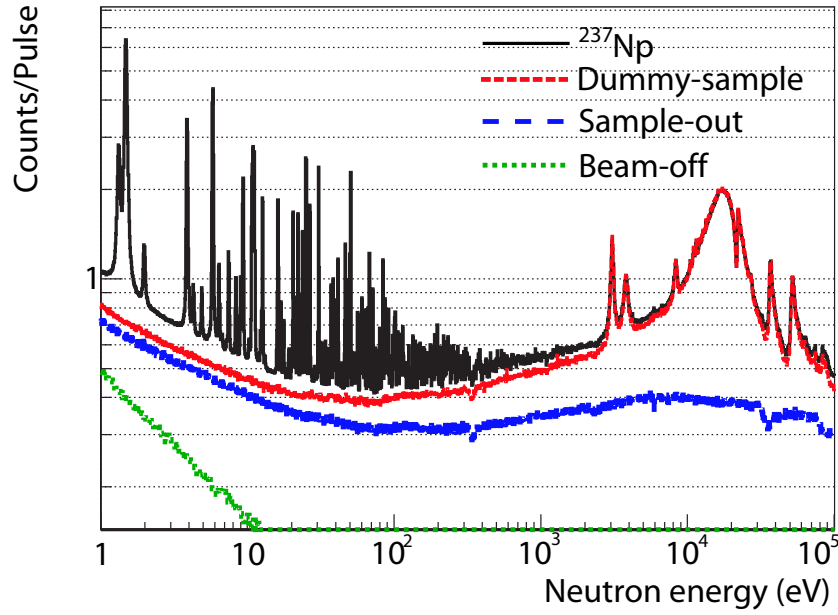


Figure 6.2: Measured counting rate distributions corresponding to ^{237}Np and the dedicated background measurements for deposited energies between 2.5 and 6 MeV, where the capture to background ratio is more favourable.

The capture to background ratio of the measured data is reduced by selecting events that satisfy some specific conditions for E_{sum} and m_{cr} . This is illustrated in Figure 6.3, which shows the distribution of deposited energy of ^{237}Np under different conditions for the crystal multiplicity. Selecting events with $m_{cr} > 2$ eliminates the major part of the background without affecting the $^{237}\text{Np}(n,\gamma)$ contribution significantly. Since most of the $^{237}\text{Np}(n,\gamma)$ events are found below 6 MeV, the events selection criteria that have been chosen for the analysis are $m_\gamma > 2$ and $2.5 < E_{sum}(\text{MeV}) < 6$.

The background related to neutron scattering in the sample has been determined, see Section 4.2.4, and found to be lower than 1% in the complete energy range. This was expected because the capture to scattering ratio in ^{237}Np is very large ($\frac{\sigma_\gamma}{\sigma_n} > 10^3$) in the energy range considered in this work.

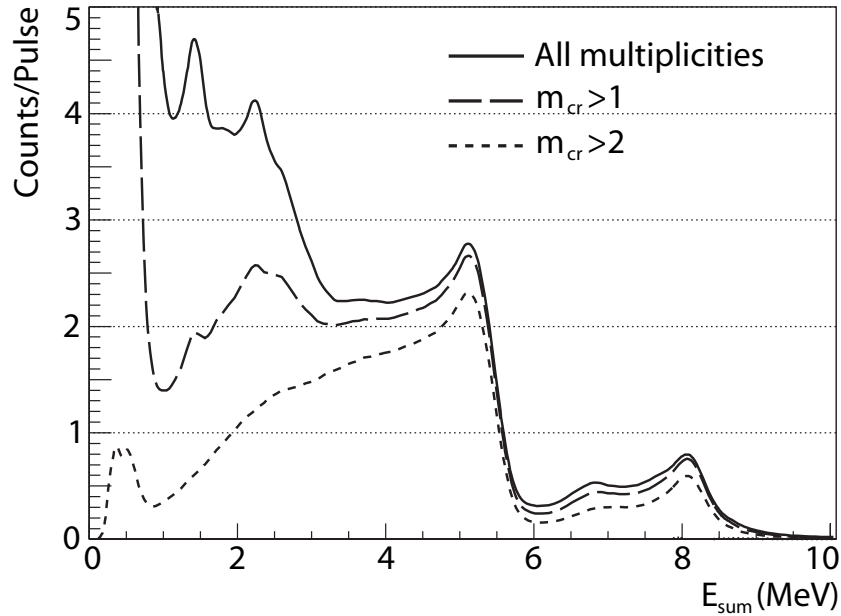


Figure 6.3: Distribution of deposited energy for the $^{237}\text{Np}(n,\gamma)$ measurement at neutron energies between 1 eV and 10 eV. The different lines correspond to different conditions on m_{cr} .

6.2.3 Calculation of the capture yield

The experimental capture yield of ^{237}Np is calculated as follows:

$$Y_{n,\gamma}^{obs} = \frac{C - B}{\varepsilon \cdot N \cdot \Phi_n}, \quad (\text{see Section 5.2.2.}) \quad (6.1)$$

where C and B are the total and background counting rates, ε is the efficiency of the detector, $\phi_n(E_n)$ is the intensity of incident neutrons recorded in the neutron monitor *SiMon*, and N is the normalization factor that relates the neutron intensity in the monitor with that of the neutrons impinging on the sample.

6.2.3.1 Determination of the detection efficiency

The detection efficiency of the TAC ε depends on the event selection conditions for E_{sum} and m_{cr} , and also on the counting rate because of the dead-time losses. The efficiency was calculated by means of a Monte Carlo simulation of the complete neutron capture measurement by: (i) producing the electromagnetic cascades following the (n,γ) reaction in ^{237}Np , (ii) generating the capture cascades with the experimental time-of-flight distribution, and (iii) calculating detector response to the generated cascades. The details and validation of the Monte Carlo code, the dead-time correction model and the capture cascade generator are given in Section 5.1.

Compared to the $^{197}\text{Au}(n,\gamma)$ measurement (see Figure 6.4), the counting rate of ^{237}Np is

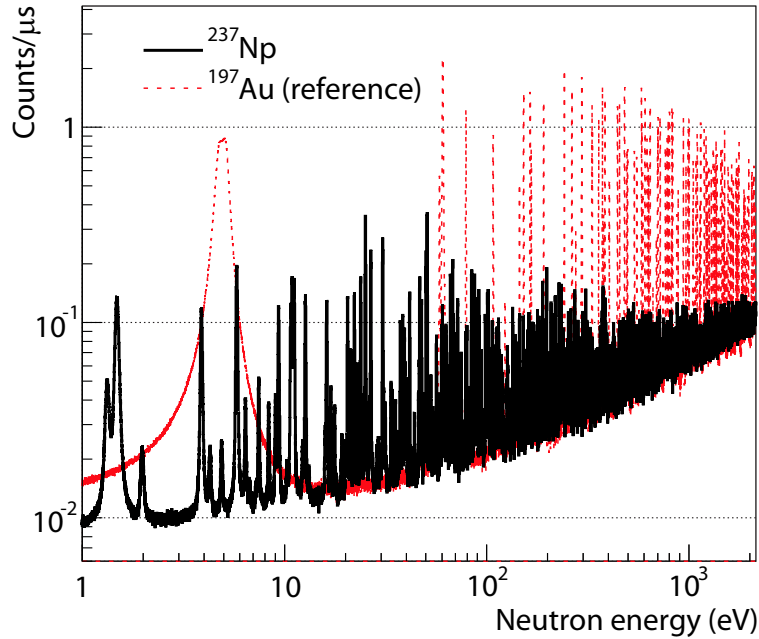


Figure 6.4: Counting rate recorded during the $^{237}\text{Np}(n,\gamma)$ measurement. The counting rate of the $^{197}\text{Au}(n,\gamma)$ measurement is shown for comparison, since the dead-time correction method was validated with this measurement in section 5.1.

much lower and therefore the effect of dead-time losses on the detection efficiency are expected to be lower than in the reference case of ^{197}Au . Indeed, it is found that the dead-time corrections are less than 1.5% at low neutron energies and completely negligible at high neutron energies.

The capture cascades for the MC simulation are produced from the available information for the nuclear level scheme and the transition probabilities up to a few hundred keV [115], and the statistical models of level density LD (Back Shifted Fermi Gas model) and transition probabilities (PSF combined with the Brink hypothesis) that describe the characteristics of the compound at high excitation energies.

The initial set of input parameters for the LD and PSF was obtained from RIPL-2 database [41]. However, it was necessary to adjust the parameters of the PSF so that the simulation was able to reproduce the experimental data. This adjustment required mainly to increase the intensity of M1 with respect to E1 transitions. A more detailed discussion of the investigations on Photon Strength Functions of actinides with the TAC is given in Ref. [119].

The simulated distributions of deposited energy and multiplicity are compared to the experimental data in Figure 6.5 at the example of the resonance at 1.32 eV. Simulated and experimental data are in excellent agreement. the simulation could be further validated by comparing the experimental and simulated distributions of deposited energy in the TAC for each value of m_{cr} . The latter distributions are also very sensitive to the characteristics of the capture cascade. The comparison between the simulated and experimental data showed some differences at low

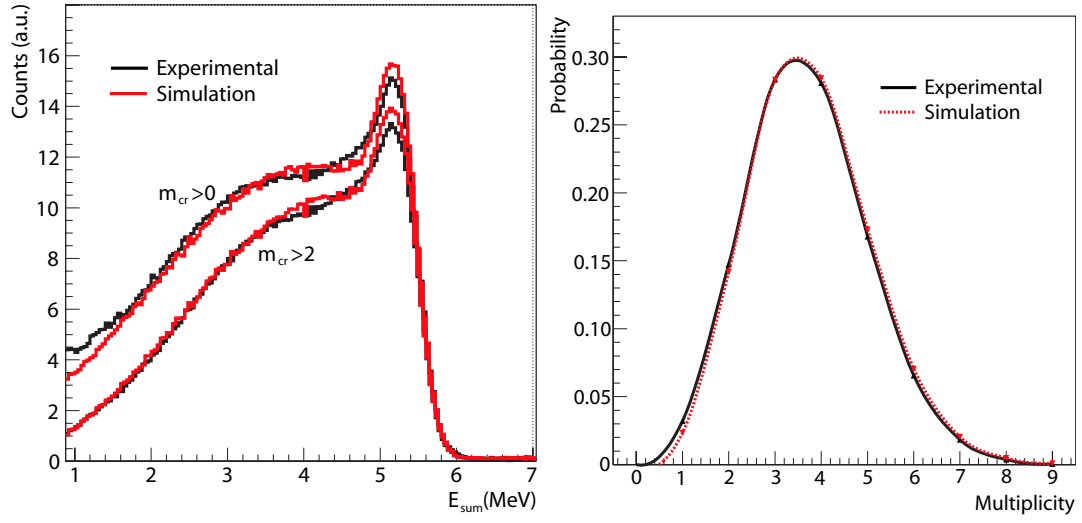


Figure 6.5: Comparison of the simulated distributions of deposited energy (left) and crystal multiplicity (right) with the experimental data.

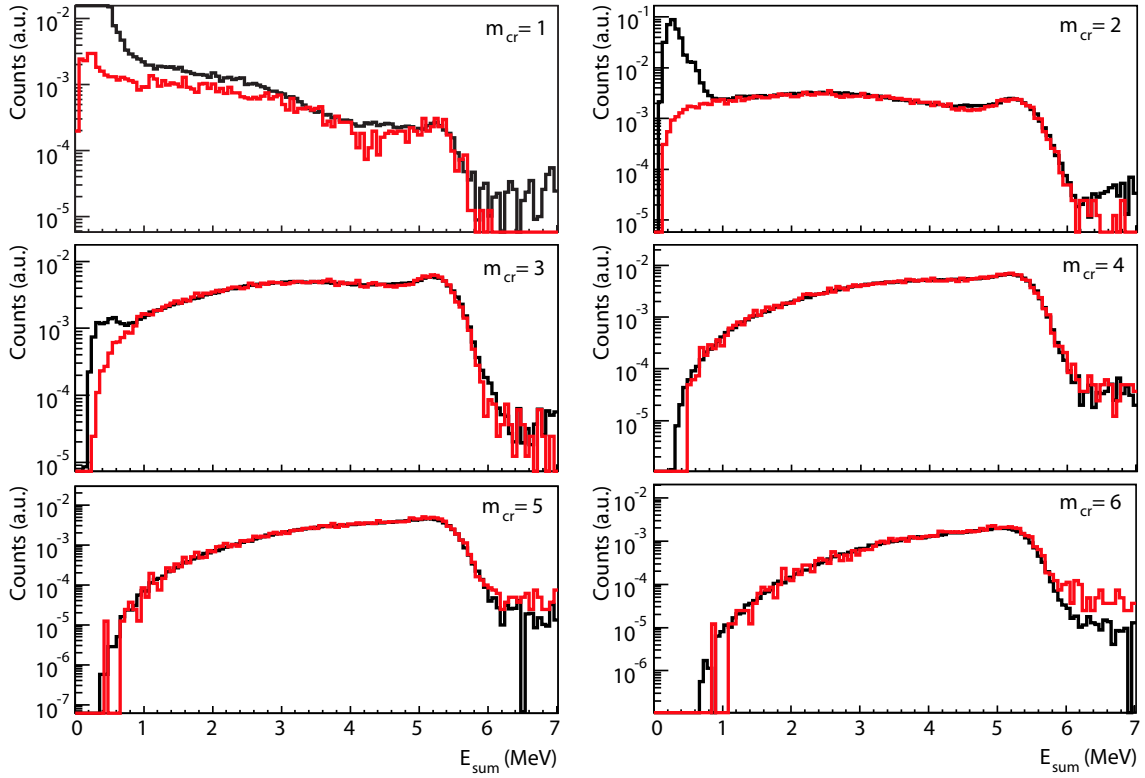


Figure 6.6: Comparison of the simulated and experimental distributions of deposited energy for different values of the crystal multiplicity.

energies and low multiplicities which are related to the difficulty of subtracting the background from the experimental data.

The detection efficiency was calculated for all combinations of E_{sum} and m_{cr} . For the specific conditions chosen for the analysis, $2.5 < E_{sum}(\text{MeV}) < 6$ and $m_{cr} > 2$, the calculated value was

$$\varepsilon = 0.70 \pm 0.02, \quad (6.2)$$

The uncertainty was estimated by performing a series of calculations with different but compatible sets of parameters for the PSF, a procedure that is discussed in detail in Section 5.1.6.

6.2.3.2 Normalization

The normalization factor needed for the calculation of the capture yield, which relates the neutron intensity measured by the neutron monitor *SiMon* with that of the neutrons impinging on the sample, has been determined in two different ways. The two normalization methods result in two different capture yields that were analyzed independently.

- In the first case, the capture yield is calculated by using the detection efficiency given by the MC simulations, the incident neutron intensity as monitored with the *SiMon* and the normalization factor obtained via the saturated resonance of ^{197}Au at 4.9 eV, see Section 5.2.2. It is assumed that the ^{237}Np sample has the same geometry and is placed at the same position as the ^{197}Au sample. In this way the capture yield is calculated without using any external reference such as capture cross sections at thermal energies or total cross section values from external measurements.
- In the second case, the experimental capture yield was analyzed in combination with the most reliable transmission data [132]. In this analysis, the normalization factor of the capture data is a free parameter that produces a capture yield compatible with the total cross section of the evaluations, which is assumed to be accurate within 3%. If accurate transmission data are available, as in this case, this method eliminates the systematic uncertainty associated with the calculated detection efficiency (2.5%). Since the thin target approximation is valid, the systematic uncertainty related to the sample mass (3%) is also eliminated.

In this work both methods have been used and two capture yields have been analyzed. However, the results presented in this chapter correspond to the analysis of the capture yield normalized to transmission because it is more accurate and, what is more important, because the combined analysis of capture and transmission data provide more accurate and less correlated resonance parameters.

6.2.3.3 Uncertainty in the capture yield

The sources of uncertainty associated with the calculation of the experimental capture yield are related to the calculation of the detection efficiency, the dead-time correction model, the possible misalignment of the sample with respect to the ^{197}Au reference measurement, the shape of the neutron fluence and the normalization factor. Since the analysis of the experimental capture yield is based in the calculation of a theoretical yield from resonance parameters, the sources of uncertainty in this calculation should also include the uncertainty of the isotopic mass of ^{237}Np in the sample.

Most of these components have been already discussed and correspond to uncertainties of 2.5% for the detection efficiency, 3% for the normalization via the ^{197}Au measurement, 3% for the sample mass, 2% for the energy dependence of the neutron fluence, and 3% for the transmission measurement [132]. The contribution from the calculation of dead-time losses is negligible.

Concerning the uncertainties related to the alignment of the ^{237}Np sample with respect to the ^{197}Au sample, it can be assumed that the maximum difference in the positioning of the samples in the TAC is below 1 mm. Such a displacement would affect the fraction of the beam intercepted by the sample, which has been calculated from the information on the beam profile presented in Section 3.1.2. Figure 6.7 shows the beam profile in a 2D histogram with the sample positioned in the centre. For a maximum misalignment of 0.5 mm, the uncertainty in the fraction of the beam intercepted by the sample is 1%.

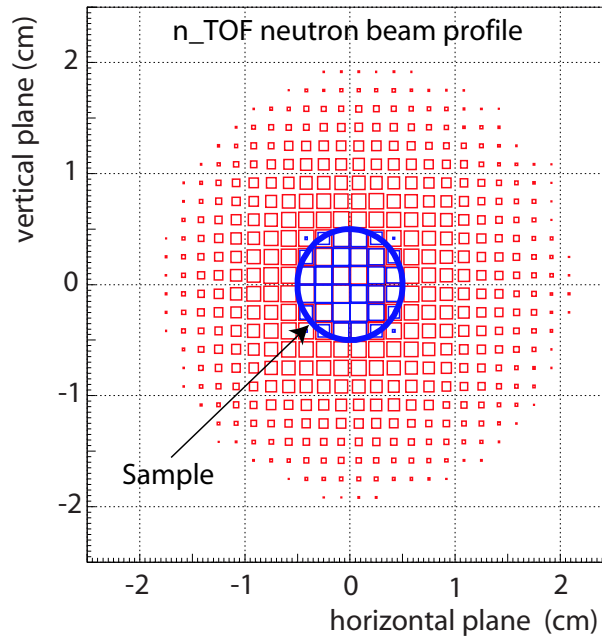


Figure 6.7: Calculation of the fraction of the beam intercepted by the sample from the information on the beam profile.

Analysis of the Resolved Resonance Region

The sources of uncertainty that are propagated to the capture yield using each of the normalization methods are different. These are summarized in Table 6.4, where the final uncertainty is calculated as the square root of the sum of squares of the partial uncertainties.

	Sources of uncertainty						Total
	Norm.	ε	Sample mass	Alignment	$\Phi_n(E_n)$	uncertainty	
Method 1	3%	2.5%	3%	1%	2%	< 5.4%	
Method 2	3%	-	-	-	2%	< 3.6%	

Table 6.4: *Partial and total uncertainties of the capture yield of ^{237}Np normalized to either the saturated resonance of ^{197}Au at 4.9 eV, or the total cross section of ^{237}Np from a reference transmission measurement.*

When the yield is normalized to transmission data, the systematic uncertainties related to scaling factors in the measurement (detection efficiency, mass of the sample and alignment) vanish. Both methods for calculating the capture yield provide an accuracy better than 5.4%, the second being of only 3.6%. The capture cross sections resulting from the analysis of the two yields have been found to be compatible within uncertainties, which validates the techniques developed for the determination of the different corrections and is a proof of the robustness of the analysis.

6.3 Analysis of the Resolved Resonance Region

6.3.1 Methodology for the resonance analysis

The capture yield and the transmission data in the Resolved Resonance Region (1 eV to 500 eV) were analyzed using the SAMMY code. The Reich-Moore approximation of the R -matrix formalism was adopted. The resonance parameters obtained after each SAMMY fit were used as initial values for a following fit until convergence was reached. In this specific context, convergence means that the resonance parameters do not vary significantly from one fit to the following and hence the χ^2 value remains stable. Starting from the resonance parameters in the evaluated data libraries, the convergence was usually reached after two or three iterations analyzing transmission and capture sequentially.

The transmission data included in the analysis correspond to the measurements of disk shaped sample 5 cm in diameter with a mass of 39 g that was irradiated at the 50 m time-of-flight station of the GELINA facility. Full details of the measurements are given in Refs. [132, 142]. The data were supplied by G. Noguere from CEA-Cadarache [143] and correspond to the measurement between 0.3 eV and 2 keV, which covers the complete energy range investigated in this work. The original flight times of the transmission data (50 m flight path) were re-

calibrated to the present capture data (185 m flight path) taking into account that the deviation $D(E)$ between two neutron energy scales corresponding to two different TOF experiments is given by [76]:

$$D(E) = aE + bE^{3/2}, \quad (6.3)$$

where the coefficient a is related to the flight path and b to the time-of-flight. The a and b parameters were obtained from the comparison of resonance energies fitted with SAMMY. The analysis of the resonances at lower energy gave $a=0.0002$ and $b\simeq 0$. These values are consistent with an uncertainty of the order of 1 cm in the flight path length of the measurement at GELINA.

The following items summarize the methodology used for the resonance analysis:

1. Thermal cross section and negative resonances.

Following the trend from recent investigations (see Section 6.1.2), the thermal cross section value $\sigma_{th}=181$ barns of the JEFF-2.2 evaluation was adopted in the present work. Since this choice affects the analysis of the first resonances, the low energy region was analyzed using the two sets of thermal cross sections presented in Table 6.2 and the results were compared.

2. Scattering radius.

The scattering radius R' of the R -matrix formalism is related to the potential scattering and can be expressed as a function of the scattering radius for each channel radius R'_J by:

$$\sigma_p(E) = 4\pi(g_{J=2}R'^2_{J=2} + g_{J=3}R'^2_{J=3}) = 4\pi R'^2, \quad (6.4)$$

where $g_{J=2,3}$ are the spin statistical factors.

The potential scattering dominates the total cross section in the valleys between resonances. Therefore the values of R'_J are extracted from a fit to the transmission data in the widest possible interval between resonances, which is located between 12 eV and 35 eV. The following values were obtained:

$$R'_{J=2} = 10.71 \pm 0.25 \text{ fm} \quad \text{and} \quad R'_{J=3} = 9.79 \pm 0.18 \text{ fm}, \quad (6.5)$$

and hence

$$R' = \sqrt{g_{J=2}R'^2_{J=2} + g_{J=3}R'^2_{J=3}} = 10.18 \pm 0.20 \text{ fm}. \quad (6.6)$$

The uncertainty in R' given by the fit does not account for other sources of uncertainty such as the normalization and the background, the sample thickness and the contribution of the oxygen to the potential scattering. These were investigated in Ref. [132] giving an overall uncertainty of 0.38 fm. Hence:

$$R' = 10.2 \pm 0.4 \text{ fm}. \quad (6.7)$$

3. Resonance energies.

Due to the small spacing between resonances, these are well resolved only up to a few tens of eV. At larger energies, the overlap becomes too large and it is not possible to separate neighbouring resonances and hence to assign their energies with an accuracy better than their width. Indeed, the resonant structures observed at high energies should be considered as pseudo-resonances.

4. Resonance widths.

The reaction channels that are open in the energy region under study are elastic scattering, neutron capture and fission.

Concerning the neutron and radiative widths, both are fitted simultaneously for all resonances at low energies ($E_n < 43$ eV) from the combined analysis of the capture yield and the transmission data. At larger energies, the determination of the radiative width becomes less accurate due to the overlap between resonances and hence the average value found from the resonances at low energies is assigned to all resonances. The constant value of the radiative width is predicted by the Gaussian Orthogonal Ensemble (GOE) discussed in Section 2.1.4.

The fission in ^{237}Np is subthreshold and therefore has a small effect on the capture yield and the transmission widths. For this reason the fission widths Γ_f were fixed to the values reported by Auchampaugh et al. [131], who proved that only one fission channel is needed for an accurate description of the subthreshold fission cross section of ^{237}Np .

5. Orbital momentum and spin of the resonances.

The statistical properties of the resonance parameters allow one to express the ratio of average neutron widths of s - and p -waves as a function of their level density, strength function and penetrability factors. With this information it is found [132] that the fraction of p -wave resonances below 500 eV should be lower than 0.001 and therefore all resonances are adopted as s -waves with the spins found in the evaluations, which were determined from measurements at low temperatures where the resonances are better resolved.

6. Background.

In the specific case of ^{237}Np , the neutron sensitivity component is negligible compared to the overall background, see Section 6.2.2 for details. The remaining background, which is smooth, was fitted using SAMMY. The fitted background, using three parameters, has the shape $B = a + b \cdot E_n^{-1/2} + c \cdot E_n^{1/2}$, with E_n expressed in eV units.

The full set of resonance parameters and the associated covariance matrix will soon be available in the EXFOR database. In the meantime, the resonance parameters resulting from this analysis are listed in the ENDF format [20] in Appendix B.

6.3.2 Resonance analysis below 43 eV

The cross section from thermal energies up to 43 eV is described by 75 resonances, including the negative resonance which accounts for the thermal cross section. The negative resonance and the lowest laying resonance at 0.49 eV are out of the energy range of this measurement and hence the parameters of the JEFF-2.2 evaluation were adopted.

The energy range between 1 eV and 43 eV was divided in two intervals due to the computational power required for the fit of such a large number of resonances. The result of the fits to the capture yield and the transmission data are shown in Figures 6.8 and 6.9. The points

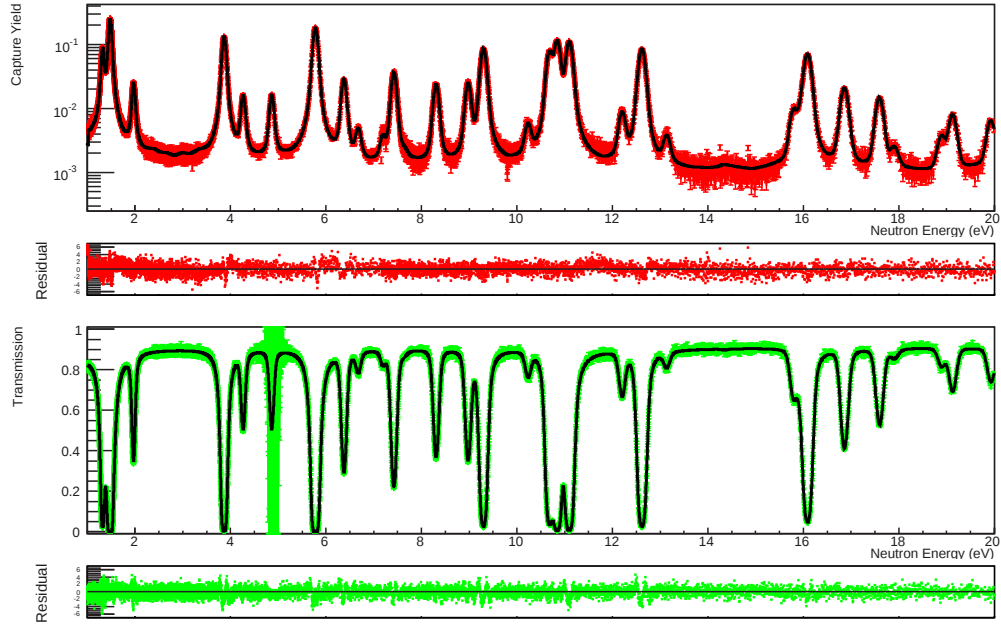


Figure 6.8: *SAMMY fits to capture (top) and transmission (bottom) in the energy range from 1 eV to 20 eV.*

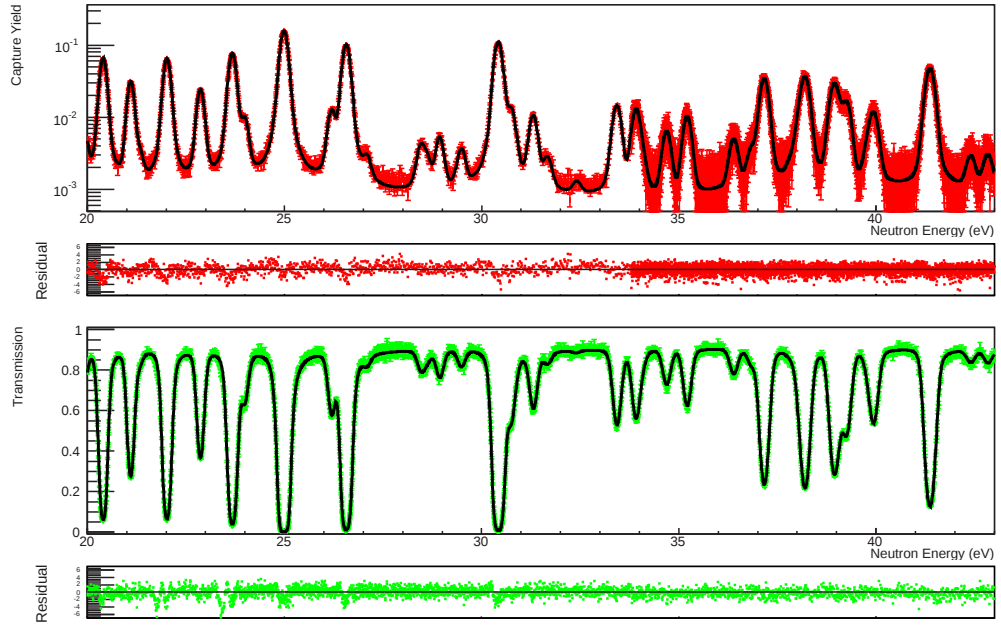


Figure 6.9: *SAMMY fits to capture (top) and transmission (bottom) in the energy range from 20 eV to 43 eV.*

correspond to the experimental data and the black lines to the yield and transmission calculated from the resonance parameters resulting from the SAMMY fits. The residual of the fit at each data point is shown in the small panels below the figures:

$$Residual = \frac{FIT_i - DATA_i}{\Delta_i}, \quad (6.8)$$

where Δ_i is the statistical uncertainty of each data point.

The most significant systematic uncertainties that affect the results in this low energy range are related to the modelling of the Thermal broadening of the resonances and the choice of the negative resonance accounting for the thermal cross section:

1. Thermal broadening.

In the present case of a crystalline sample the Free Gas Model (FGM) that was used in the analysis is only a good approximation for the description of the Doppler effects. Indeed, a previous work [144] identified as the cause for the small structures in the residuals of the lowest lying resonances (see Figure 6.8). Courcelle et al. [144] showed that the Crystal Lattice Model (CLM) should be more accurate than the FGM at low neutron energies. In the present work the CLM was tested but the results compared to those of the FGM were small below 7 eV and negligible at larger energies. The differences in the radiative and neutron widths using the FGM and the CLM were only of the order of 0.5% and 1.5%, respectively.

2. Choice of thermal cross section.

The ambiguity in the choice of the thermal cross section value (see Table 6.2) has a sizable effect in the resulting cross section, mainly at low energies. The analysis of this low energy region with the two values summarized in Table 6.2 lead to similar results in terms of quality of the fit (equivalent χ^2 values). Although the thermal cross section of JEFF-3.1 is 12% lower than that of JEFF-2.2, the resulting capture cross section using the value of JEFF-3.1 is only 2-3% larger than that using the negative resonances of JEFF-2.2.

In addition to the uncertainty of each resonance parameter, the full covariance matrices were generated with SAMMY. The correlation between resonance parameters is directly given by the analysis of microscopic cross sections and can be later propagated to the study of multi-group cross sections and group-average covariance matrices, which are important for nuclear applications [135].

Figure 6.10 shows the correlation matrix between resonance parameters corresponding to the 35 resonances below 20 eV. In this energy range there are a total of 107 parameters: three for each resonance (#1 to #105) and the last two corresponding to the normalization (#106) and constant background (#107) variables, respectively. The red lines in the figure indicate a distance of nine resonance parameters, which corresponds to three consecutive resonances. Within these three resonance distance limits there are clear correlations between the neighbouring resonances, especially those that are observed as clusters in Figure 6.8.

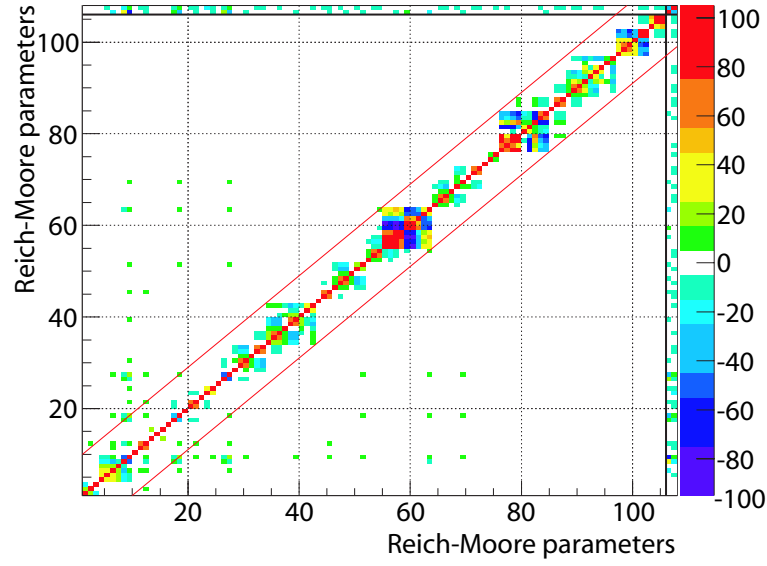


Figure 6.10: Correlation matrix for the first 35 resonances (E_n , Γ_γ and Γ_n for each resonance) including the negative resonance (first three parameters) and the normalization and constant background variables, represented by the last two parameters.

The accuracy in the determination of the radiative width is reduced as the energy increases and the overlap between resonances becomes larger. Hence the average radiative width $\langle\Gamma_\gamma\rangle$ has been calculated only below 43 eV. Figure 6.11 shows the radiation widths obtained from the

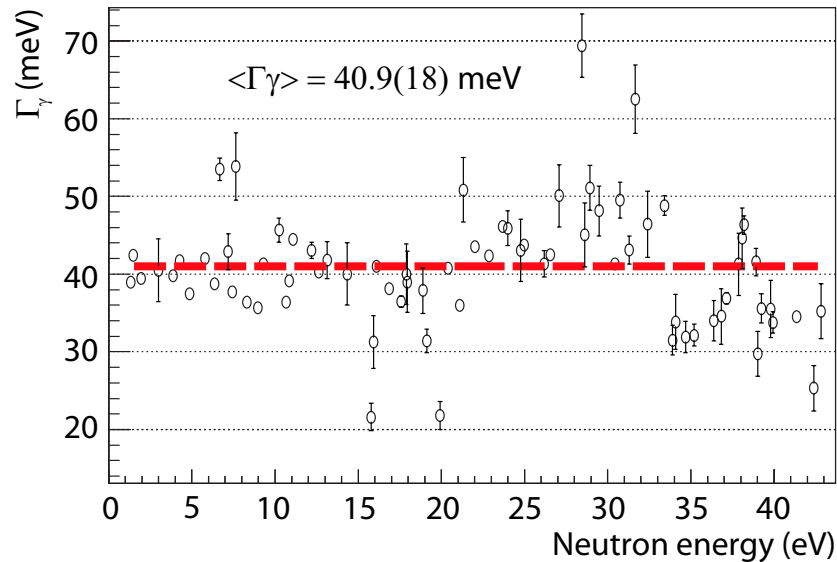


Figure 6.11: Fitted values for the radiative widths below 43 eV. The red-dashed line corresponds to the weighted mean value $\langle\Gamma_\gamma\rangle=40.9\pm1.8 \text{ meV}$.

analysis of all resonances in that energy range, where a minimum uncertainty of 1.5% related to the use of the FGM has been assigned to every value. The average value computed in the complete energy region is $\langle \Gamma_\gamma \rangle = 40.9 \pm 1.8$ meV. The significant deviations of the individual values from the average value are related mainly to the large correlation existing between the radiative and neutron widths. Such deviation does not indicate a non statistical behaviour of the radiative widths, as it is indicated by the fact that all the resonances at higher energies can be analyzed successfully using a fixed value corresponding to $\langle \Gamma_\gamma \rangle$ (see Section 6.3.3).

In the analysis at larger neutron energies, the radiative width is fixed to the average value obtained below 43 eV. Such a constant behaviour of the radiative width is expected from theory, see Section 2.1.4, and has been confirmed by the study of the low energy region. The weighted average radiation width $\langle \Gamma_\gamma \rangle$ has been computed in reduced energy intervals. The values are given in Table 6.5, where the variations in the local values of $\langle \Gamma_\gamma \rangle$ are within the expected 1.8 meV uncertainty. An exception is found only in the highest energy interval, where the overlap between resonances starts to be larger.

E_n (eV)	# Resonances	$\langle \Gamma_\gamma \rangle$ (meV)
1 - 7	10	41.0 $\pm$ 1.5
7 - 13	12	40.2 $\pm$ 2.6
13 - 19	10	39.4 $\pm$ 2.9
19 - 25	11	42.8 $\pm$ 2.8
25 - 31	9	42.2 $\pm$ 1.9
31 - 37	10	39.3 $\pm$ 8.1
37 - 43	12	36.1 $\pm$ 3.7
1 - 43	74	40.9$\pm$1.8

Table 6.5: *Weighted mean value of the radiation width in different neutron energy intervals below 43 eV.*

6.3.3 Resonance analysis between 43 eV and 500 eV

The complete interval between 43 and 500 eV could not be analyzed in a single fit due the very large number of resonances and to the size of the associated covariance matrix. For this reason, the analysis was subdivided in ten smaller intervals. The analysis of small energy intervals reduces computing time and, more important, allows one to test the consistency of the correction parameters (normalization and background) as a function of neutron energy.

In the analysis it is considered that all the radiative widths have a constant value of $\langle \Gamma_\gamma \rangle = 40.9 \pm 1.8$ meV. The number of pseudo-resonances analyzed is from the literature. The energy and the neutron width of all resonances are fitted leaving also the normalization and background as free parameters. The energy intervals 120 to 140 eV and 310 to 370 eV are analyzed from the capture yield only because in these regions several black filters were used to determine the background in the transmission data. In these two intervals the normalization

was fixed to the value interpolated from the results of the analysis of the neighbouring energy intervals.

Figures 6.12 to 6.20 display the experimental capture yield and the transmission data between 43 eV and 500 eV together with the SAMMY fits and the corresponding residuals. The capture and transmission data are compatible in the complete energy range because they can be reproduced from the same set of resonance parameters. Moreover, the result validates the use of a fixed value for the radiative width of all resonances.

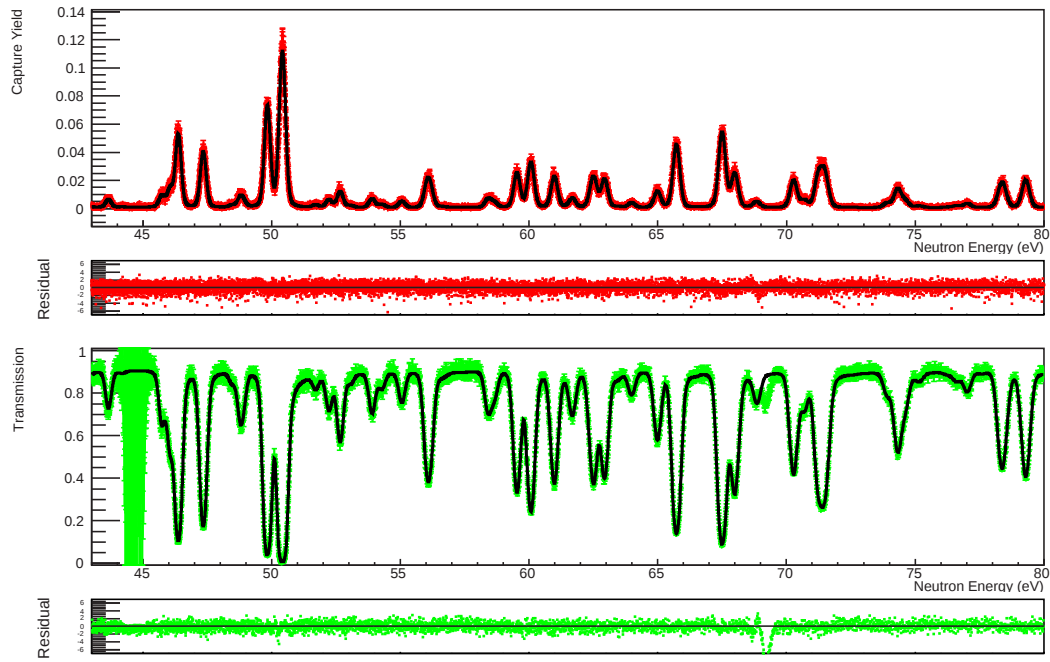


Figure 6.12: *SAMMY fits to the capture and transmission data between 43 and 80 eV.*

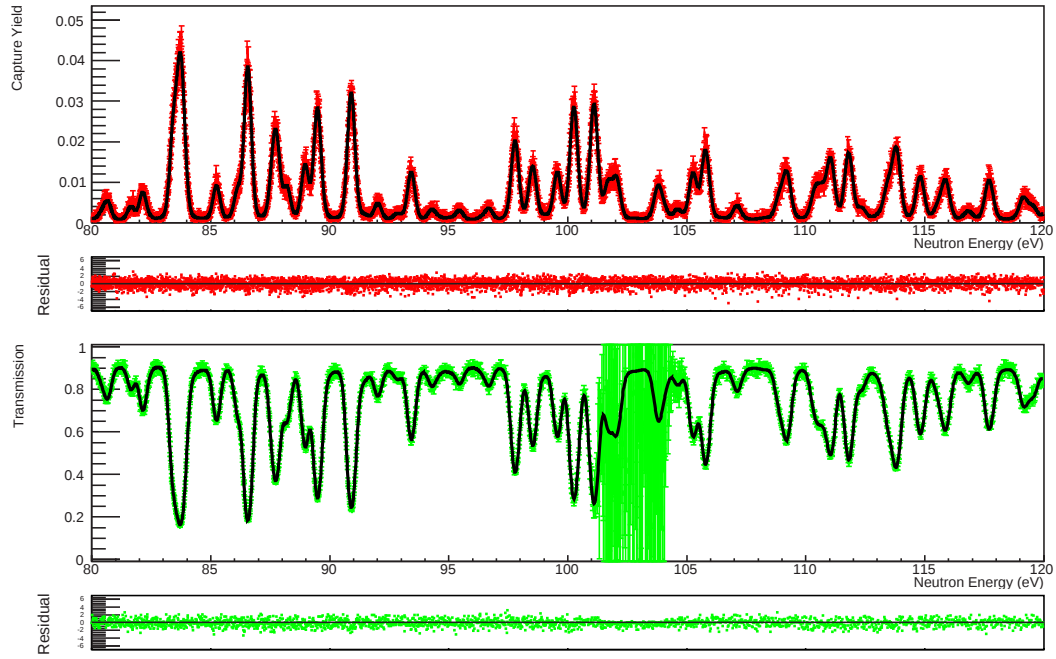


Figure 6.13: *SAMMY fits to the capture and transmission data between 80 and 120 eV.*

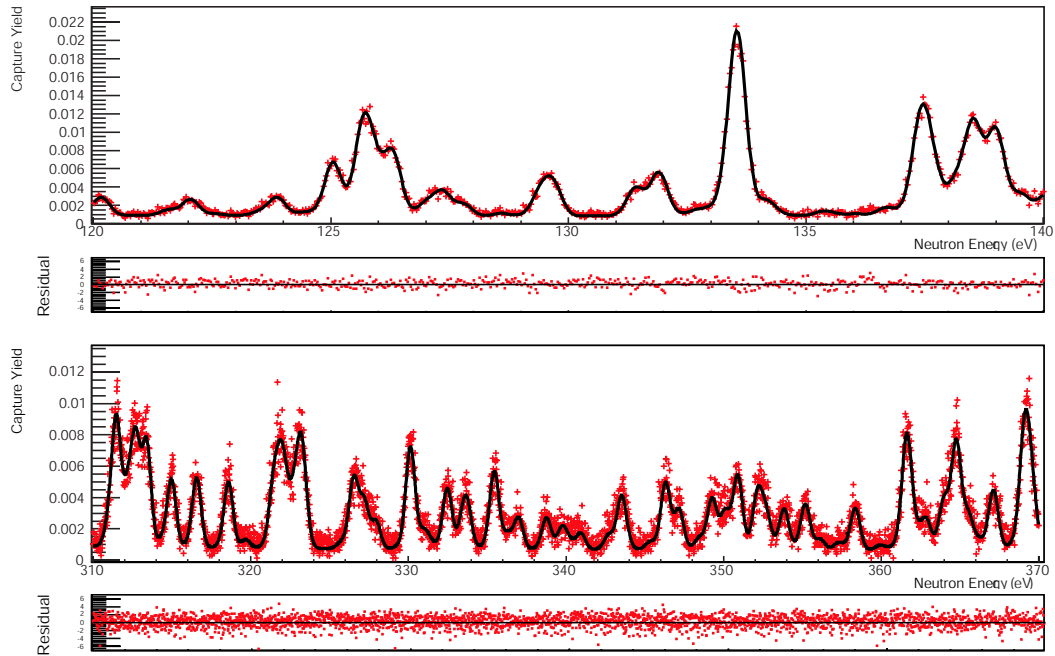


Figure 6.14: *SAMMY fits to the capture data in the intervals 120-140 eV and 310-370 eV.*

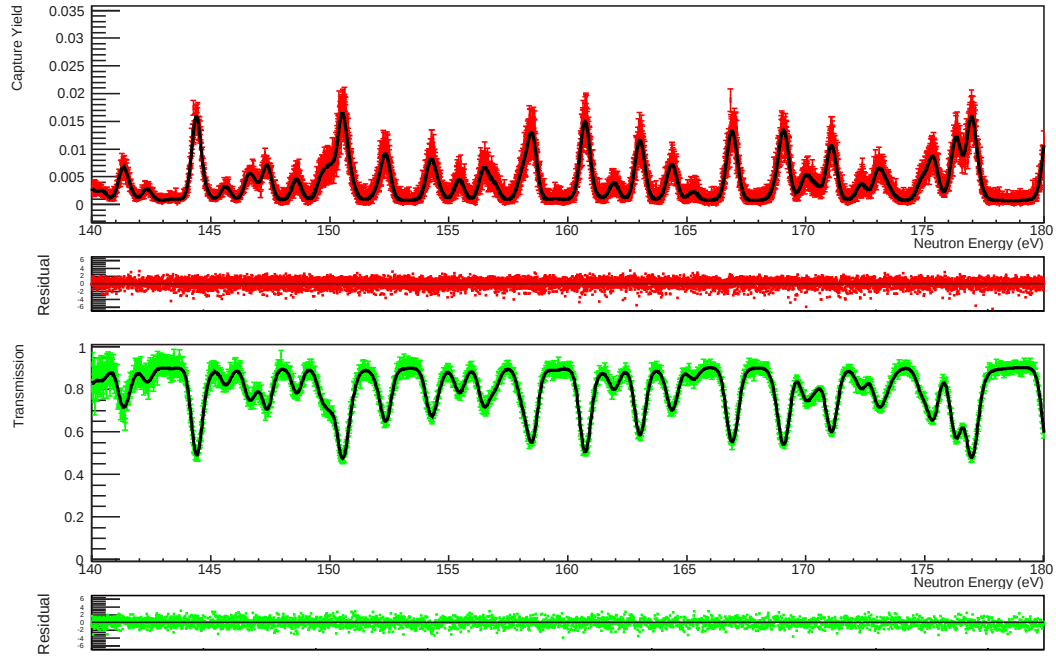


Figure 6.15: *SAMMY fits to the capture and transmission data between 140 and 180 eV.*

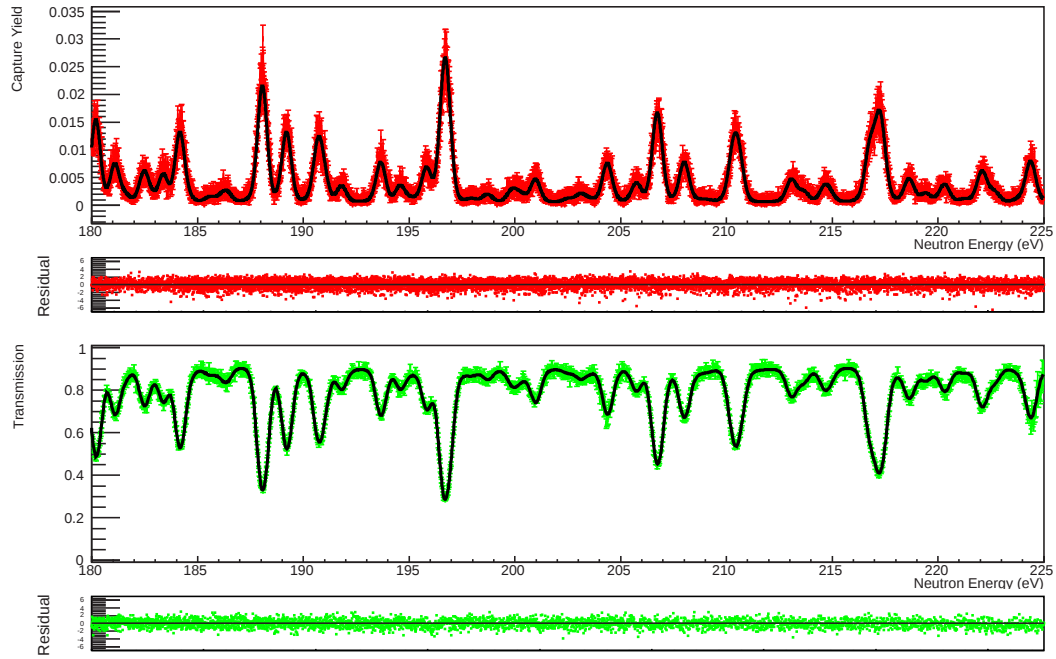


Figure 6.16: *SAMMY fits to the capture and transmission data between 180 and 225 eV.*

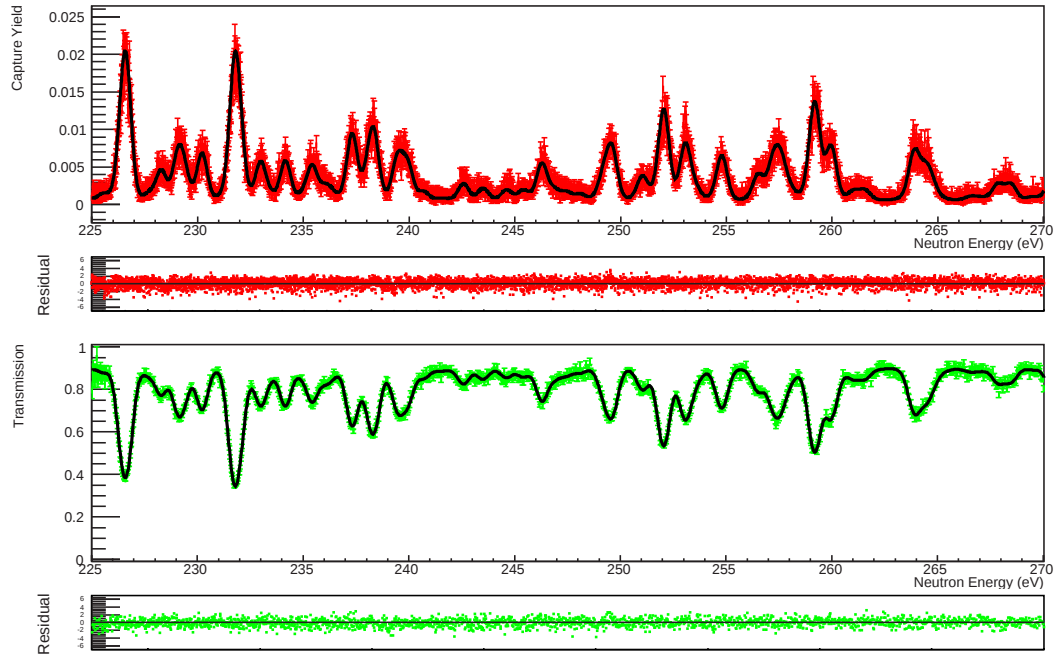


Figure 6.17: *SAMMY* fits to the capture and transmission data between 225 and 270 eV.

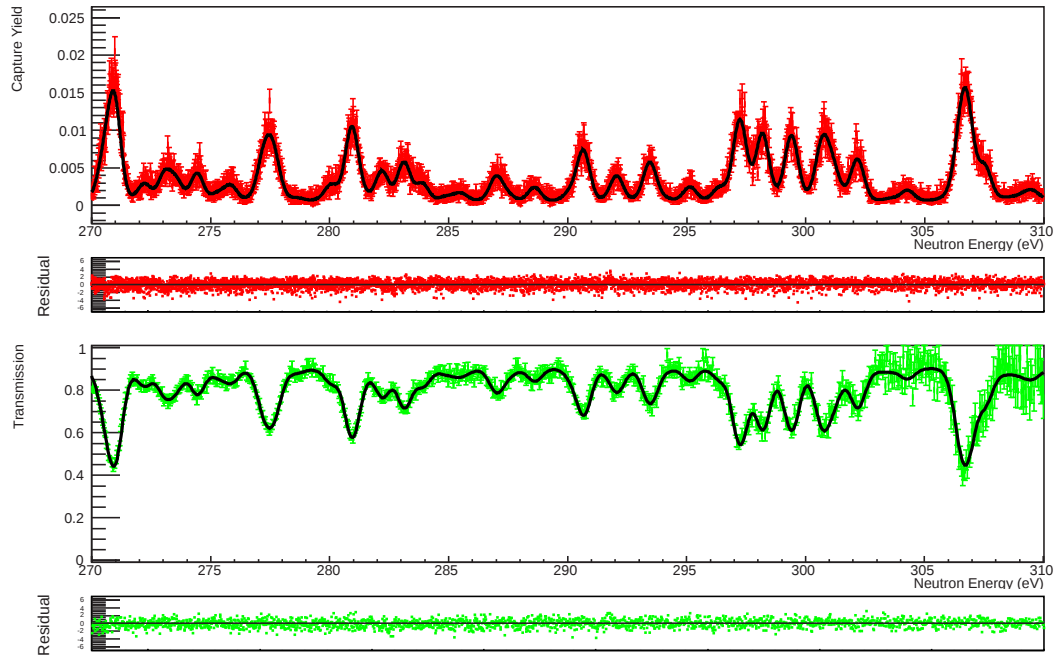


Figure 6.18: *SAMMY* fits to the capture and transmission data between 270 and 310 eV.

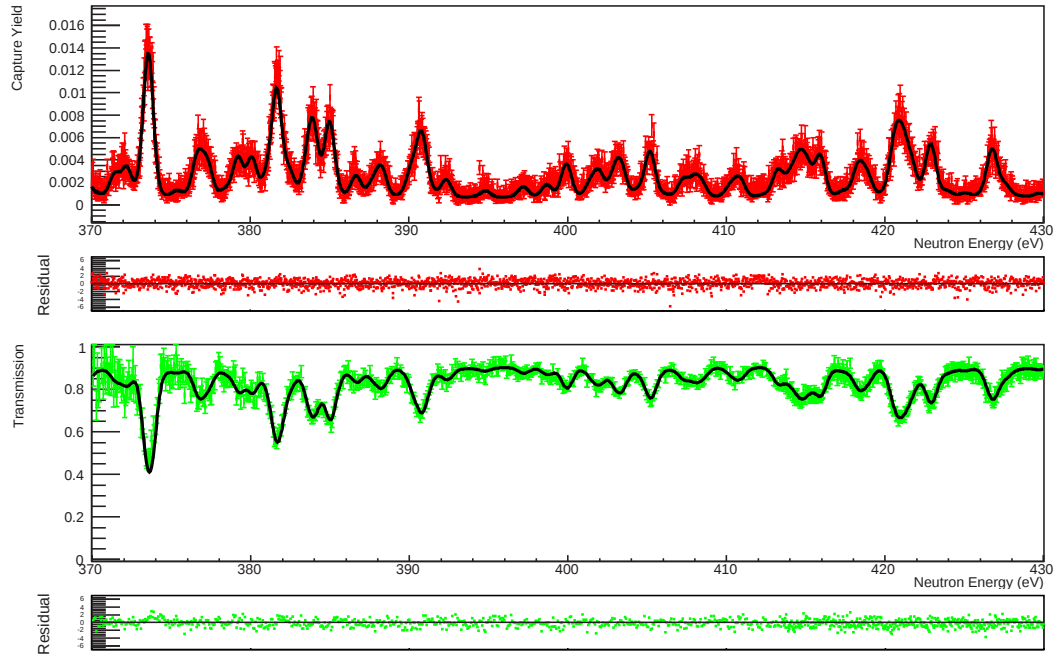


Figure 6.19: *SAMMY* fits to the capture and transmission data between 370 and 430 eV.

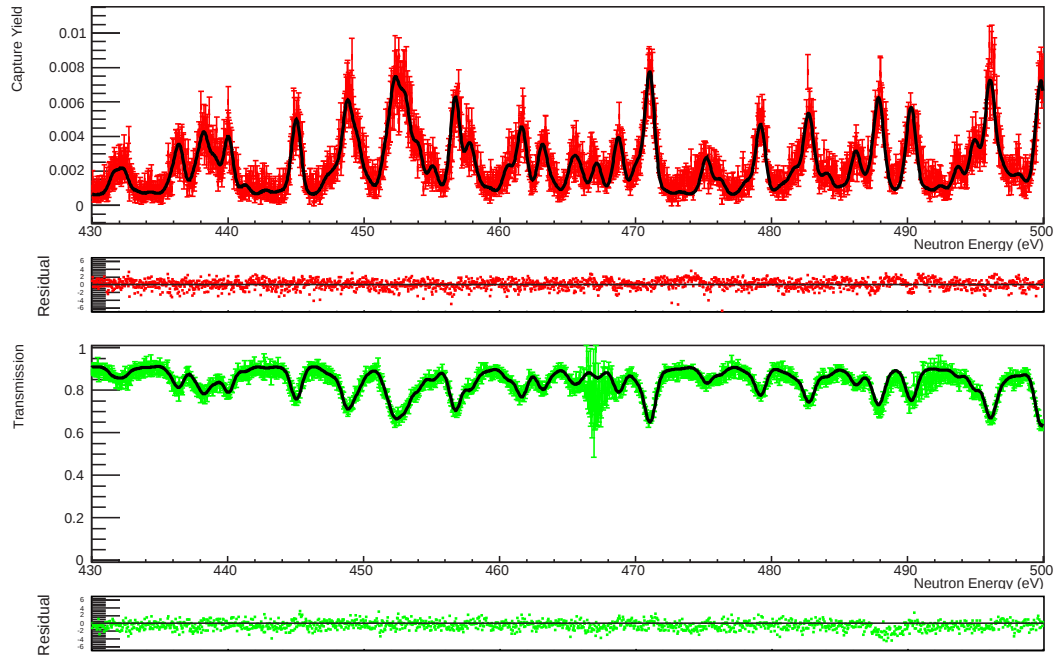


Figure 6.20: *SAMMY* fits to the capture and transmission data between 430 and 500 eV.

The analysis of each of the ten energy intervals provides an independent value of the normalization factor and the background level. Hence, it is important to confirm that all the values are consistent:

- Although the background is described by a function of the type $B = a + bE^{1/2} + cE^{-1/2}$, the fits provide a nearly constant value in each energy interval. As shown in Table 6.6 the background level decreases with energy from $3 \cdot 10^{-4}$ at 1 eV to $8 \cdot 10^{-5}$ at 500 eV, corresponding to less than 1% of the capture yield in the resonances and up to 10% in the valleys.

$E_n(\text{eV})$	Background	$E_n(\text{eV})$	Background
1 - 20	0.00031	140 - 225	0.00015
20 - 43	0.00029	225 - 310	0.00011
43 - 80	0.00021	310 - 370	0.00010
80 - 120	0.00018	370 - 430	0.00010
120 - 140	0.00016	430 - 500	0.00008

Table 6.6: *Background level, in units of yield, fitted between 1 eV and 500 eV. The uncertainty in the background given by SAMMY is lower than a unit in the last digit.*

- The normalization factors obtained in the different energy intervals are shown in Figure 6.21. The constant behaviour of the normalization, with the only exception of the value between 370 eV and 430 eV, indicates that the capture and transmission data are compatible at all energies. The reasons behind the large deviation around 400 eV does not seem to be related to large fission or scattering resonances.

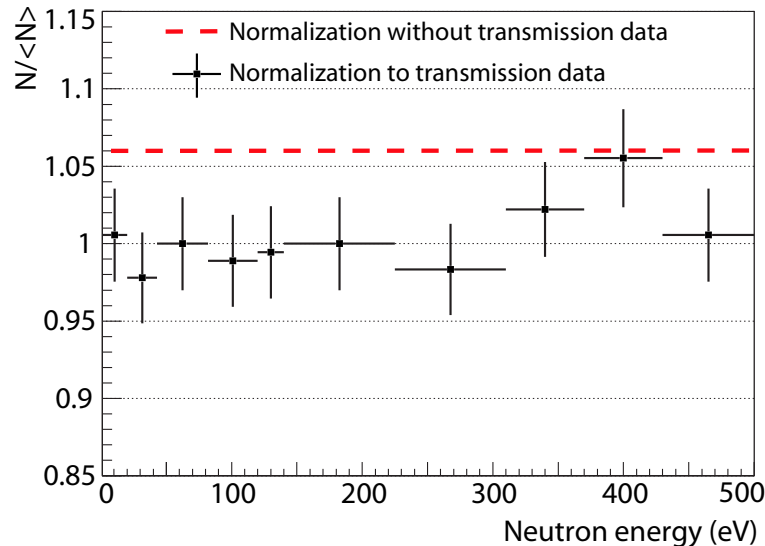


Figure 6.21: *Variation of the normalization around the mean value in different energy intervals.*

The normalization found by applying the Saturated Resonance Method to the 4.9 eV resonance in ^{197}Au is 6% larger than the values determined via de transmission (dashed line in Figure 6.21). This difference is at the limit but still within the uncertainty expected from the combination of the normalization to ^{197}Au , the mass of the sample and the detection efficiency. This is again a proof of the consistency of the different corrections that have been applied and also of all the systematic uncertainties that have been estimated.

6.3.4 Statistical properties of the resonance parameters

The investigation of the statistical properties of the resonance parameters calculated in the RRR is necessary for two reasons: (i) it gives information about the consistency of the resonance parameters, (ii) the average resonance parameters are the basis of the high energy neutron cross sections calculations using the Hauser-Feshbach Statistical Model and the Optical Potential Model.

This section presents the analysis of the statistical properties of the resonance parameters and the comparison of the results with the evaluations and previous measurements.

6.3.4.1 Average level spacing and probability distribution

The level spacing $\langle D_0 \rangle$ is defined as the mean distance between next neighbouring resonances. It is calculated from the slope of the cumulative distribution of the number of resonances as a function of neutron energy, which is shown in Figure 6.22. The expected linear behaviour is only followed up to an energy of ~ 120 eV, indicating that resonances start to overlap significantly at higher energies and hence may not be properly identified.

The slope of the linear fit to the cumulative distribution in Figure 6.22 corresponds to the value:

$$\langle D_0 \rangle = 0.56 \pm 0.02 \text{ eV}, \quad \text{where} \quad \frac{\Delta \langle D \rangle}{\langle D \rangle} = \sqrt{\frac{1}{N} \left(\frac{4}{\pi} - 1 \right)}. \quad (6.9)$$

This result is compatible within uncertainties with previously reported values summarized in Table 6.7.

	This work	RIPL-2	Mughabghab	JEFF-3.1	JEF-2.2
$\langle D_0 \rangle$ (eV)	0.56 ± 0.02	0.57 ± 0.03	0.052 ± 0.04	0.58	0.56

Table 6.7: *Average level spacing of this work and previous evaluations.*

The average level spacing can also be calculated for resonances of the same spin from the partial cumulative distributions shown in Figure 6.23. The values from the linear fits give

$\langle D_0^{J=2} \rangle = 1.23 \pm 0.09$ eV and $\langle D_0^{J=3} \rangle = 1.025 \pm 0.07$ eV. The nuclear level density is calculated as the inverse of the average level spacing, resulting in the values give in Table 6.8.

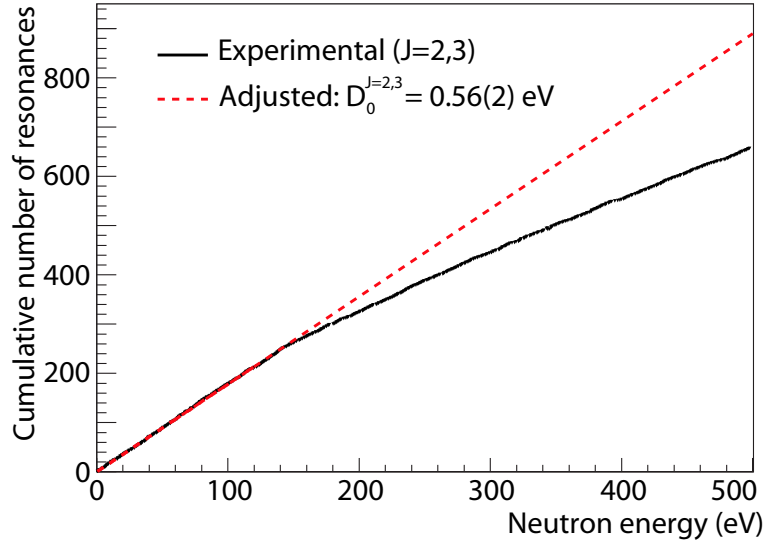


Figure 6.22: Cumulative number of observed levels (black). The red line corresponds to a linear fit in the low energy region.

	$\langle D_0^J \rangle$ (eV)	ρ_0^J (eV $^{-1}$)
J=2	1.23 ± 0.09	0.82 ± 0.06
J=3	1.025 ± 0.07	0.97 ± 0.06
J=2,3	0.56 ± 0.02	1.79 ± 0.09

Table 6.8: Level density calculated from the *n*-TOF resonance parameters.

An important test for the distribution of neighbouring levels is provided by the comparison of the experimental distribution and the expected Wigner distribution (see Section 2.1.4). For resonances with the same total angular momentum and parity with an average level spacing $\langle D_0 \rangle$, the Wigner distribution becomes

$$p(x)dx = \frac{\pi x}{2} \exp\left(-\frac{\pi x^2}{4}\right) dx \quad \text{with } x = \frac{D_i}{\langle D_0 \rangle}. \quad (6.10)$$

Using the average level spacing in Table 6.8, the Wigner distribution has been calculated and compared to the measured distribution for each spin group. The results are shown in Figure 6.24. Although the distributions of both spin groups follow reasonably well the Wigner shapes, the agreement for J=3 resonances is better than for J=2. The difference could be related either to the fact that *J*=2 resonances are weaker than those with *J*=3 and therefore more difficult to identify, or to an incorrect spin assignment of the weaker resonances. A capture measurement at low temperature could help to identify and solve the problem.

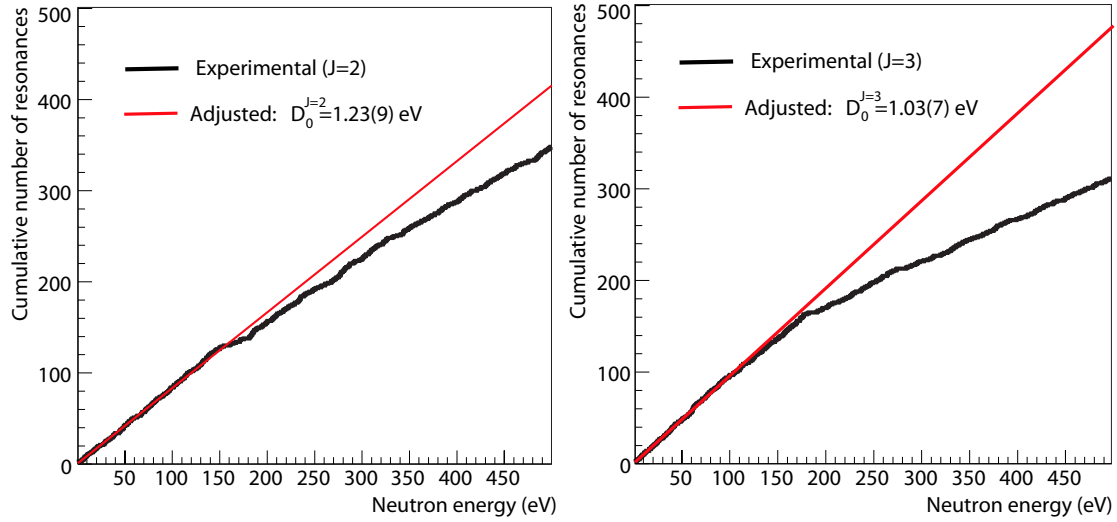


Figure 6.23: Measured cumulative number of levels for each spin and the corresponding linear fit.

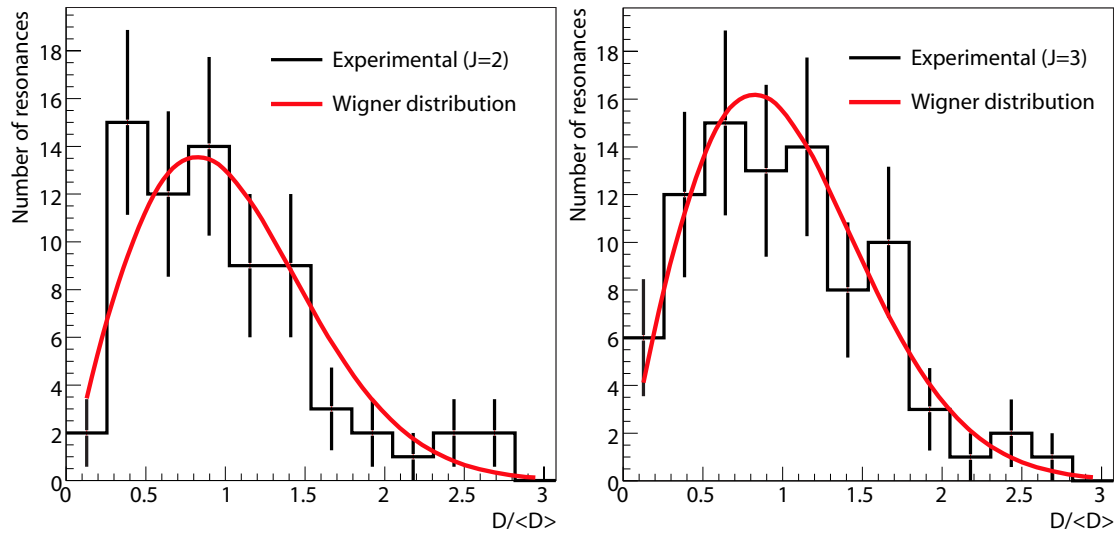


Figure 6.24: Level spacing distribution of the n -TOF resonance parameters below 90 eV compared to the expected Wigner distribution.

6.3.4.2 Radiative width

The average radiation width has been determined from 74 resonances in the energy range below 43 eV. The average value $\langle \Gamma_\gamma \rangle = 40.9 \pm 1.8$ meV is in good agreement with previous evaluations (Table 6.9). The smaller uncertainties quoted in the RIPL-2 and the Mughabghab compilation are not realistic since the effect of using the Free Gas Model instead of the Crystal

Lattice Model for the modelling of the Doppler broadening was not considered.

	This work	RIPL-2	Mughabghab[121]	JEFF-3.1	JEF-2.2
$\langle \Gamma_\gamma \rangle$ (meV)	40.9 ± 1.8	40.8 ± 1.2	40.8 ± 1.0	40.0	40.0

Table 6.9: Average radiation width of this work and previous evaluations.

6.3.4.3 Neutron strength function

The neutron strength function S_0 is related to the average total cross section and is defined as the ratio of the average reduced width and the level spacing. The work of Feshbach, Porter and Weisskopf in 1954 [145] was crucial in the systematic investigation of the values of S_0 as a function of the atomic mass. The prediction for minor actinides with $A \simeq 240$ is a value of $S_0 \simeq 10^{-4}$.

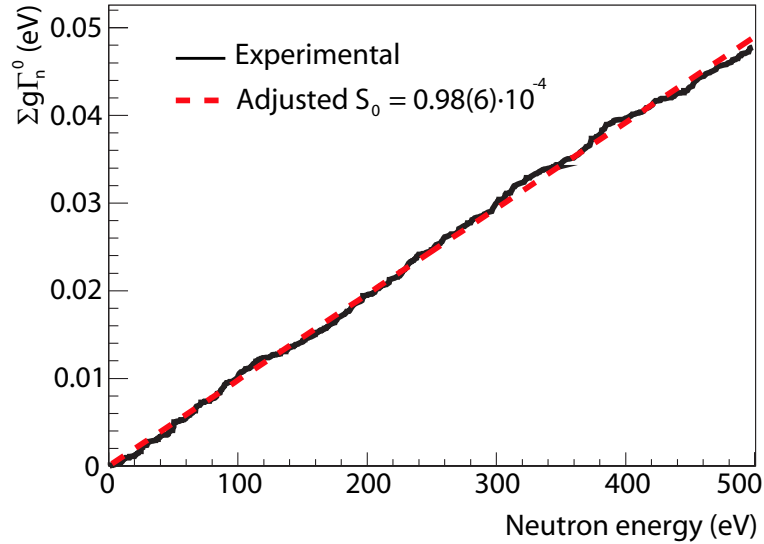


Figure 6.25: Cumulative distribution of the reduced width of ^{237}Np from its resonance parameters. The experimental value of the strength function S_0 corresponds to the slope of the linear fit (red dotted line).

The strength function can be calculated from experimental information,

$$S_0 = \frac{\langle g\Gamma_n^0 \rangle}{\langle D_0^{J=2,3} \rangle} = \frac{\Sigma_E^{E+\Delta E} g\Gamma_n^0}{\Delta E} \quad (6.11)$$

where the reduced width Γ_n^0 is defined, in the case of s -wave resonances, as $\Gamma_n^0 = \Gamma_n / \sqrt{E}$. This means that S_0 can be calculated from the slope of the cumulative distribution of $g\Gamma_n^0$ as a

function of neutron energy. This distribution is shown in black in Figure 6.25 with the red line corresponding to a linear fit that yields:

$$S_0 = (0.98 \pm 0.06) \cdot 10^{-4}. \quad (6.12)$$

The strength function has been calculated in the complete energy range. Even though a significant number of resonances is missing above 100 eV, the calculation is still valid because the strength function is an average quantity related to the total area of the resonances which, even in the case of resonance multiplets, does not depend on the number of resonances used. In order to confirm this assumption, the strength function has been calculated in several energy intervals. The results listed in Table 6.10 exhibit only variations of a few percent, compatible within uncertainties with the statistical uncertainty related to the number of levels included in the calculation.

E (eV)	$N_{measured}$	$S_0(10^{-4})$
0-100	177	0.962 ± 0.11
100-200	145	0.969 ± 0.13
200-300	120	0.989 ± 0.14
300-400	109	0.999 ± 0.14
400-500	106	0.968 ± 0.15
1-500	657	0.98 ± 0.06

Table 6.10: *Strength function calculated for different energy intervals.*

The average value $S_0(10^{-4}) = 0.98 \pm 0.06$ is in good agreement with the values reported that are summarized in Table 6.11.

	This work	RIPL-2	Mughabghab	JEFF-3.1	JEF-2.2
$S_0 (10^{-4})$	0.98 ± 0.06	0.97 ± 0.07	1.02 ± 0.06	1.0	0.99

Table 6.11: *Values of the strength function (s-wave) calculated in this work and given in the different evaluations.*

6.3.4.4 Neutron width distributions

The reduced neutron widths for a single J value are expected to follow the Porter-Thomas (P-T) distribution, i.e. a χ^2 distribution with only one degree of freedom ($\mu=1$):

$$P_{PT}(x) = \frac{1}{\sqrt{2\pi x}} e^{-x/2}, \quad (6.13)$$

with $x = \Gamma_n^0 / \langle \Gamma_n^0 \rangle$. However, the comparison between the experimental data and the expected P-T distribution is usually illustrated from the integral of the P-T distribution as a function of x_i :

$$N(x_i) = N_0 \int_{x_i}^{\infty} P_{PT}(x) dx = N_0 (1 - \text{erf} \sqrt{x_i/2}) \quad (6.14)$$

where N_0 is the total number of resonances.

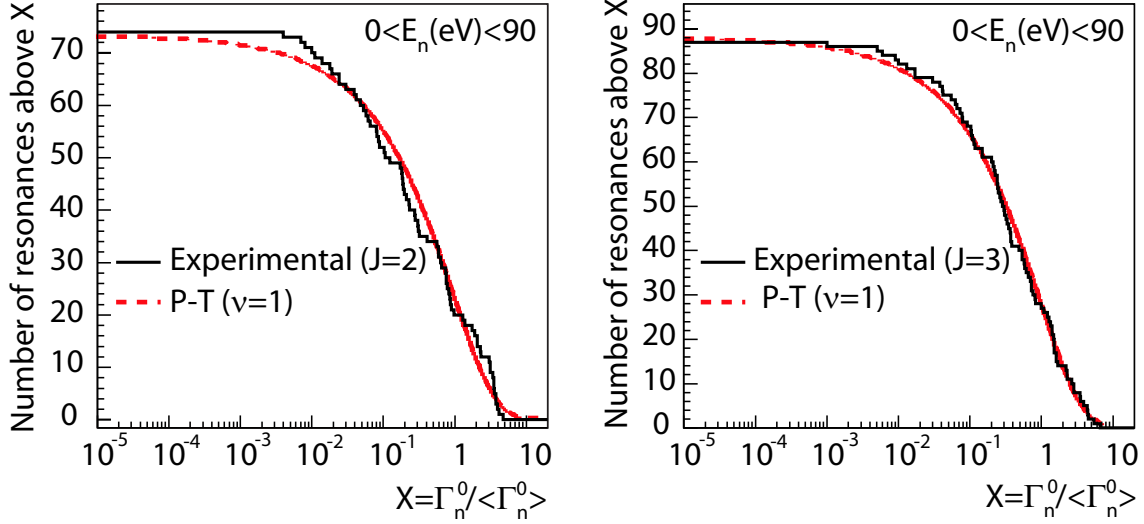


Figure 6.26: Comparison between the experimental and expected integral distributions of the neutron widths.

Since resonances start to be missed above ~ 90 eV, the distribution of neutron widths has been studied only in the interval between 0 and 90 eV. Figure 6.26 shows the experimental and calculated number of resonances with $\Gamma_n^0 > x_i \langle \Gamma_n^0 \rangle$ as a function of x_i . The number of resonances N_0^J has been calculated from the partial level spacing values summarized in Table 6.8.

It is observed that, for both J=2 and J=3 resonances, the experimental data is in excellent agreement with the expected behaviour. This result confirms the consistency of the resonance parameters in the energy region of small resonance overlap.

6.4 Analysis of the Unresolved Resonance Region

Above 500 eV the resonant structures are still visible but the degree of overlap is too large for a meaningful fit. Therefore the energy region between 500 eV and 2 keV was analyzed using the formalism for the URR with capture and elastic scattering as open reaction channels. The contribution of sub-threshold fission to the total cross section is below 0.1% and does not play a significant role in the analysis of average capture and transmission data and has been neglected.

In the URR it is more suitable to analyze the average cross sections instead of indirect quantities such as reaction yields or transmission coefficients. The average cross sections have been analyzed using the implementation of the FITACS code [146], which is based on Hauser-Feshbach calculations with width fluctuations [147], into SAMMY.

6.4.1 Averaging the cross sections

The capture yield is related to the capture cross section and the target thickness n by

$$\langle Y(E_n) \rangle = f(E_n)n\langle\sigma_\gamma(E_n)\rangle \quad (6.15)$$

where the thin target approximation has been assumed. The factor $f(E_n)$ accounts for self-shielding and multiple scattering effects and has been calculated with the SESH code [148]. SESH generates individual resonances according to user-supplied level densities and neutron strength functions and takes the sample geometry into account. The results have shown that the size of the corrections is smaller than 1% in the complete energy range, which was expected since the self shielding factor $(1-e^{-n\sigma_t})$ is at maximum 0.6%. Therefore, the average capture cross section is directly calculated from the measured yield after the subtraction of the point-wise background using equation 6.15. Since the normalization of the capture yield has been shown to be accurate and consistent in the RRR, the same normalization was applied in the URR.

The transmission is related to the total cross section by

$$\langle T \rangle = \langle e^{-n\sigma_{tot}} \rangle = e^{-n\langle\sigma_{tot}\rangle} \langle e^{-n(\sigma_{tot}-\langle\sigma_{tot}\rangle)} \rangle \quad (6.16)$$

where all but n are energy dependent quantities. The second term on the right-hand side of this equation is a correction term due to fluctuations of the cross section and is calculated using the SESH code, resulting again to be lower than 1%. It is important to subtract the contribution of the oxygen in the NpO_2 sample (0.0104 at/barn) from the measured transmission data because the ^{16}O and ^{237}Np total cross sections are of the same order of magnitude, which is not the case in the capture channel.

6.4.2 Analysis with SAMMY/FITACS

The parameters that can be adjusted in the SAMMY/FITACS code for each partial wave ℓ are the neutron strength function S_ℓ , the level spacing D_ℓ , the average radiation width $\langle\Gamma_\gamma\rangle_\ell$, and the distant level parameter R_ℓ^∞ , calculated from the potential scattering radius as $R' = a \cdot (1 - R^\infty)$ where a is the nuclear radius: $a=1.23A^{1/3}+0.8$ fm. In this energy range it is sufficient to include only the first two partial waves, s and p .

The initial values for the s -wave parameters and their uncertainties are those found in the RRR, that is $S_0=(0.98\pm0.06)\cdot10^{-4}$, $\langle\Gamma_\gamma\rangle_0=40.9\pm1.8$ meV and $R_0^\infty=-0.249\pm0.012$ fm, corresponding to $R'=10.2\pm0.4$ fm.

The p -wave parameters are fixed to:

- $S_1=2 \cdot 10^{-4}$ [132, 121];
- D_1 is scaled to D_0 by the number of allowed spin states: $D_1=D_0/2$, assuming a $(2J+1)$ spin dependence of the level density;
- $\langle \Gamma_\gamma \rangle_1 = \langle \Gamma_\gamma \rangle_0 = 40.9$ meV.

In principle the capture cross section is more sensitive to $\langle \Gamma_\gamma^0 \rangle$ while the total cross section is more sensitive to R^∞ . For this reason it is again important to analyze both total and capture cross sections together to determine the best s -wave parameters and their correlations.

Given these input parameters, S_0 , $\langle \Gamma_\gamma \rangle_0$ and R_0^∞ are fitted keeping all other parameters fixed. The parameters obtained from the fit to the capture and transmission data are presented in Table 6.12, together with their statistical uncertainties and correlations. There are significant correlations between the fitted parameters, although these are lower than those of previous works where only transmission was analyzed.

Param.	Fitted value	Correlation		
		S_0	R^∞	$\langle \Gamma_\gamma^0 \rangle$
S_0	$(1.04 \pm 0.06) \cdot 10^{-4}$	1		
R^∞	-0.106 ± 0.005 fm	0.8	1	
$\langle \Gamma_\gamma^0 \rangle$	40.7 ± 0.5 meV	-0.4	-0.3	1

The value of R^∞ corresponds to $R'=9.4 \pm 0.5$ fm.

Table 6.12: Average resonance parameters calculated in the URR for s -waves with their associated statistical uncertainties and correlation coefficients.

The experimental capture and total average cross sections data are shown in Figure 6.27 together with the cross sections found with SAMMY.

The fitted neutron strength function $S_0=1.04 \pm 0.06$ is 6% larger than the result in the RRR, which is within the estimated uncertainty. The same situation is found for $\langle \Gamma_\gamma^0 \rangle$, that agrees within uncertainties with the value found in the RRR. The case of the channel radius parameter R' is interesting because it has been found that the fit does not converge if the value $R'=10.5$ fm from the evaluations is used. The fitted channel radius $R'_{URR} = 9.4 \pm 0.5$ fm is 8% smaller than the value obtained in the analysis including the transmission in the low energy range where $R'_{RRR} = 10.2 \pm 0.4$. An average value of $R'_{RRR/URR} \sim 9.8 \pm 0.6$ is consistent with the results in the resolved and unresolved region.

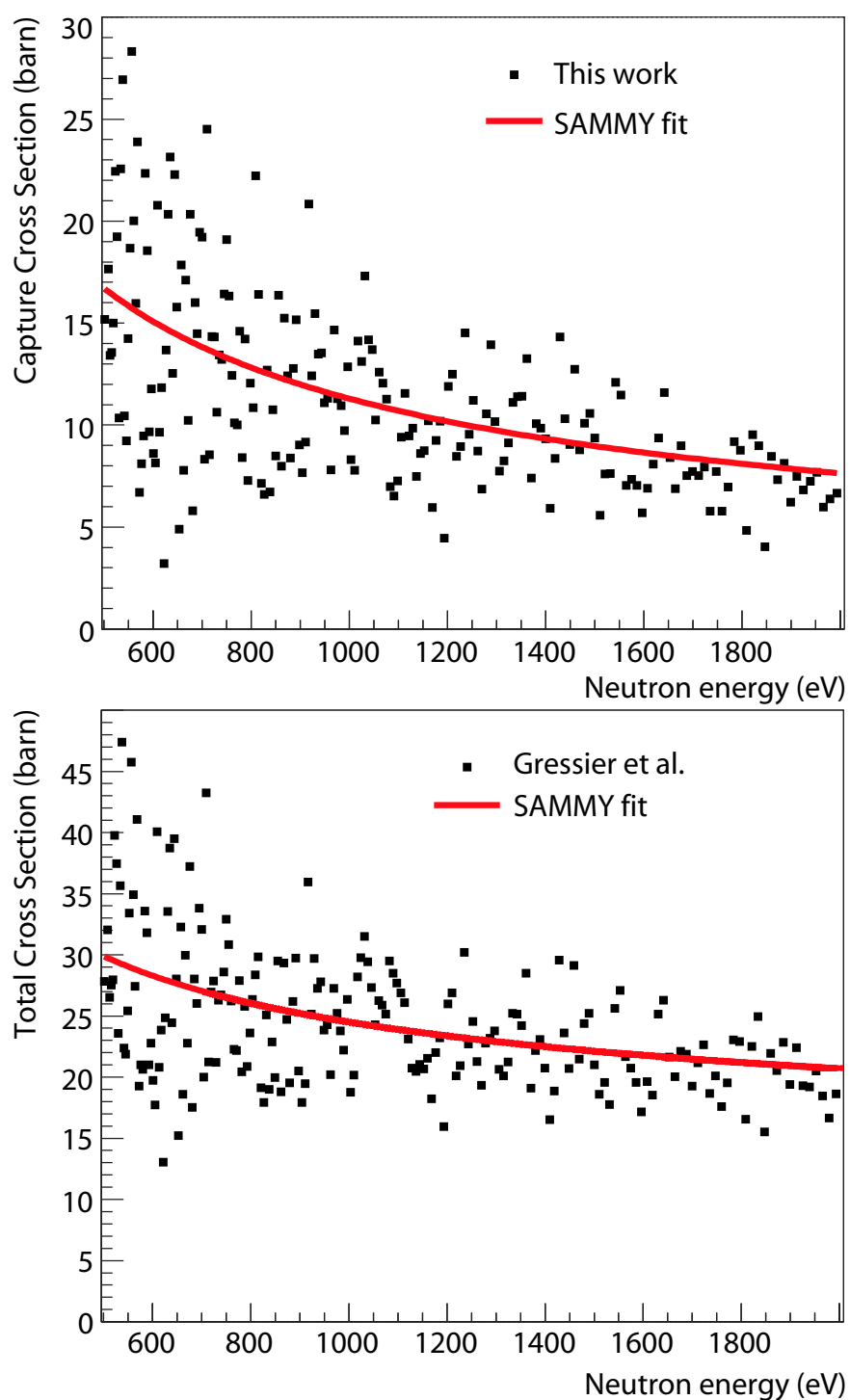


Figure 6.27: ^{237}Np capture (top) and total (bottom) average cross sections in the URR. The corresponding SAMMY fit is shown as a red line.

6.5 Comparison with previous measurements and the evaluated cross sections

6.5.1 Resolved Resonance Region

The result of the analysis of the RRR between 1 and 500 eV is a complete set of resonance parameters for 657 resonances, which is given in Appendix B. Due to the large number of resonances and the overlap between them, the results can be better understood in terms of average quantities. In particular, the results from this work in the RRR have been compared to previous measurements and evaluations by looking at the average cross section in different energy intervals:

$$\langle\sigma_\gamma\rangle_{i,f} = \frac{\int_{E_i}^{E_f} \sigma_\gamma dE}{E_f - E_i}, \quad (6.17)$$

which is computed using NJOY-99 [149].

Other authors have compared their results in selected energy intervals below 100 eV [124, 136]. Hence, the same intervals have been chosen in this work. However, the good energy resolution of the n_TOF data allows one to investigate the average cross sections calculated from resonance parameters up to larger neutron energies. Therefore, two additional energy intervals have been analyzed: 100-280 eV and 280-500 eV.

E_1 - E_2 (eV)	$\langle\sigma_\gamma\rangle_{Thiswork}$	$\frac{JEFF-3.1}{Thiswork}$	$\frac{Hoffman}{Thiswork}$	$\frac{Weston}{Thiswork}$	$\frac{Kobayashi}{Thiswork}$	$\frac{Scherbakov}{Thiswork}$
1-2.8	158.6±4.8	1.01	-	1.01	1.16	1.00
2.8-5.2	66.9±2.0	1.01	-	1.03	1.16	1.04
5.2-8	104.6±3.1	1.00	-	1.00	1.16	1.01
8-16	91.5±2.7	1.00	-	1.01	0.94	1.04
16-28	97.9±3.0	1.01	0.442	0.99	1.11	1.03
28-45	46.0±1.4	1.03	0.554	1.03	1.10	1.11
45-73	66.8±2.0	1.06	0.492	1.01	1.37	1.05
73-100	43.3±1.3	1.02	0.546	1.02	1.53	1.01
100-280	27.4±0.8	1.02	0.599	-	-	-
280-500	16.9±0.5	1.01	0.498	-	-	-

Table 6.13: Average capture cross section of ^{237}Np from 1 to 500 eV calculated with NJOY-99 and compared to the values from previous measurements and evaluations. Values in bold indicate that the difference exceeds the uncertainty of the present results.

The average cross section values of this work and their comparison with the values from previous measurements and evaluations are summarized in Table 6.13, where the values in bold indicate that the difference is beyond the uncertainty in the present results. It should be noted that the values of the JENDL-3.3 and the ENDF/B-VII evaluations are the same than those of the JEFF-3.1 evaluation. The experimental values correspond to the measurements of Hoffman

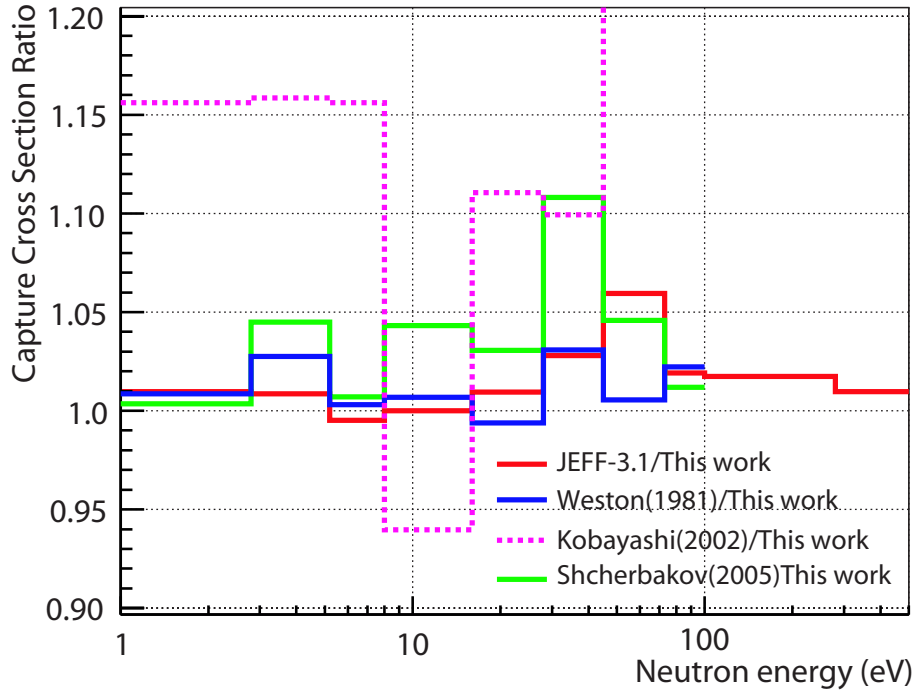


Figure 6.28: Ratios of the n -TOF average capture cross section of ^{237}Np from 1 to 500 eV and those of previous measurements and evaluations (see Table 6.13).

et al. [129], Weston et al. [124], Kobayashi et al. [133], and Scherbakov et al. [136]. The ratios reported in the table are illustrated in Figure 6.5.1.

Looking at the ratios listed in Table 6.13 and displayed in Figure 6.5.1, it can be concluded that the evaluated cross sections, as well as the data by Weston and Scherbakov, are in good agreement (within 5%) with the results of this work. On the contrary, the data of Kobayashi and Hoffman are in clear disagreement in the major part of the resolved resonance region. In particular:

- The results of this work and the evaluated cross sections are in agreement within a few percent in the complete resolved resonance region. The evaluations are always equal to or larger than n -TOF with maximum differences of 3%, except between 45 and 73 eV, where the difference is 6%.
- The data of Hoffman et al., available only above 16 eV, are in clear disagreement with this work. In the complete energy range they are 40% to 50% lower than the present values.
- The data of Weston, available only up to 100 eV, are in remarkable agreement with the present results. The differences between the two data sets are always smaller than 3%.
- The data of Kobayashi et al. are in clear disagreement with this work. At low energies (1-8 eV) they are 16% larger; between 8 and 16 eV they are 11% lower; and at larger

energies, they are always larger (as much as 53%).

- The data of Scherbakov et al., available only up to 100 eV, are in overall agreement with the present values, except in the energy interval between 28 and 45 eV where they are 11% larger. In the other energy intervals they differ less than 5%, but are systematically larger.

The point-wise cross sections of the different measurements are compared to the present results in Figures 6.29 to 6.30. The data of Scherbakov correspond to average cross sections only and are therefore not shown. Concerning the energy calibration between the different measurements, it seems that the data of Esch et al. have a shift to lower energies while the data of Kobayashi et al. show a shift to larger energies.

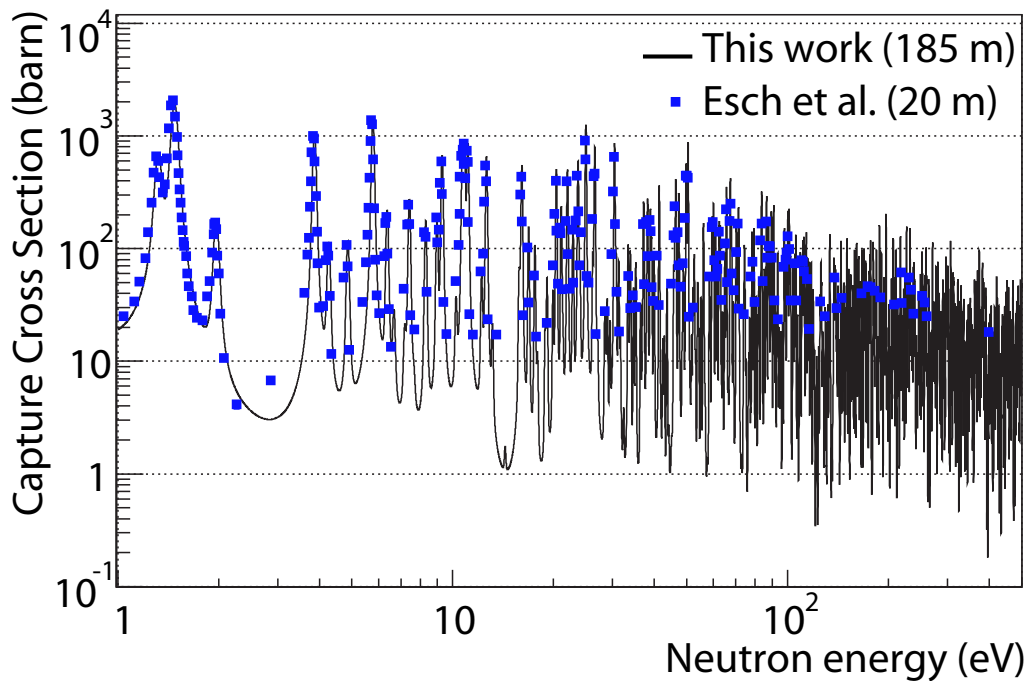


Figure 6.29: *n*-TOF neutron capture cross section compared to the data of Esch et al.

The figures illustrate the better resolution of the *n*-TOF data at these energies compared to all previous measurements. This instrumental feature of the *n*-TOF data serves to increase the limit of the resolved resonance region in the cross section of some minor actinides that were previously measured at shorter flight paths. Although the resonances overlap significantly at high energies, it is important to perform high resolution measurements because the uncertainty caused by the background is minimized with respect to those of the average cross sections measurements. In the latter measurements, the background becomes the largest source of uncertainty. For instance, the background introduces an uncertainty of 6% and 5% in the average cross section measurements of Weston et al. [150] and Scherbakov et al. [136]. Esch et al. [134] do not provide a quantitative estimation of this source of uncertainty.

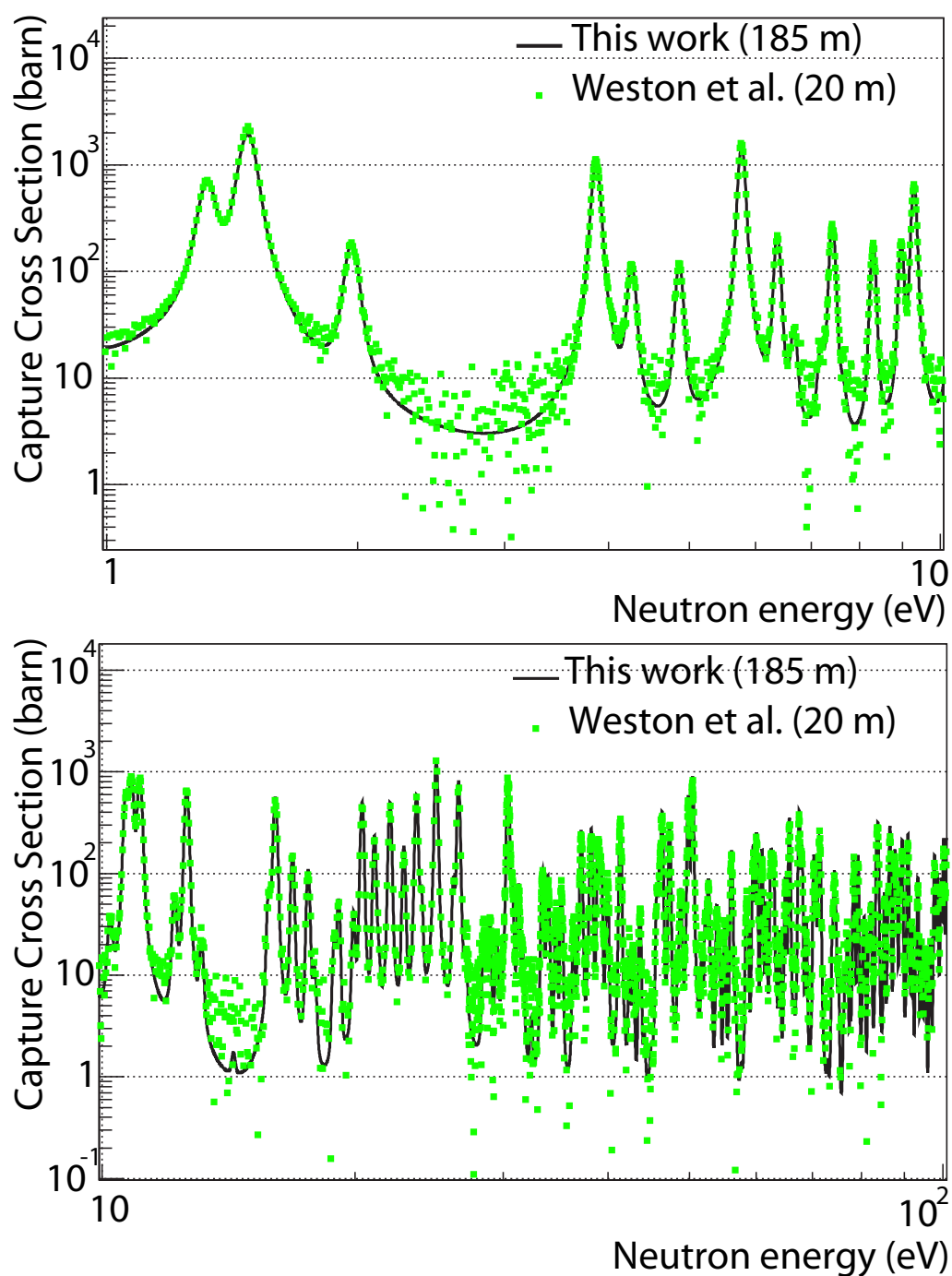


Figure 6.30: n -TOF neutron capture cross section compared to the data of Weston et al. between 1 eV and 100 eV.

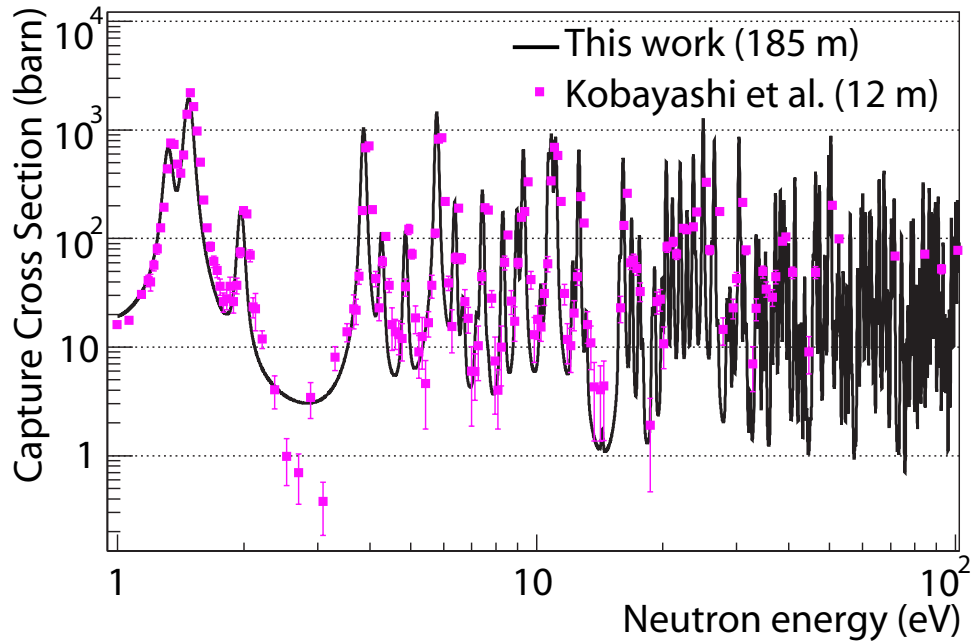


Figure 6.31: n -TOF neutron capture cross section compared to the data of Kobayashi et al.

6.5.2 Unresolved Resonance Region

The experimental data sets available in the URR are the same as for the RRR: Hoffman et al., Weston et al., Kobayashi et al. and Esch et al.; with the only exception of the data of Scherbakov et al., that range up to 100 eV.

Figure 6.32 shows the capture cross section resulting from the analysis with SAMMY of the capture yield measured in this work compared with those of the evaluations and the previous measurements. The cross section of this work agrees with the evaluations and therefore with the data of Weston et al. On the contrary, there are significant differences with respect to all other experimental values.

The largest differences are found with respect to the measurement of Hoffman et al. which are between 30% to 50% lower than all other measurements. The cross section of Kobayashi et al. is clearly above this work, but the difference decreases with neutron energy from 25% at 500 eV to only 10% at 2 keV. The situation is the opposite for the recent measurement of Esch et al., whose cross section agrees with this work and the evaluations around 500 eV, but decreases too rapidly with neutron energy and becomes 25% lower than this work at the 2 keV limit. This behaviour of the cross section shape of Esch et al. could be related to an accordion effect due to an offset in the time-of-flight of a few μ s, which corresponds to ~ 100 eV in the keV region. For instance, the same time-of-flight offset could explain the energy displacement observed at low energies (see Figure 6.29). However, no references have been found concerning the time-energy calibration applied in the measurements at DANCE.

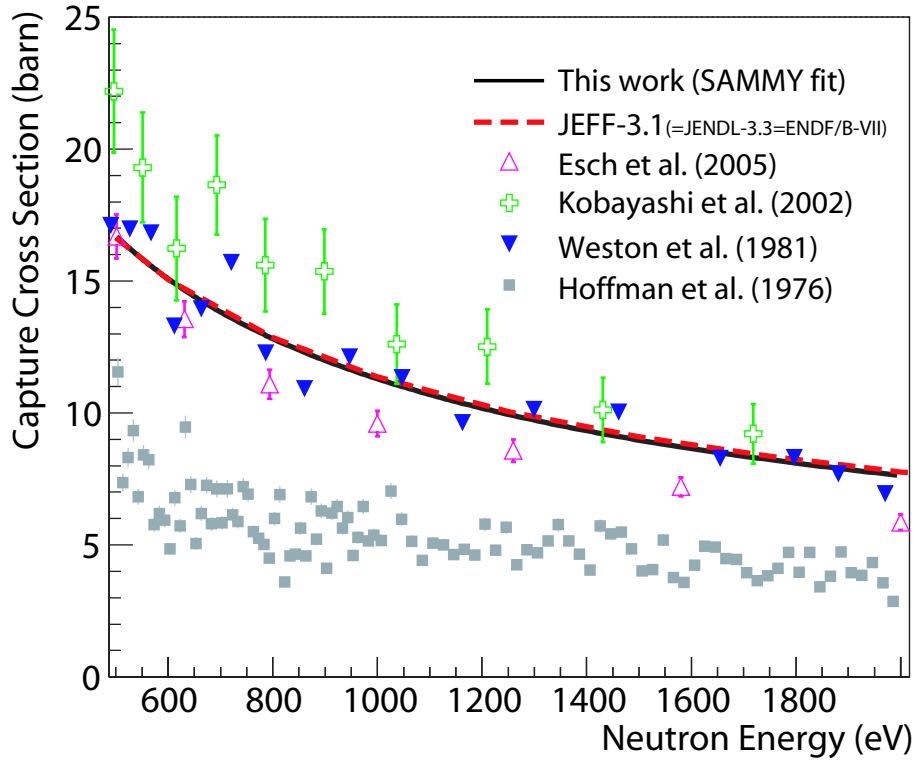


Figure 6.32: Comparison of the ^{237}Np URR capture cross section of the n_{TOF} data compared with previous data and evaluations.

6.6 Summary and conclusions

The neutron capture cross section of ^{237}Np has been measured using the n_{TOF} Total Absorption Calorimeter (TAC) in the energy range between 1 eV and 2 keV, which includes the complete resolved resonance region. The measurement provided, with good statistics, the highest resolution neutron capture cross section data to date.

The background has been highly reduced by setting specific conditions on the deposited energy and the crystal multiplicity of the detected events: $2.5 < E_{\text{sum}}(\text{MeV}) < 6$ and $m_{\text{cr}} > 2$. The detection efficiency of the TAC under those conditions has been determined to be 70% by means of Monte Carlo simulations using GEANT4. The accuracy reached is 2.5%. In this measurement, the counting rate was kept below $0.5 \mu\text{s}^{-1}$ in order to minimize the effect of dead-time losses.

The capture yield was determined in two different and independent ways: (i) using the normalization by the Saturated Resonance Method applied to the reference resonance at 4.9 eV in ^{197}Au , and (ii) normalizing the capture data to the most reliable transmission data. The second method provided a more accurate capture yield and, most importantly, the analysis

of both capture and transmission reduced the correlations between the resonance parameters Γ_γ and Γ_n . However, the capture cross section obtained with one or the other normalization method have been shown to be compatible within uncertainties, giving confidence to the various techniques developed for the calculation of the capture yield. Indeed, it would be possible to obtain reliable capture yields also for the cases where transmission data are not reliable or even missing.

The RRR has been analyzed between 1 eV and 500 eV, and is described by 557 resonances. The combined analysis of capture and transmission has shown that both data sets are compatible in the complete RRR. The analysis of the energy region below 43 eV has provided the values of the scattering radius R' and the average radiative width $\langle\Gamma_\gamma\rangle$ that are kept constant for the analysis of resonances at higher energies. The resulting covariance matrix indicate that there are sizable correlations between the resonance parameters of neighbouring resonances. However, these correlations are, in most cases, limited to next neighbouring resonances. On the other hand, the normalization and background correction parameters exhibit negative correlations of -0.25 on average, with the capture and neutron widths of most resonances. The statistical analysis of the resonance parameters indicates that the radiative width follows a constant behaviour within uncertainties, the distribution of the distance between neighbouring resonances is compatible with the expected Wigner distribution (even when resonances of different spins are analyzed individually), and the neutron strength function S_0 does not show any dependence on neutron energy.

The URR has been studied between 500 eV and 2 keV. The capture yield was analyzed together with the transmission data with the SAMMY/FITACS code. The analysis has shown that both data sets are compatible and that it is possible to determine a set of average resonance parameters ($\langle\Gamma_\gamma\rangle$, R' and S_0) compatible within uncertainties with the values obtained from the statistical analysis of the resonance parameters in the RRR. The combination of the results from the resolved and unresolved resonance regions provide a set of recommended values for the statistical description of the cross section that is given in Table 6.14, where the values of different evaluated data libraries are included for comparison.

	This work	JEFF-3.1	
		JENDL-3.3	RIPL-2
		ENDF/B-VII	
Level spacing (eV)	0.56 ± 0.02	0.58	0.57 ± 0.03
$\langle\Gamma_\gamma\rangle$ (meV)	40.8 ± 1.8	40.0	40.8 ± 1.2
Strength function ($\times 10^{-4}$)	1.03 ± 0.08	1.0	0.97 ± 0.07
R' (fm)	9.8 ± 0.6	10.5	-

Table 6.14: *Average resonance parameters of ^{237}Np determined in this work compared to the values in the evaluations.*

The capture cross section measured and analyzed in this work has been averaged with NJOY-99 and compared to those from previous measurements and to evaluated data libraries. Overall,

the results from this work are compatible with the most recent evaluated cross sections. There is also a reasonable agreement with the data of Scherbakov et al. below 100 eV and Weston et al. in the complete energy range. Compared to other results in literature there are significant differences beyond the estimated uncertainties of the corresponding measurements. This is the case for the data of Kobayashi et al., which deviate largely in both the resolved and unresolved resonance regions; and for the data of Esch et al., which are in agreement with the present cross section around 500 eV but show significant differences at higher energies.

In summary, this work has provided a set experimental data for the capture cross section of ^{237}Np in the energy region between 1 eV and 2 keV. All sources of uncertainty have been carefully investigated and the uncertainties reported are realistic and provide confidence in the overall accuracy of the capture yield (4%). The measured capture data are compatible with the most reliable transmission data, which have been included in the analysis in order to provide a complete set of resonance parameters, the associated covariance matrix, and a set of statistical parameters suitable for the calculation of the cross section at higher neutron energies. The compatibility with the transmission data and the agreement with the evaluated capture cross sections indicate that the uncertainty of 15% in the capture cross section of ^{237}Np estimated previous to this work is significantly overestimated. A value of 4% in the region under study seems more reasonable in view of the present results. Concerning the resonance parameters, the results of this work will most likely be the basis of the new evaluation for the JEFF library.

Chapter 7

Measurement and analysis of the $^{240}\text{Pu}(n,\gamma)$ cross section

At present, all the evaluated cross sections of ^{240}Pu in the resolved resonance region are based exclusively on transmission and fission data. This is related to the lack of high resolution neutron capture cross section measurements. For instance, the most reliable neutron capture measurement to date was performed by Weston et al. [150, 151] but provided only average cross sections. The comparison of the evaluated capture cross sections of ^{240}Pu reveals significant differences, even though they are all based on the same set of transmission data.

Energy range	σ_f	σ_{el}	σ_γ
<0.54 eV	50%	5%	0.5%
0.54 - 4 eV	20%	10%	3%
4 - 22.6 eV	20%	10%	8%
22.6 eV - 67.4 keV	20%	10%	10%
67.4 - 183 keV	20%	10%	15%
183 - 498 keV	20%	10%	25%
498 keV - 2.23 MeV	10%	10%	30%
2.23 - 20 MeV	5%	10%	30%

Table 7.1: *Uncertainties in the neutron cross section of ^{240}Pu .*

The systematic comparison between the experimental and evaluated data [22] provides (see Table 7.1) an estimate of the uncertainty in the neutron cross sections of ^{240}Pu as a function of neutron energy. The capture cross section fairly accurate between thermal energies and a few eV ($< 3\%$), but the uncertainties increase to 10%, 15%, 25% and 30% as the neutron energy reaches 20 eV, 67.4 keV, 183 keV and 498 keV, respectively. These large uncertainty are responsible for the loss of accuracy in the calculation of macroscopic parameters of ADS [25].

The goals of this work are to reduce the uncertainty in the cross section of ^{240}Pu by providing the nuclear data community with the first high resolution capture data in the resolved resonance region, and to perform an analysis of the experimental data for obtaining an accurate set of resonance parameters and covariance matrices.

7.1 Previous measurements and evaluations

There have been a few measurements of the ^{240}Pu capture cross section in different energy regions. These experiments as well the fission and the total cross section measurements are briefly discussed.

7.1.1 Previous measurements

In 1959, Cote et al. [152] measured and analyzed for the first time the total cross section of ^{240}Pu between 1 eV and 40 eV. They measured the total cross section of ^{239}Pu using several samples and inferred the cross section of ^{240}Pu from the resonant structures corresponding to the ^{240}Pu impurities of 0.6% and 5.1% in their samples.

In 1972, Hockenbury et al. [153] performed a set of dedicated capture, fission and transmission measurements at the RPI facility. They used liquid scintillators for the detection of neutron capture reactions in the neutron energy range between 20 eV and 30 keV. The capture data were normalized to the strong resonances measured in transmission at low neutron energies.

In 1977, Weston and Todd [150] measured at ORELA the capture cross section of ^{240}Pu . They used total energy detectors and surrounded the sample by a 1.3 cm thick lead shield in order to reduce the background caused by the activity of the sample. The results of their analysis were average values of the cross section in a large number of intervals between 200 eV and 350 keV, with an overall uncertainty that ranged from 7% to 13%. The results were in agreement within uncertainties with those of Hockenbury et al.

The most recent total cross section data are those of Kolar et al. [154], who measured three ^{240}Pu samples at the GELINA facility in 1968. The measurement with the thickest sample (0.00416 atoms/barn) was performed at the 50 m time-of-flight station with the neutron monitor (BF_3) placed at 100 m from the neutron source. The transmission data are available in the EXFOR database and correspond to neutron energies between 20 eV and 5.7 keV.

The first measurement of the subthreshold fission cross section of ^{240}Pu was performed by Leonard et al. [155] in 1960. Since then, additional measurements were carried out by Auchampaugh et al. [156] in 1975 and Weston et al. [157] in 1984.

7.1.2 Evaluated cross sections

The most recent cross section evaluations of ^{240}Pu are briefly discussed in the following

- **ENDF/B-VII (2006).** The neutron cross section of the ENDF/B-VII evaluation corresponds to the work by Weston and Arthur [151] in 1987 and covers the resolved resonance region up to 5.7 keV. The resonance at 1.06 eV was analyzed from several transmission and fission measurements. From 20 to 5.7 eV the neutron and radiation widths are essentially those from Weigmann et al. [158] based on the transmission data of Kolar et al. [154]. The fission widths correspond to the weighted averages of several fission measurements, mainly those of Weston et al. [157] and Auchammpaugh et al. [156] .
- **JEFF-3.1 (2006).** The neutron cross section of the JEFF-3.1 evaluation in the resolved resonance region ranges up to 5.7 keV and is based on the work of Bouland et al. [76]. Bouland et al. analyzed in 1997 the available transmission and fission data and provided a set of resonance parameters from 1 eV to 5.7 keV. Their analysis of the first resonance at 1.056 eV included the previous results by Spencer et al. [159] and the fission data of Leonard et al. Between 20 eV and 5.7 keV, the analysis included the transmission data of Kolar et al. and the fission data of Weston et al.
- **JENDL-3.1 (2002).** The JENDL-3.3 evaluation is also based in the analysis of Bouland et al. [76]. However, the upper limit of the resolved resonance region was shifted down to 2.7 keV. Another change with respect to the analysis of Bouland et al. is that the capture widths of the resonances at -3 eV and 1.06 eV were slightly modified for better reproduction of the thermal value.

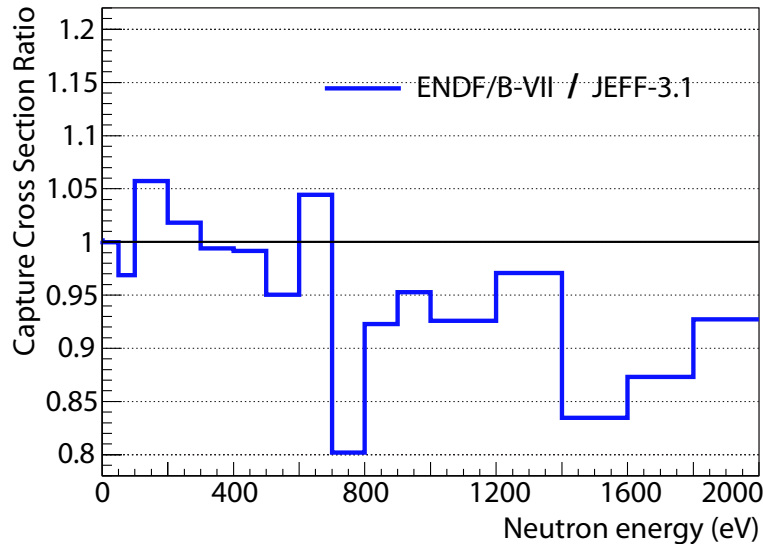


Figure 7.1: Comparison between the capture cross sections of the JEFF-3.1 (=JENDL-3.3) and ENDF/B-VII evaluations.

The evaluated ^{240}Pu cross sections of the main libraries are based mainly on transmission measurement of Kolar et al. [154]. Nevertheless the evaluations differ significantly due to the analysis techniques and the assumptions adopted by the evaluators. Figure 7.1 illustrates the differences between the evaluated capture cross sections of ^{240}Pu in the energy range of interest. The evaluations are compatible within 5% below 700 eV, but show large discrepancies (10% on average) at higher energies. The largest difference (19%) is found between 700 eV and 800 eV where there is a cluster of strong fission resonances.

7.2 The $^{240}\text{Pu}(n,\gamma)$ measurement at n_TOF

7.2.1 The ^{240}Pu sample

The ^{240}Pu sample was a pellet of 51.2 mg in mass and 10 mm in diameter, which was pressed from oxide powder at IPPE Obninsk [96]. The pellet was deposited between two aluminium foils (0.15 mm thick) and encapsulated in sealed titanium (0.2 mm thick) such that the complete assembly was certified with the *ISO-2919 norm*, as required by the safety authorities at CERN. For easy handling, the sample was mounted between two Kapton foils of 25 μm stretched over a carbon fibre ring as shown in Figure 3.11. The main characteristics of the sample are summarized in Table 7.2.

$^{240}_{94}\text{Pu}$	$A+1S_n$	5241.52 keV
	I^Π	0^+
	τ	6563 years
Sample	Chemical form	$^{240}\text{PuO}_2$
	Activity (MBq)	458 ± 58
	Pu mass (mg)	51.2 ± 1.5
	Thickness (atoms/barn)	$(1.64 \pm 0.05) \cdot 10^{-4}$
	Diameter (cm)	1.0
	Abundance (^{240}Pu)	0.88 ± 0.05
	Abundance (^{239}Pu)	0.11 ± 0.02
	Abundance (others)	0.01 ± 0.005

Table 7.2: $^{240}\text{Pu}(n,\gamma)$ measurement and sample properties. The isotopic abundances were determined from a spectroscopic analysis performed at CERN.

The isotopic composition of the sample provided by the manufacturer turned out to be not very accurate. Since a preliminary analysis of the capture data revealed a significant but undeclared ^{239}Pu impurity, the isotopic composition was measured with an accuracy of 5% by γ spectroscopy at CERN. The total activity of the sample was measured to be 458 ± 58 MBq. Part of this activity corresponded to ^{239}Pu (10 MBq), ^{241}Pu (2.2 MBq) in equilibrium with its daughter ^{237}U , and ^{241}Am (1.3 MBq), the daughter of ^{241}Pu . However, most of the ^{241}Am must

have a different origin because the decay of ^{241}Pu would produce only 1 kBq. Traces of the daughters of the previously mentioned isotopes were also found: ^{236}U (23 MBq) and ^{233}Pa (400 kBq). The isotopic composition resulting from this analysis was 88% of ^{240}Pu , 11% of ^{239}Pu , and less than 1% for the rest.

7.2.2 The $^{240}\text{Pu}(n,\gamma)$ measurement

The measurements with the TAC provide a list of detected events, each characterized by its deposited energy (E_{sum}), crystal multiplicity (m_{cr}), time-of-flight (TOF) and the corresponding neutron energy (E_n).

Figure 7.2 shows the energy deposited in the TAC during the measurement of the ^{240}Pu sample. The contribution from $^{240}\text{Pu}(n,\gamma)$ reactions is observed up to 6 MeV, just above $S_n(^{241}\text{Pu})=5.24$ MeV. The background caused by the activity of the sample is observed only at low energies ($E_{\text{sum}} < 0.5$ MeV). The background measured in the beam-off and sample-out runs is common to every measurement with the TAC and is discussed in Section 4.2.2. The characteristic background caused by the sample assembly corresponds, at low neutron energies, to capture reactions in titanium producing two peaks, above 6 MeV, whose positions are given by the neutron separation energies of the titanium isotopes. At higher energies, neutron scattering on titanium becomes the dominant source of background.

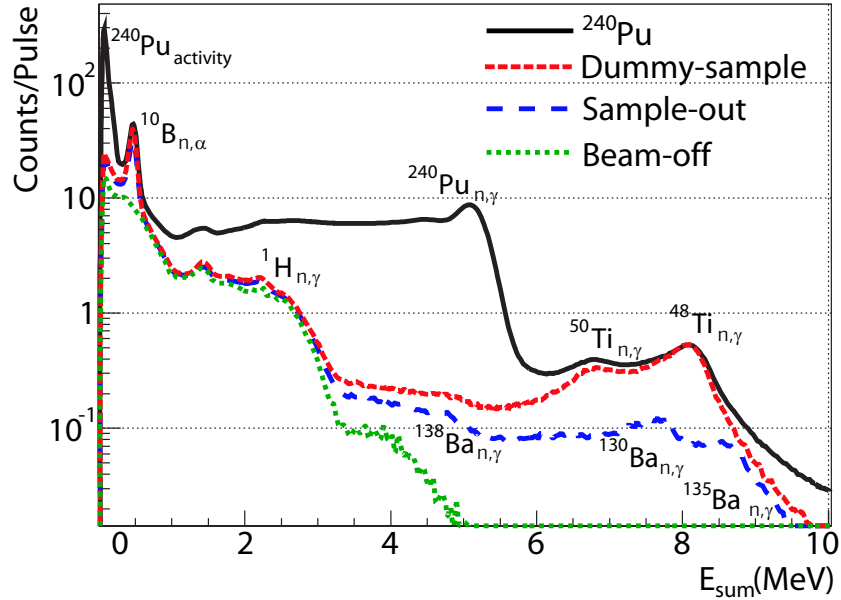


Figure 7.2: Measured deposited energy distributions during the ^{240}Pu and the dedicated background measurements. The data correspond to neutron energies between 1 eV and 10 eV.

Figure 7.3 shows the counting rate as a function of neutron energy recorded during the ^{240}Pu

and the dedicated background measurements. The data correspond to events with deposited energies between 2.5 MeV and 6 MeV, where the capture to background ratio is more favourable (see Figure 7.2). The resonant structures in ^{240}Pu are identified above the background up to a few keV. However, due to a decrease in the capture cross section of ^{240}Pu and the existence of strong titanium resonances in the keV region, the capture data of the ^{240}Pu measurement could be analyzed only below 2 keV.

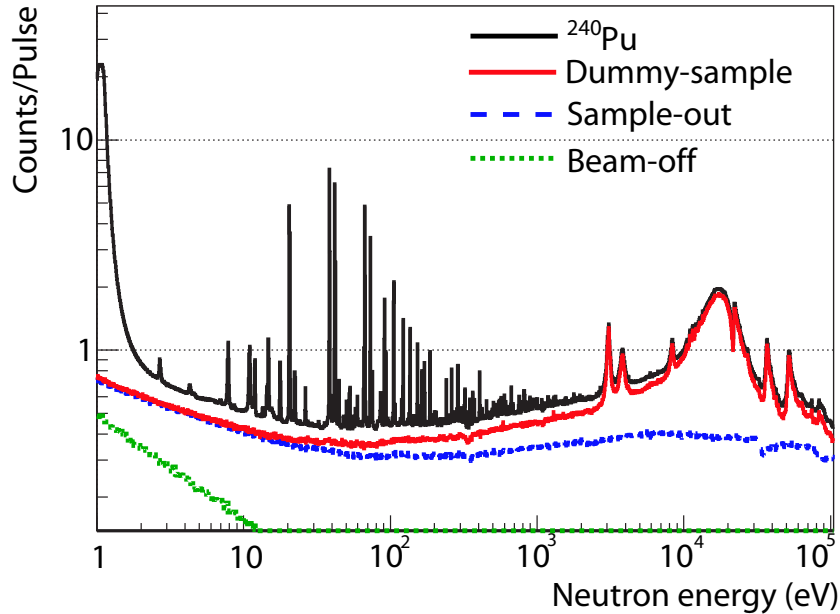


Figure 7.3: Measured counting rate distributions during the ^{240}Pu and the dedicated background measurements. The data correspond to events with deposited energies between 2.5 MeV and 6 MeV, where the capture to background ratio is more favourable.

The segmentation of the detector provides a powerful tool for the reduction of the different sources of background. For instance, the α and β activity of the crystal, that contribute up to several MeV, produce single crystal events. Their contribution could, therefore, be significantly reduced by the condition $m_{cr} > 1$. This is illustrated in Figure 7.4, which shows the distribution of deposited energy of ^{240}Pu for different crystal multiplicities. A threshold of two in m_{cr} eliminates the major part of the background without significantly affecting the contribution corresponding to capture reactions in ^{240}Pu . Since all of the $^{240}\text{Pu}(n,\gamma)$ events are found below 6 MeV, the event selection conditions that have been chosen for the analysis are $m_{cr} > 2$ and $2.5 < E_{sum}(\text{MeV}) < 6$.

The neutron scattering cross section of ^{240}Pu at low energies is lower than that for capture, while above a few hundred eV it can be as much as 10 times larger at the peak of the resonances. As a consequence, some resonances present a sizable neutron sensitivity resonant background due to scattering reactions in the sample.

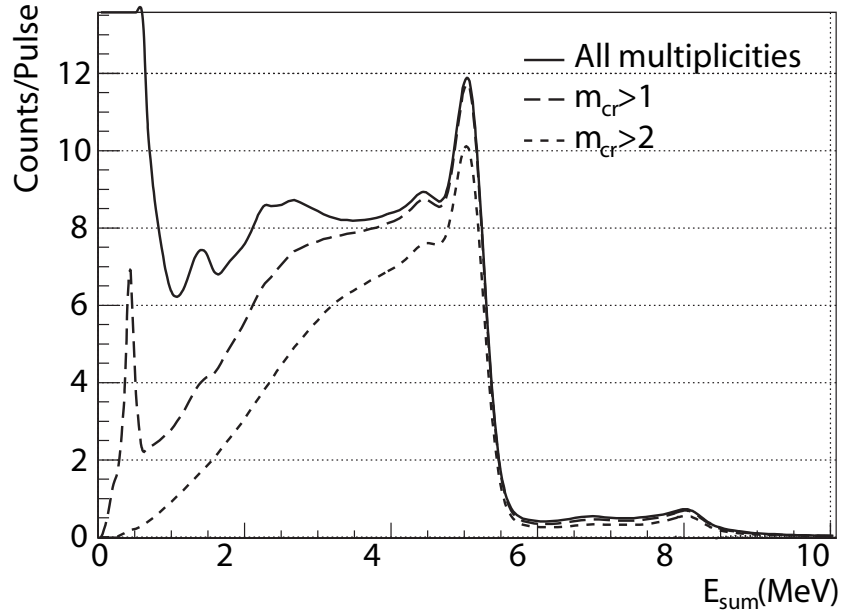


Figure 7.4: Distribution of deposited energy for the $^{240}\text{Pu}(n,\gamma)$ measurement for different values of the lower threshold in crystal multiplicity. The events correspond to neutron energies between 1 eV and 10 eV.

Figure 7.5 shows the overall background (red line) as a function of time-of-flight in two regions: ~ 300 eV (left) and ~ 800 eV (right). While the background in the left panel is completely flat, a resonant neutron scattering background contributing as much as 10% is observed below some resonances in the right panel. The point-wise background as a function of time-of-flight (see Section 4.2.4) is included properly in SAMMY for the resonance analysis.

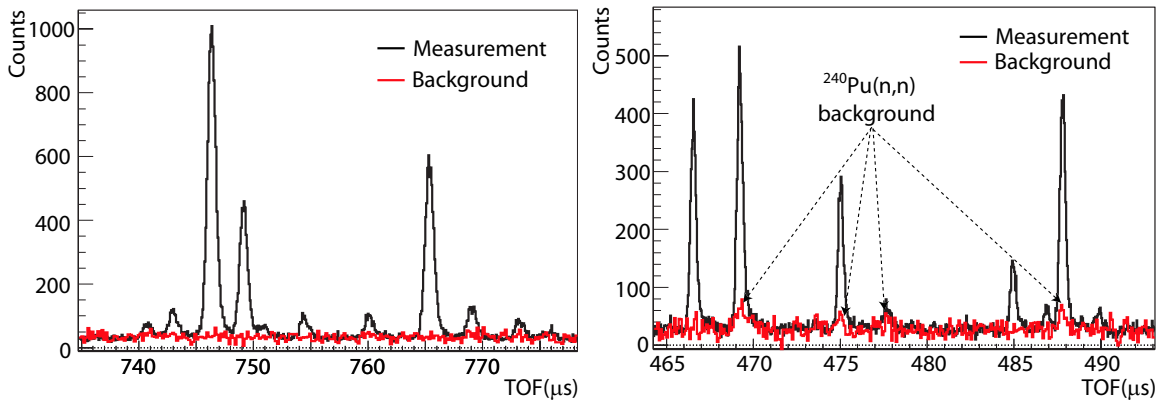


Figure 7.5: Distribution of events as a function of time-of-flight ($m_\gamma > 2$ and $2.5 < E_{\text{sum}}(\text{MeV}) < 6$). Black: total counting rate. Red: overall background.

7.2.3 Calculation of the capture yield

The experimental capture yield is determined as

$$Y_{n,\gamma}^{exp} = \frac{C - B}{\varepsilon \cdot N \cdot \Phi_n}, \quad (\text{see Section 5.2.2.}) \quad (7.1)$$

where the detection efficiency ε is calculated by MC simulations, the neutron beam intensity Φ_n is given by the Silicon Monitor *SiMon*, and the normalization factor N is determined by the Saturated Resonance Method (see Section 5.2.2). The latter has been applied to both, the 4.9 eV resonance of ^{197}Au and the 1.06 eV resonance of ^{240}Pu .

7.2.3.1 Determination of the detection efficiency

The value of ε is determined by means of a Monte Carlo simulation of the complete neutron capture measurement. In the simulation, the capture cascades that are generated at the centre of the TAC follow the experimental counting rate distribution as a function of time-of-flight in such a way that it is possible to estimate the signal losses in the individual crystals related to the dead-time effects. The detection efficiency resulting from the simulation is a function of the event selection conditions on E_{sum} and m_{cr} and also of the neutron energy through the time-of-flight dependent counting rate. A detailed discussion of the Monte Carlo simulation and the dead-time correction model is given in Section 5.1.

The capture cascades of the MC simulation are produced from the experimental information on the nuclear level scheme and transition probabilities of ^{241}Pu available at low excitation energies, and from statistical model calculations of level density LD (using the Back Shifted Fermi Gas model) and transition probabilities (PSF combined with the Brink hypothesis) at high excitation energies. The initial set of input parameters for the LD and PSF was obtained from the RIPL-2 database [41]. To reproduce the experimental data it was, however, necessary to adjust the PSF parameters by increasing the intensity of the M1 with respect to E1 transitions. A more detailed discussion of the PSF investigations for actinides isotopes with the TAC is given in Ref. [119].

The simulated and measured distributions of deposited energy and crystal multiplicity are compared in Figure 7.6, where the experimental data correspond to the ^{240}Pu resonance at 20 eV. The agreement between the simulation and experimental data is excellent, validating in this way the calculation of the detection efficiency by the MC simulations.

Further proof of the reliability of the MC simulations is provided by comparing the experimental and simulated distributions of deposited energy for each single value of the crystal multiplicity. These distributions, shown in Figure 7.7, are very sensitive to the particular characteristics of the capture cascades. The discrepancies observed at low deposited energies in the distributions for low crystal multiplicities are related to the difficulty of subtracting the background from the experimental data the first is clearly dominant.

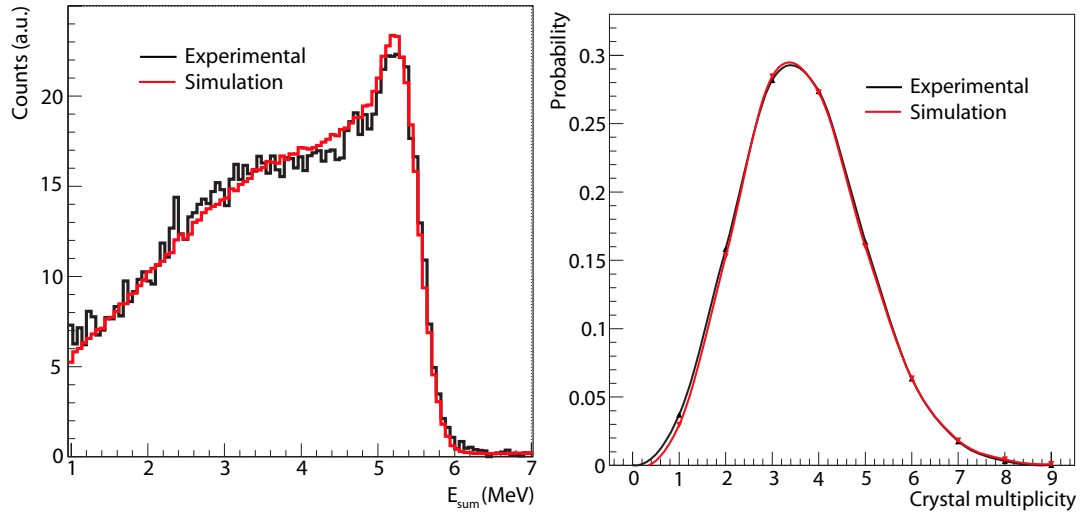


Figure 7.6: Simulated and experimental distributions of E_{sum} (left) and m_{cr} (right).

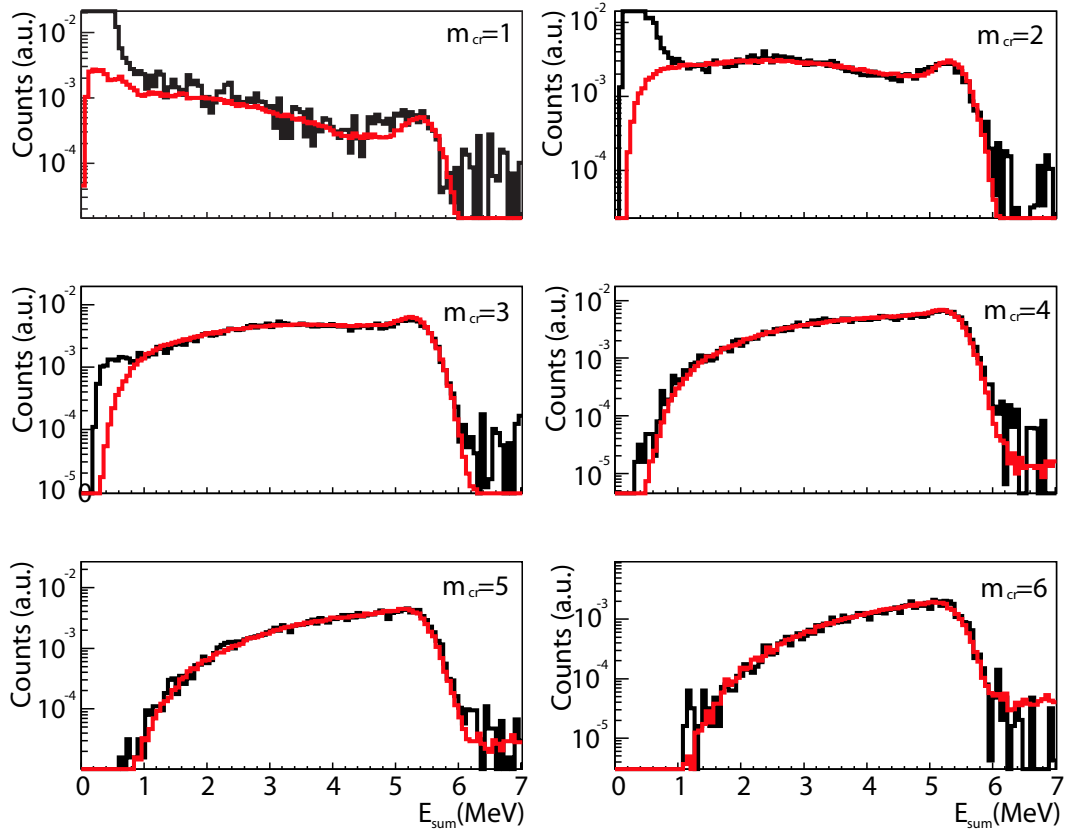


Figure 7.7: Simulated and experimental distributions of E_{sum} for specific values of m_{cr} .

The detection efficiency was calculated for all combinations of E_{sum} and m_{cr} as the ratio of the detected to initial events. For the analysis conditions $2.5 < E_{sum}(\text{MeV}) < 6$ and $m_{cr} > 2$ and a counting rate of $0.6 \mu\text{s}^{-1}$, the detection efficiency for the resonance at 20 eV is

$$\varepsilon = 0.64 \pm 0.02, \quad (7.2)$$

where the uncertainty has been estimated by performing a series of calculations with different but compatible sets of PSF parameters (see Section 5.1.6 for details on this procedure).

The detection efficiency is calculated as a function of neutron energy by distributing the initial events of the simulation according to the experimental counting rate distribution showed in Figure 7.8. The corresponding distribution for the ^{197}Au measurement is shown for comparison, since the dead-time correction model was validated for that case. The counting rate of ^{240}Pu is comparable to the ^{197}Au reference only below 100 eV and remains below $0.5 \mu\text{s}^{-1}$ at higher energies.

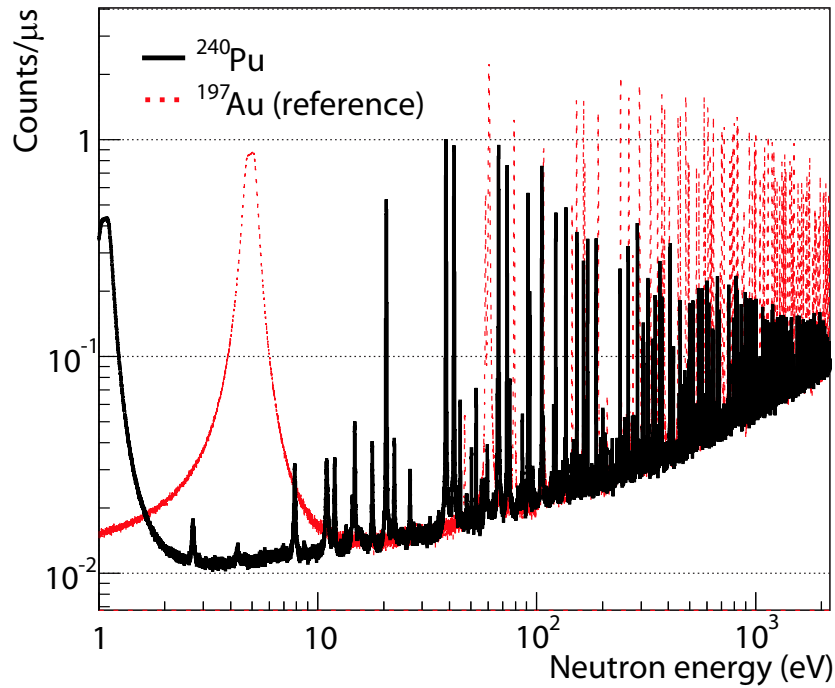


Figure 7.8: *Experimental counting rate of ^{240}Pu (black) used for the distribution in time-of-flight of the initial events in the simulation. The corresponding distribution for ^{197}Au (grey) is shown as a reference.*

The dead-time losses at low energies are always less than 10% and become negligible above 100 due to the low counting rate and the small width of the resonances in terms of time-of-flight.

7.2.3.2 Normalization

The Saturated Resonance Method [120] (see Section 5.2.2) is a very accurate method for the calculation of the normalization factor N that relates the beam intensity recorded in the neutron monitor and that of the neutrons impinging on the sample. The saturated resonance of ^{197}Au at 4.9 eV is commonly used at n_TOF to determine the normalization factor; nevertheless, in this particular measurement the normalization can be also performed by means of the saturated resonance of ^{240}Pu at 1.06 eV.

Figure 7.9 shows the experimental and calculated capture yields at the saturated resonances of ^{197}Au (left) and ^{240}Pu (right) at 4.9 eV and 1.06 eV, respectively. Each experimental yield has been scaled to its own normalization factor, which are in agreement within 1.5%. The average value from both normalizations, with an accuracy better than 2%, has been used for the calculation of the experimental yield.

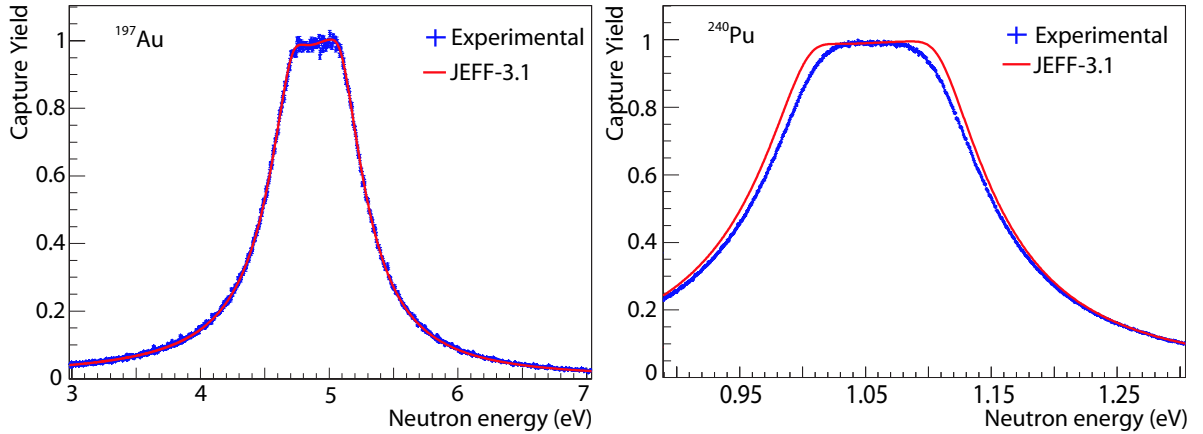


Figure 7.9: Saturated resonances of ^{197}Au (left) and ^{240}Pu (right) at 4.9 eV and 1.06 eV, respectively. The markers correspond to experimental data scaled by a normalization factor N such that they agree with the calculated yield (JEFF-3.1) in the flat top of the resonances. The normalization factors differ by less than 2%.

The right panel in Figure 7.9 indicates that there are significant differences between the experimental and calculated yields of ^{240}Pu at the tails of the resonance. These are related to the powder structure of the sample, which shows grain sizes comparable to the thickness of the sample itself [160]. For this reason, the mass can not be considered uniformly distributed in the sample, as it is assumed in the SAMMY calculation of the yield from resonance parameter from JEFF-3.1. The uncertainty introduced by the uniform mass distribution approximation is discussed in the following section.

7.2.3.3 Uncertainty in the experimental capture yield

The systematic uncertainties in the calculation of the capture yield come from the non-uniform mass distribution of the sample, the isotopic abundance of ^{240}Pu in the sample, the normalization factor, the energy shape of the neutron flux, the neutron scattering background, the detection efficiency and the dead-time losses.

- The analysis of cross section measurements using thin powder samples should take into account that the size of the grains that form the sample (1-10 μm) is comparable to the thickness of the sample itself. This has been underlined in the work of Kopecky et al. [160] for the case of the transmission measurement on ^{242}Pu and ^{240}Pu . Concerning the present measurement, the mean number of layers in the sample is only ~ 7 and therefore the simplified self-shielding correction in the form of $(1-e^{-n\sigma})$ is accurate only when the correction is small. Figure 7.10 shows the size of the self-shielding correction as a function of neutron energy. Since the present analysis cover only the energy region above 0.11 keV, the 20% uncertainty of the correction implies that the associated correction remains below 4%.

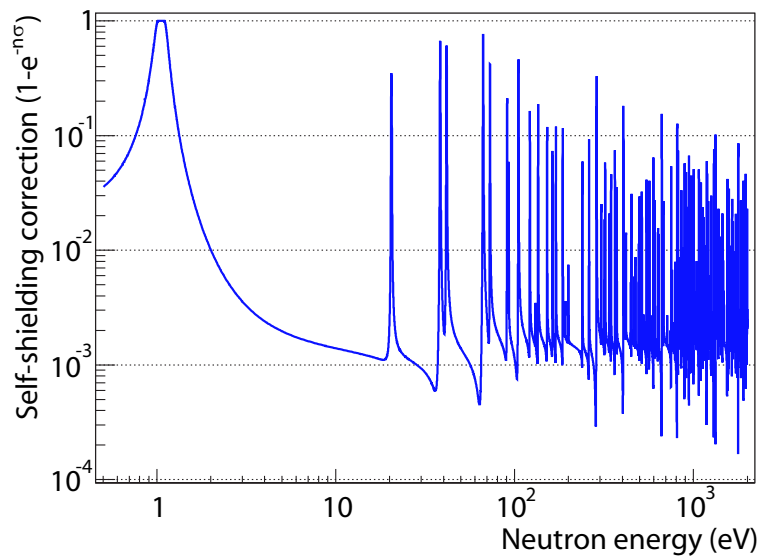


Figure 7.10: Calculation of the self-shielding correction on the ^{240}Pu sample as a function of neutron energy.

- The results of the spectroscopic analysis of the sample at CERN provided an estimation of the isotopic abundance of ^{240}Pu in the sample with an accuracy of 5%. The value of the isotopic abundance of ^{240}Pu was adjusted by means of a combined analysis of the capture data with the most reliable transmission data available (Kolar et al. [154]). The isotopic content of ^{240}Pu was fitted from the analysis in the high energy region, where the thin target approximation is valid and the capture yield is directly proportional to the number of ^{240}Pu nuclei in the sample. The value of the isotopic abundance obtained in this way is

0.84(2), which is more accurate but compatible with the value found in the spectroscopic analysis 0.88(5).

- The uncertainties of the normalization factors calculated via the saturated resonances of ^{197}Au and ^{240}Pu are 3%. Due to their good agreement the average value was adopted in the analysis with an accuracy better than 2%.
- The uncertainty of the detection efficiency from the MC simulations is 2.5%. Due to the low counting rate recorded during the measurement, the additional uncertainty related to the dead-time losses is lower than 1.5% at low neutron energies and negligible above ~ 300 eV.
- Since the neutron sensitivity of the TAC is below 1% (see Section 4.2.4), the neutron scattering background is sizable only for those resonances with a large scattering to capture ratio. In the energy range analyzed there are only 15 out of 164 resonances with $\sigma_n > 3\sigma_\gamma$, for which the uncertainty introduced in the capture yield should be at maximum 2%.
- The uncertainty related to a possible misalignment of the sample with respect to the ^{197}Au reference sample is below 1%, which is confirmed by the agreement between the normalization factors calculated with the saturated resonances of ^{197}Au and ^{240}Pu .

E_n (keV)	Partial uncertainties						Total uncertainty
	Norm.	Isotopic abund.	ε	DT losses	Mass distribution	$\Phi_n(E_n)$	
0.11 - 0.3	2%	2%	2.5%	<1.5%	<4%	2%	< 6.0%
0.3 - 2	2%	2%	2.5%	-	<1%	2%	< 4.4%

Table 7.3: *Partial and total uncertainties of the experimental capture yield of ^{240}Pu . An uncertainty related to the neutron scattering background and lower than 2% should be added in the case of resonances with $\sigma_n > 3\sigma_\gamma$.*

The partial and total uncertainties in the calculation of the capture yield are summarized in Table 7.3, where the final uncertainty is calculated as the square root of the sum of squares of the partial uncertainties. In the complete energy range, the accuracy of the capture yield is better than 6% which is a significant improvement compared to the accuracy reached in previous measurements.

7.3 Analysis of the Resolved Resonance Region

7.3.1 Methodology for the resonance analysis

The cross section resonances observed in the experimental capture yield of ^{240}Pu are well resolved in the complete energy range. Hence it has been possible to analyze them individually

as well as all together in order to find correlations between neighbouring resonances and long range correlations associated to the determination of the background.

The SAMMY code with the Reich-Moore approximation of the R' -matrix theory was used for the resonance analysis of the capture yield between 110 eV and 2 keV. The transmission data of Kolar et al. were included in the analysis in such a way that the resulting resonance parameters are capable of reproducing both capture and transmission data. Prior to the analysis, the original flight times of the transmission data (50 m flight path) were recalibrated to the n_TOF data (185 m flight path).

The procedure described in the previous chapter for the analysis of ^{237}Np was followed for the analysis of ^{240}Pu with only a few differences:

- All of the resonances are considered to be s -wave and therefore all spins are $1/2^+$, since ^{240}Pu is an even-even nucleus with spin 0^+ . The existence of p -wave resonances in this energy range is neglected because the ratio of the penetration factors for s - and p -waves at 1 keV is only 0.005 and the experimental data do not allow to distinguish between p -waves and very small s -waves.
- The radiative width of the prominent resonances can be determined accurately in the complete energy range because the resonances do not overlap. For the small resonances, the radiative width is fixed to the average value determined from the prominent ones.
- The number of resonances in the energy range is small enough to perform single fits in the complete energy range. However, preliminary fits were performed in reduced energy intervals to obtain a set of initial values of the resonance parameters and also for the calculation of the background as a function of energy.
- The ^{239}Pu impurities of the samples used at n_TOF and GELINA are large enough so that the most prominent ^{239}Pu resonances show up. The neutron width of the observed ^{239}Pu resonances was fitted in a first iteration and then kept fixed for the analysis of the ^{240}Pu resonances. Although in some cases it has not been possible to reproduce all the resonances of ^{239}Pu , they are small compared to those of ^{240}Pu and do not have a significant effect in the results.

7.3.2 Results of the resonance analysis

A total of 158 resonances of have been analyzed between 110 eV and 2 keV in ^{240}Pu . The combined resonance analysis of the capture and transmission data has shown that both data are compatible in the complete energy range. In a first preliminary analysis of the high energy range, the resonance parameters were kept fixed and the isotopic abundance of ^{240}Pu in the capture sample was fitted. The resulting value was fixed for the rest of the analysis.

The fits were performed in 14 energy intervals each containing a few tens of resonances. The resonances that were not reproduced in the first iteration were analyzed individually for

obtaining an appropriate set of starting values. Figures 7.11 to 7.24 show the experimental capture and transmission data: the points correspond to the experimental values and the black lines to the capture yield and transmission calculated from the resonance parameters resulting from the fits with SAMMY. The residual of the fit at each data point is shown in the small panels:

$$Residual = \frac{FIT_i - DATA_i}{\Delta_i}, \quad (7.3)$$

where Δ_i is the statistical uncertainty at each data point.

The background level has been fitted in each of the 14 energy intervals of the analysis. The value found in each energy range is given in Table 7.4, where it is observed that the background level remains nearly constant for all energies. The background level represents a contribution of 1% for the resonances at low energies ($E_n < 500$ eV) and as much as 5% for those at higher neutron energies.

$E_n(\text{keV})$	Background	$E_n(\text{keV})$	Background
0.11 - 0.25	0.00019(1)	1.00 - 1.25	0.00017(1)
0.25 - 0.50	0.00018(1)	1.25 - 1.50	0.00022(1)
0.50 - 0.75	0.00016(1)	1.50 - 1.75	0.00017(1)
0.75 - 1.00	0.00017(1)	1.75 - 2.0	0.00020(1)

Table 7.4: *Background level, in units of yield, in each of the 14 energy intervals of the analysis.*

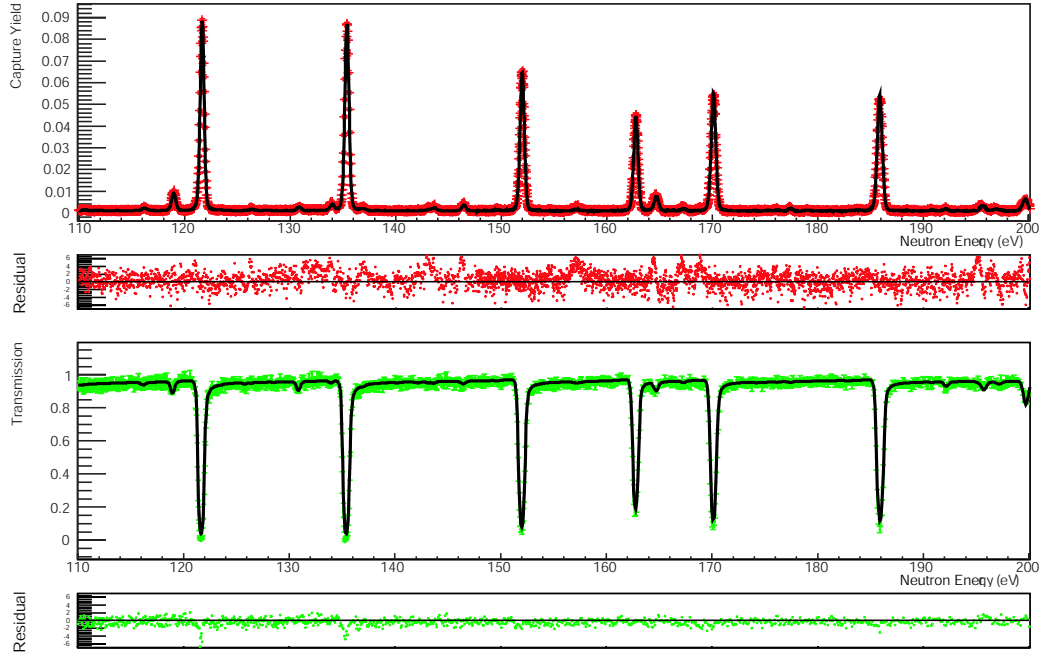


Figure 7.11: *SAMMY fits to capture and transmission between 110 eV and 200 eV.*

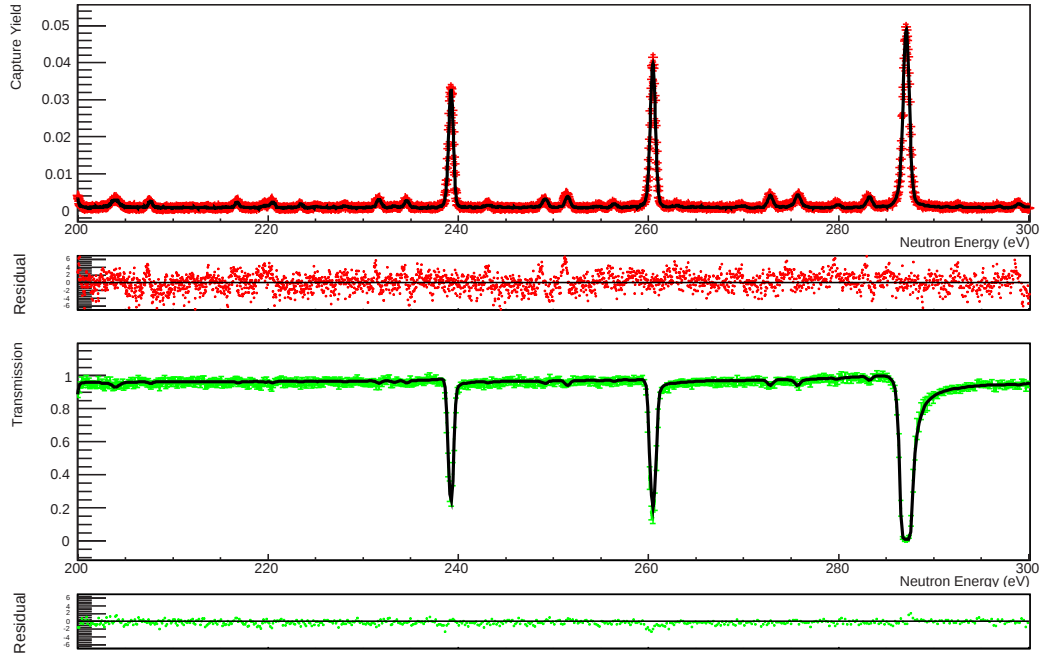


Figure 7.12: *SAMMY fits to capture and transmission between 200 eV and 300 eV.*

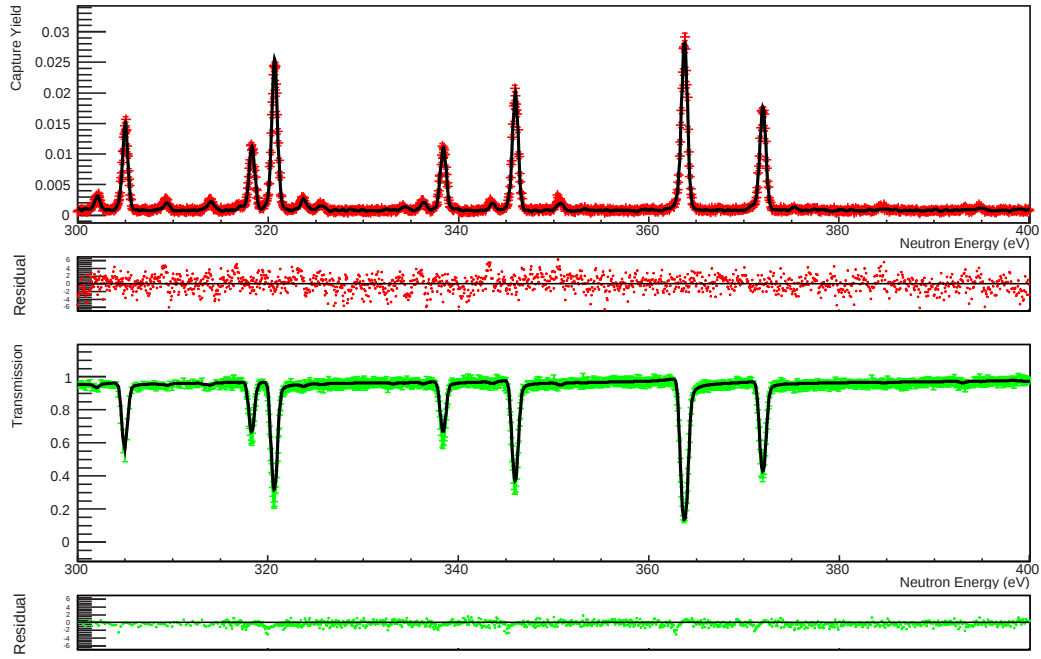


Figure 7.13: *SAMMY fits to capture and transmission between 300 eV and 400 eV.*

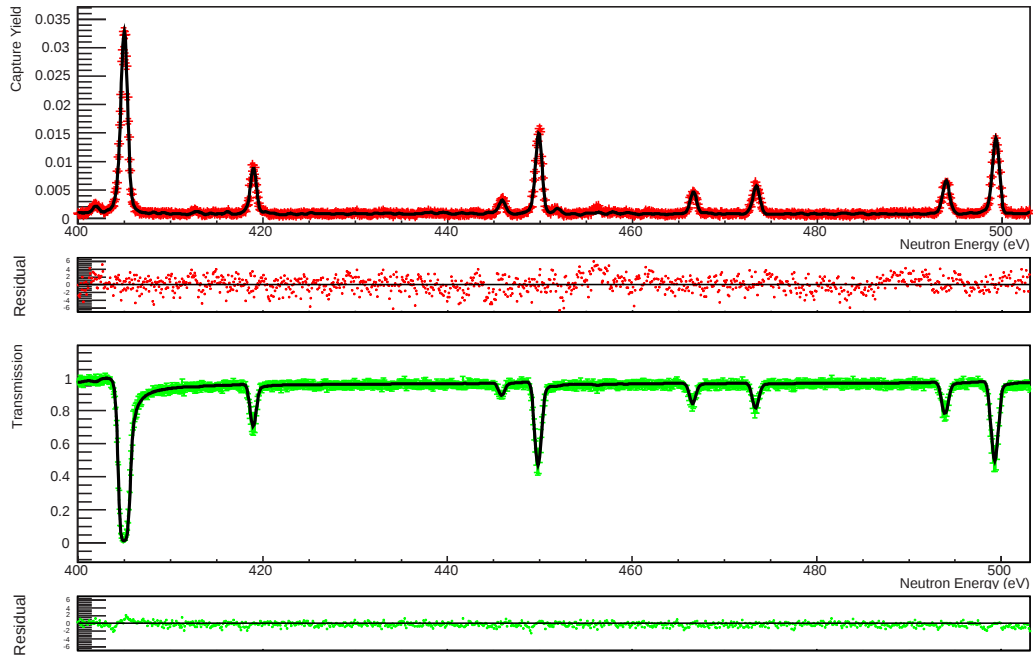


Figure 7.14: *SAMMY fits to capture and transmission between 400 eV and 500 eV.*

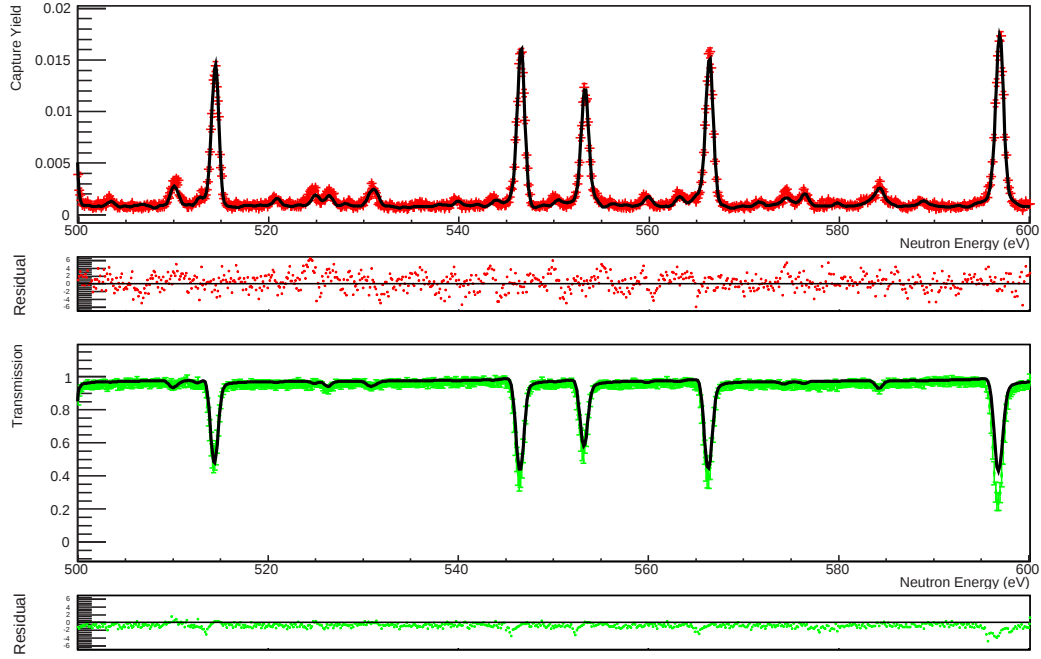


Figure 7.15: *SAMMY fits to capture and transmission between 500 eV and 600 eV.*

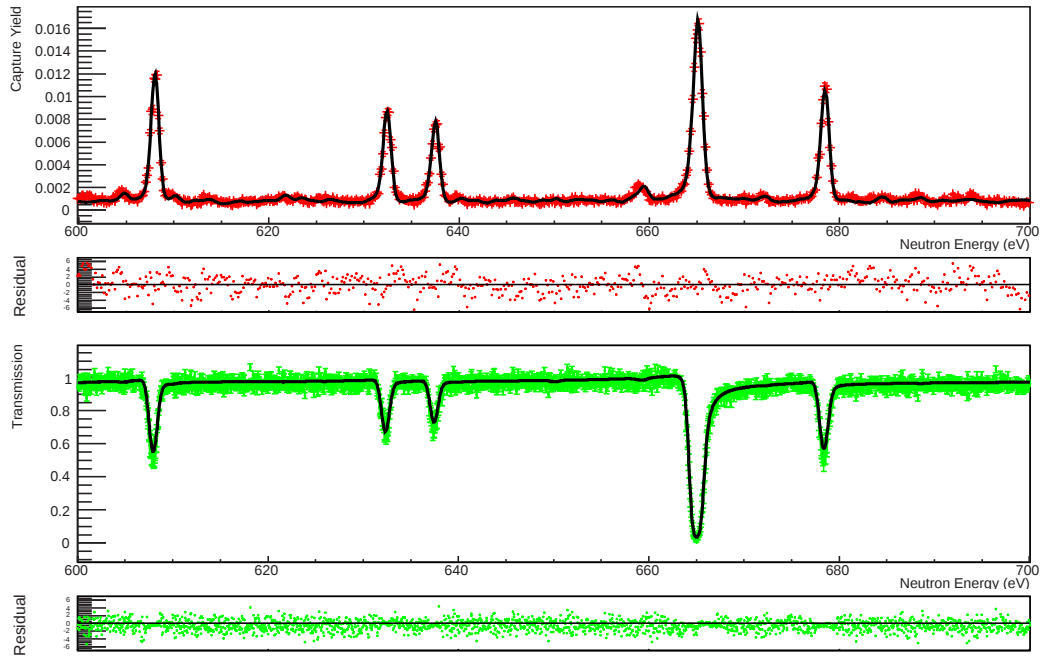


Figure 7.16: *SAMMY fits to capture and transmission between 600 eV and 700 eV.*

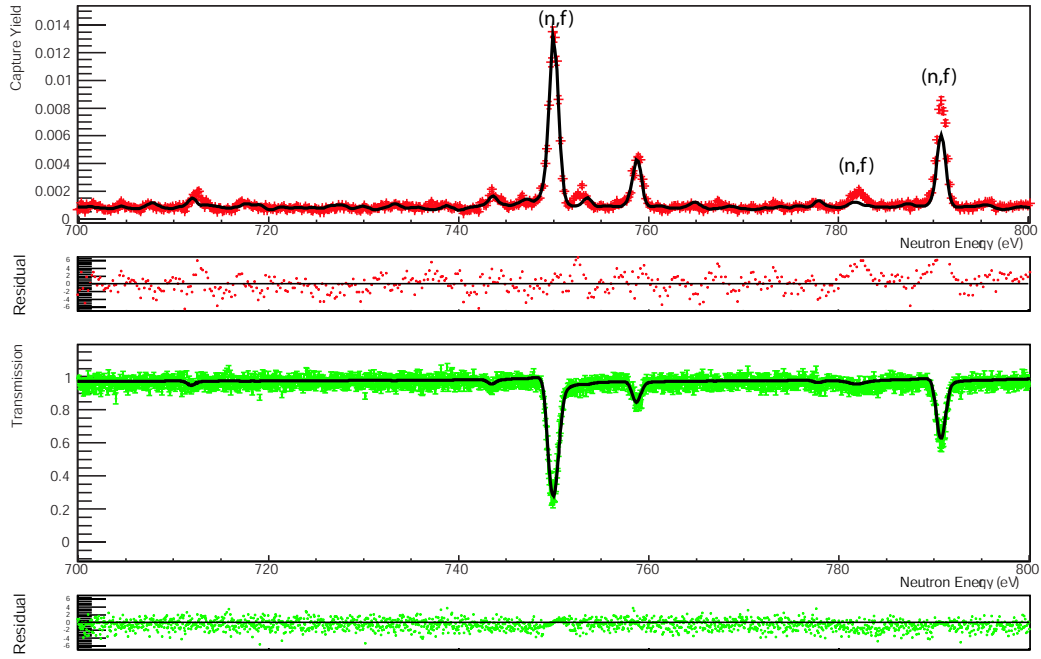


Figure 7.17: *SAMMY fits to capture and transmission between 700 eV and 800 eV.*

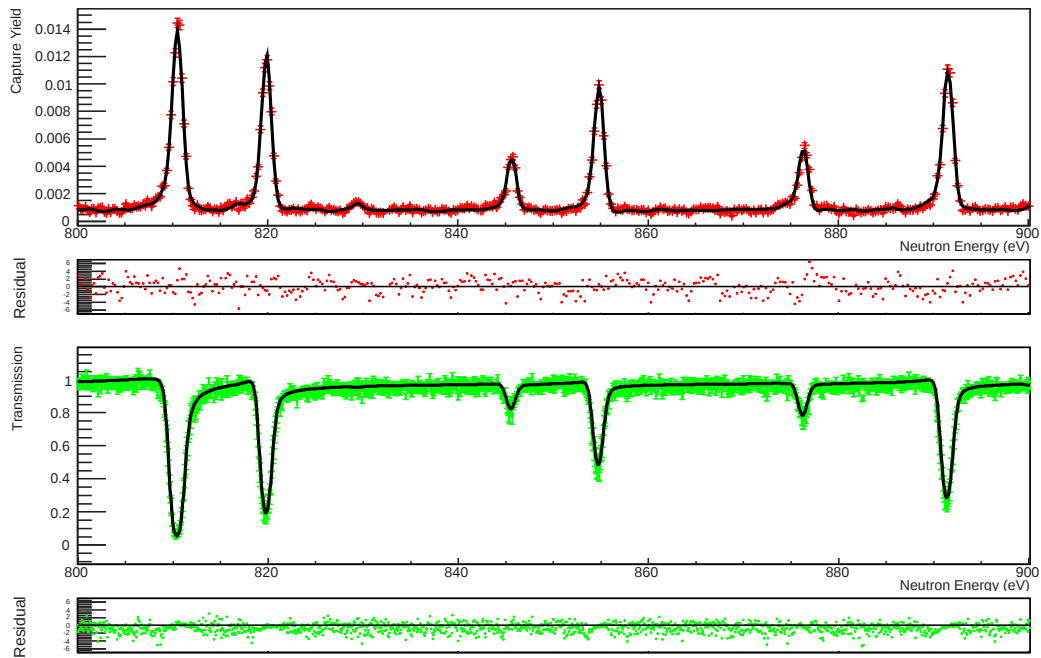


Figure 7.18: *SAMMY fits to capture and transmission between 800 eV and 900 eV.*

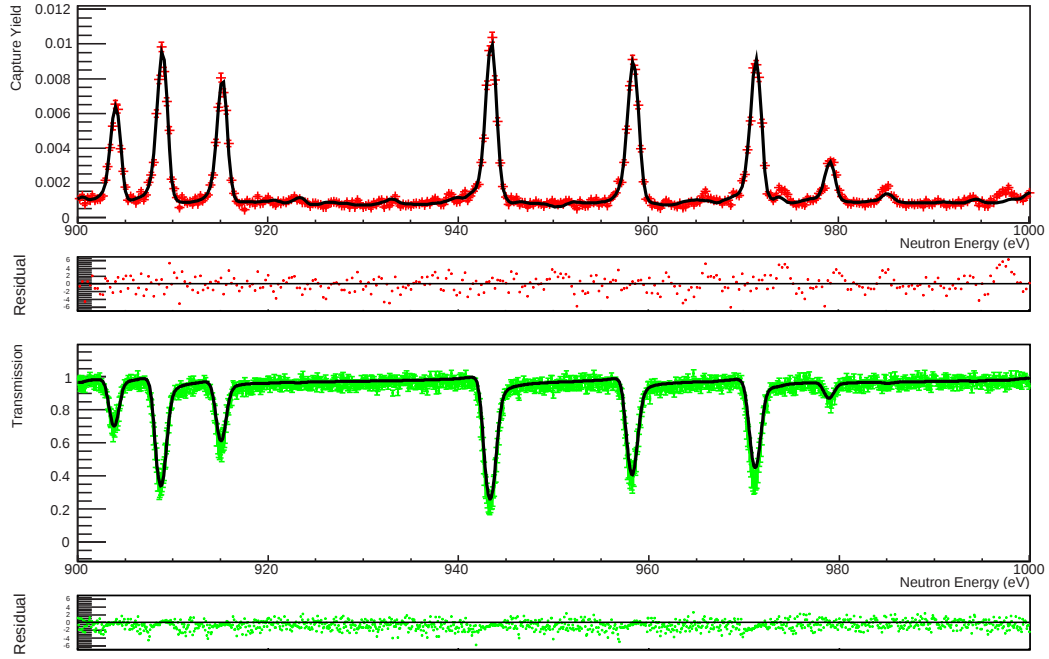


Figure 7.19: *SAMMY fits to capture and transmission between 900 eV and 1000 eV.*

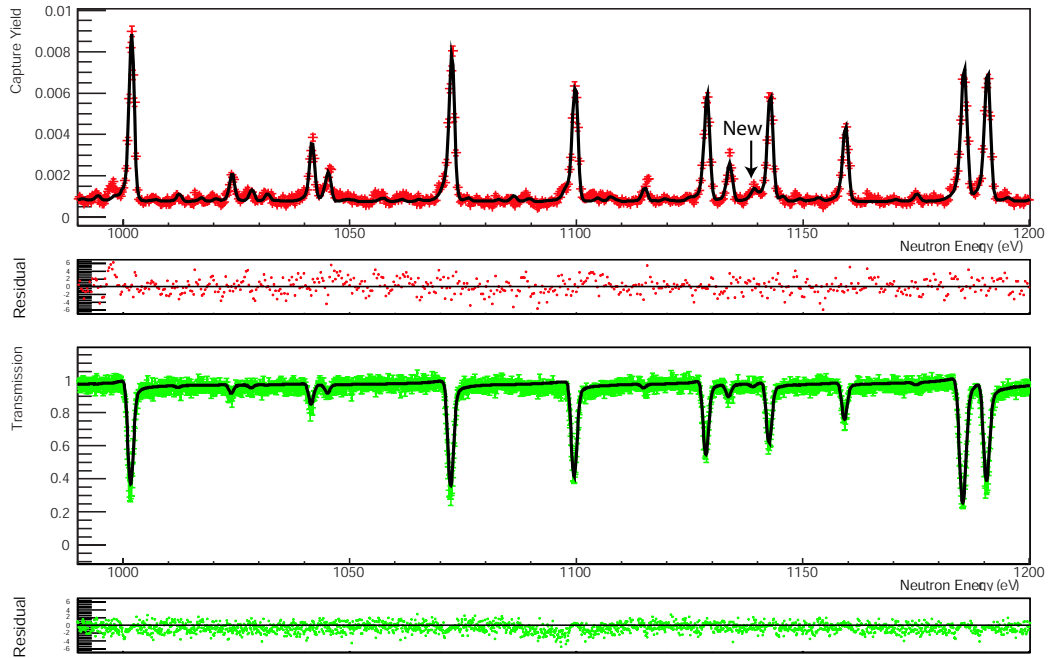


Figure 7.20: *SAMMY fits to capture and transmission between 1000 eV and 1200 eV.*

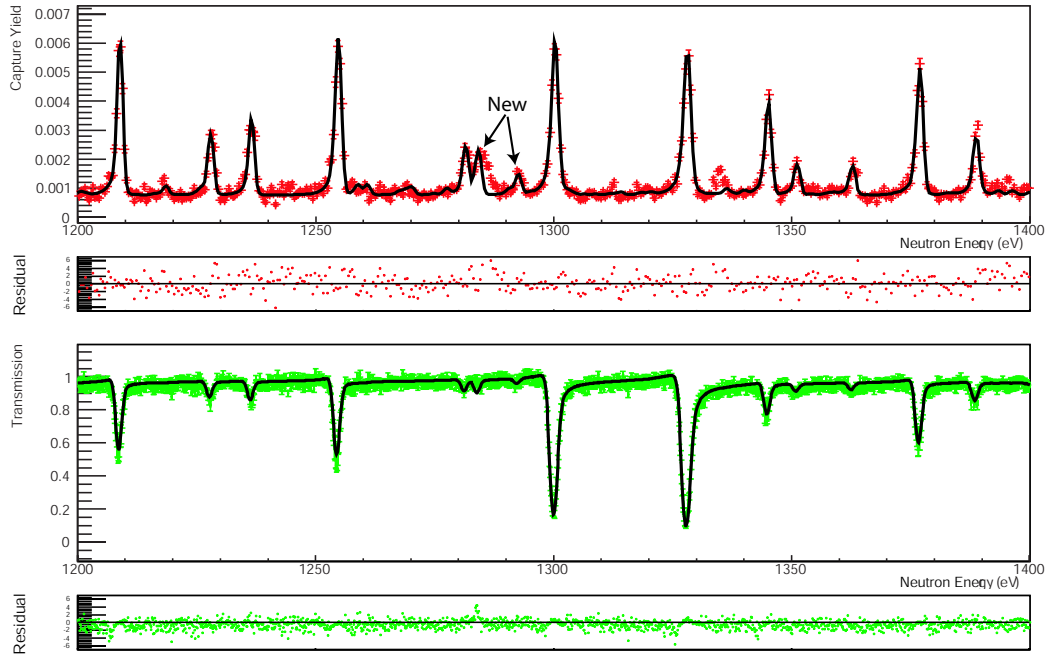


Figure 7.21: *SAMMY fits to capture and transmission between 1200 eV and 1400 eV.*

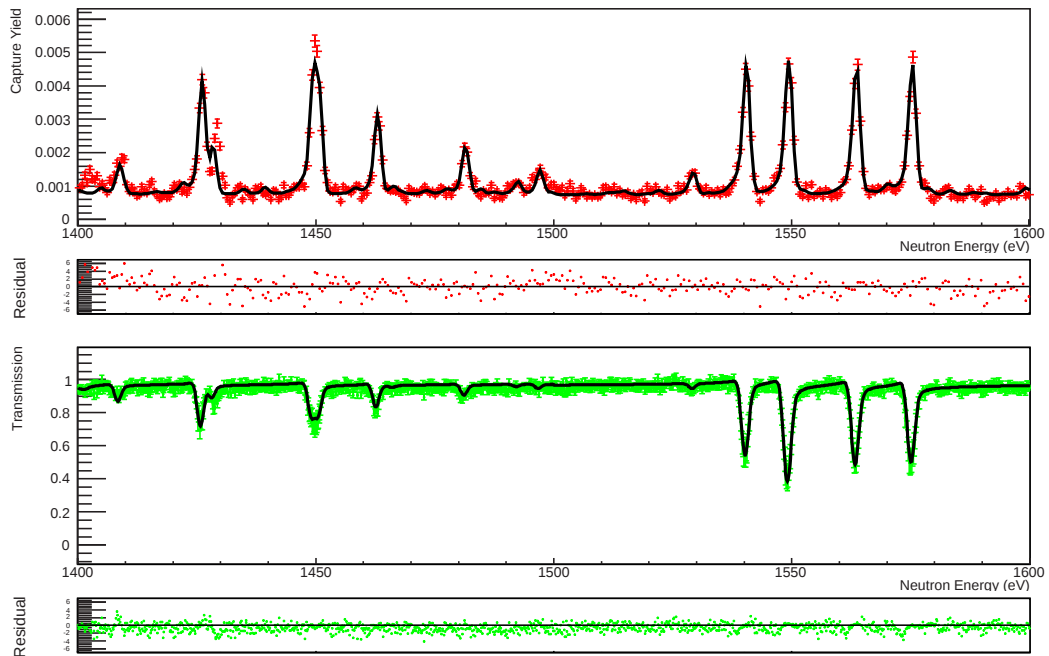


Figure 7.22: *SAMMY fits to capture and transmission between 1400 eV and 1600 eV.*

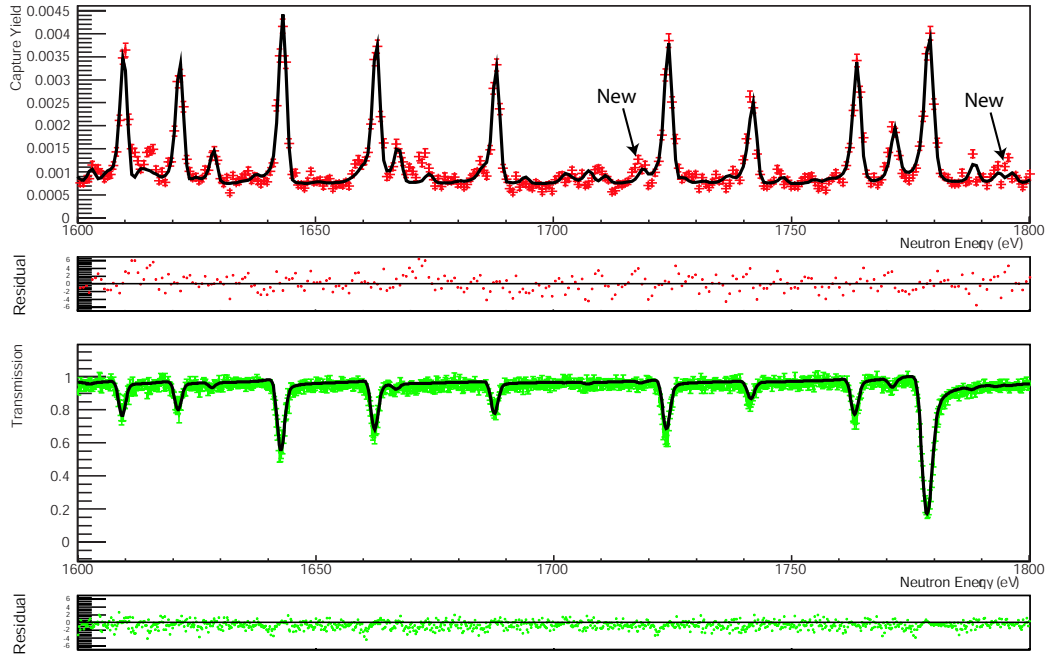


Figure 7.23: SAMMY fits to capture and transmission between 1600 eV and 1800 eV.

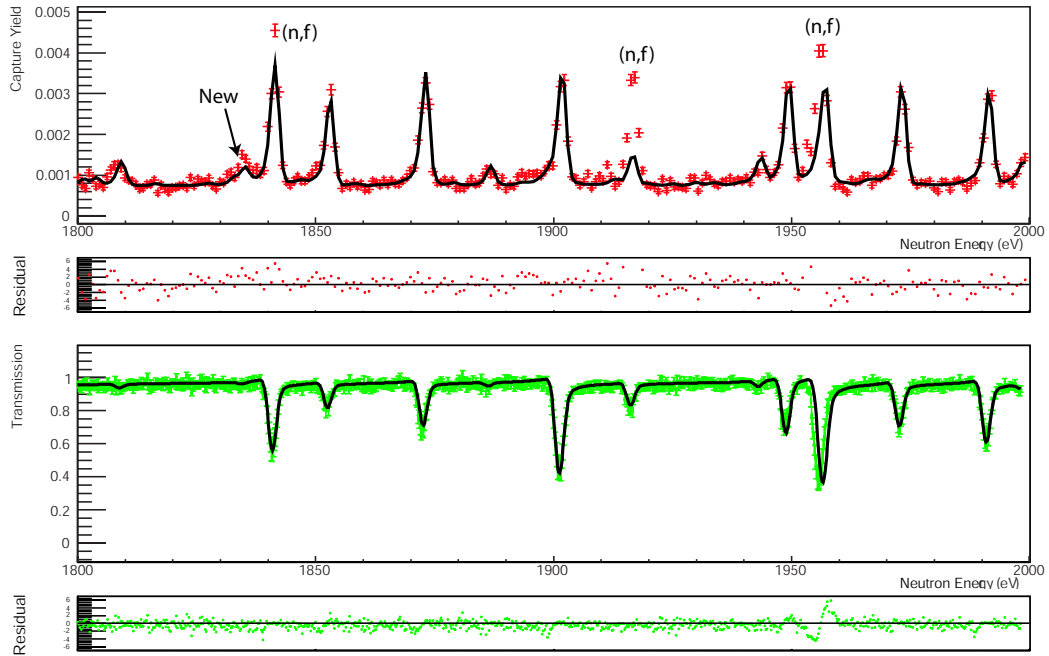


Figure 7.24: SAMMY fits to capture and transmission between 1800 eV and 2000 eV.

7.3.2.1 Correlation between resonance parameters

The correlation between the resonance parameters (RP) has been studied in the complete energy range. The correlations are null for most of the neighbouring resonances, and very small (<0.05) in some specific cases. This could be expected since all the resonances are well isolated, which was not the case for ^{237}Np cross section discussed in Chapter 6. The situation is different concerning the parameters within each resonance: when the neutron and capture widths are comparable the correlation is sizable and can range up to -0.9 , whereas it is again negligible if one of the widths is significantly larger than the other

The full set of resonance parameters and the associated covariance matrix will be available in the EXFOR database. In the mean time, the resonance parameters resulting from the analysis are listed in ENDF format [20] in Appendix C.

7.3.2.2 Resonances with small strength

The number of resonances considered between 110 eV and 2 keV is different from evaluation to evaluation. For instance, the JEFF-3.1 and JENDL-3.3 evaluations contain 158 resonances while only 119 are considered in ENDF/B-VII. The missing 39 resonances were proposed by Bouland et al. from a careful investigation of the transmission data of Kolar et al. However, in most cases the resonances in the transmission data were so small that they could only be guessed.

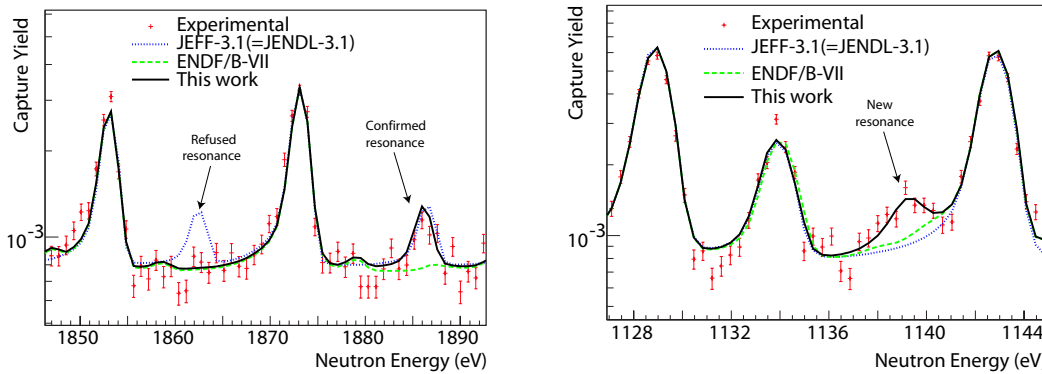


Figure 7.25: Comparison of the experimental capture yield and the calculations from JEFF-3.1(=JENDL-3.1) and ENDF/B-VII for some small resonances in ^{240}Pu . Left: the present data allow confirming or refusing the predicted resonances. Right: some small resonances have been observed for the first time in the present measurement.

The high statistics of the present capture data provides the means for confirming or refusing the existence of some of these weak resonances and to improve the accuracy of these resonance

parameters. Figure 7.25 illustrates some of the situations that have been found in the comparison of the evaluations with the experimental data for small resonances. In the energy region shown in the left panel, the JEFF-3.1 cross section includes two resonances that were not observed in previous evaluations (ENDFB/B-VII). In this case, the high statistics of the present data allows one to confirm the existence of one of the resonances and to reject the other. The case shown in the right panel illustrates one of the resonances that have been observed for the first time in this work.

Overall, from the 39 resonances proposed by Bouland et al., 18 were confirmed, 14 were rejected and 7 can neither be confirmed nor rejected because they are either very small or they fall very close to a strong ^{239}Pu resonance. These doubtful resonances are accepted in the present analysis. In addition, a total of 6 new resonances have been observed for the first time.

7.3.2.3 Resonances with a large fission to capture ratio

The efficiency of the TAC for detecting fission events is lower but comparable to that for detecting capture events. Hence, the resonant background caused by fission reactions becomes significant for resonances with a sizable fission cross section.

It has been found that the fission background becomes sizable and affects the accuracy of the analysis for those resonances with a fission to capture ratio larger than ~ 0.15 . In the energy region of interest there are a total of 25 of these resonances, but only eight of them contribute significantly to the overall capture cross section. These resonances are listed in Table 7.5, where their radiative and fission widths as well as their fission over capture cross section ratios are given. All of them are clearly identified in Figures 7.11 to 7.24.

Energy (eV)	Γ_γ	Γ_f	σ_f/σ_γ
750.06	32.4	11.3	0.35
782.44	31.2	1858	60
790.84	23.1	13.9	0.60
810.54	37.2	13.9	0.37
1426	29.8	5.48	0.18
1841.5	33.1	10.3	0.31
1916.6	31.8	95.6	3.0
1956.4	30.8	24.3	0.79

Table 7.5: *Prominent resonances of ^{240}Pu with a sizable fission over capture ratio.*

Figure 7.26 illustrates the effect of a large fission contribution to the experimental capture yield. The points and the black line correspond to the experimental and the expected capture yield, respectively. The difference between both gives a qualitative idea of the magnitude of the resonant background caused by fission.

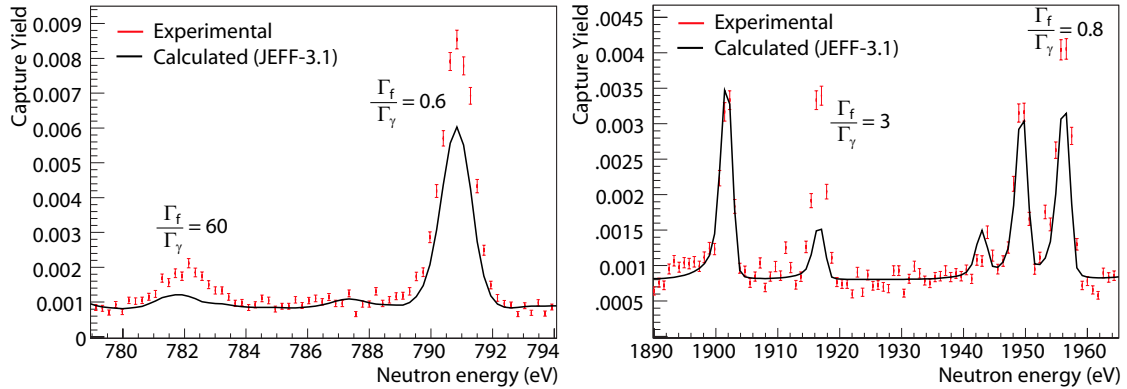


Figure 7.26: *Experimental and expected capture yield for a set of resonances with a large fission contribution: 782 eV, 790 eV, 1916 eV and 1956 eV (see Table 7.5). The difference between both gives a qualitative idea of the magnitude of the resonant background caused by fission.*

In the future measurements with the TAC the fission background should not be an issue because it is foreseen to use a sample equipped with a fission tagging system for identifying the fission reactions (see Section 8.3).

7.3.3 Statistical properties of the resonance parameters

The consistency of the resonance parameters resulting from the analysis of the RRR has been confirmed by means of a statistical analysis. Such an analysis provides a set of average resonance parameters that can be used to calculate the neutron cross sections at higher energies using the Hauser-Feshbach statistical model and the Optical Potential Model.

7.3.3.1 Average level spacing and probability distribution

The level spacing is calculated as the mean distance between next neighbouring resonances. It is given by the inverse of the slope of the cumulative distribution of levels as a function of neutron energy. This distribution is shown in the left panel in Figure 7.27 where the slope is $\rho=0.0794 \text{ eV}^{-1}$ and hence the average level spacing is

$$D_0 = 12.6 \pm 0.6 \text{ eV}. \quad (7.4)$$

The resonances suffering from strong self-shielding or large fission to capture contribution are not excluded from the calculation since their energy can be determined accurately.

In Table 7.6 the present level spacing is compared to the different evaluations. Bouland et al. (JEFF-3.1 and JENDL-3.3) recommend a value of $12.06 \pm 0.05 \text{ eV}$ while Weston et al. (ENDF/B-VII and Mughabghab) recommend a value of $13.6 \pm 0.7 \text{ eV}$. The present value

$D_0=12.6\pm0.6$ eV falls between these values, with differences that are at the limit of the estimated uncertainties.

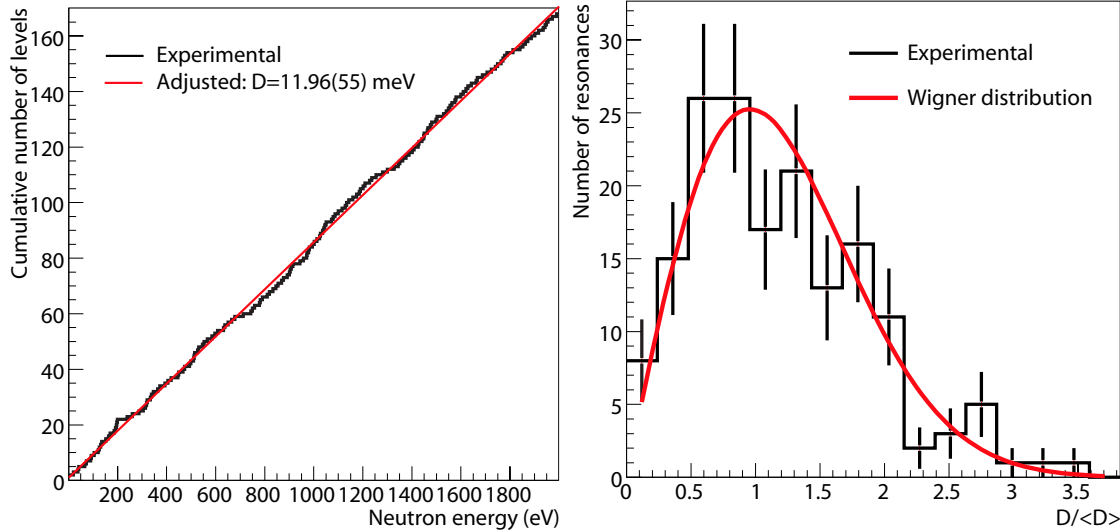


Figure 7.27: *Left: Number N of resonances identified below E as a function of E . Right: Experimental and theoretical distributions of next neighbour resonance distances.*

The Wigner law can be used as a consistency check for the distance distribution of neighbouring levels:

$$p(x)dx = \frac{\pi x}{2} \exp\left(-\frac{\pi x^2}{4}\right) dx \quad \text{with } x = \frac{D_i}{\langle D_0 \rangle}. \quad (7.5)$$

The comparison in the right panel of Figure 7.27 shows that the experimental distribution follows the Wigner distribution reasonably well.

		JEFF-3.1	ENDF/B-VII	
	This work	JENDL-3.3	Mughabghab	RIPL-2
		Bouland et al.	Weston et al.	
D_0 (eV)	12.6 ± 0.6	12.06 ± 0.05	13.6 ± 0.7	12.4 ± 0.7

Table 7.6: *Average level spacing from this work and those of the recent evaluations.*

7.3.3.2 Radiative width

The radiative widths for the 40 most prominent resonances are shown in Figure 7.28, where the solid line corresponds to the average value $\langle \Gamma_\gamma \rangle = 31.0 \pm 1.5$ meV. This value is in agreement within uncertainties with the values found in previous works, which are summarized in Table 7.8.

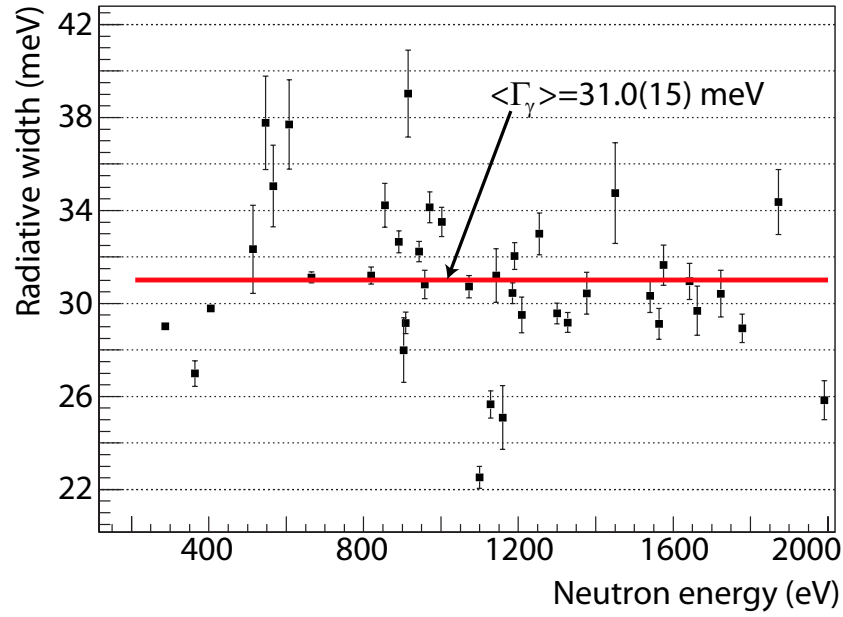


Figure 7.28: Radiative widths of the 40 most prominent resonances between 110 eV and 2 keV.

In order to identify a possible non-statistical behaviour of the radiative width, average values have been determined in several energy intervals. The results are summarized in Table 7.7 and confirm a constant behaviour of the radiative width at all energies except in the energy interval between 500 eV and 750 eV. This could be related to the fission clusters that exist in this interval.

Energy (keV)	$\langle \Gamma_\gamma \rangle$ (meV)
0.11 - 0.25	-
0.25 - 0.50	28.6
0.50 - 0.75	34.8
0.75 - 1.00	32.4
1.00 - 1.25	29.0
1.25 - 1.50	31.4
1.50 - 1.75	30.4
1.75 - 2.00	29.7
0.11 - 2.0	31.0 ± 1.5

Table 7.7: Average radiation widths $\langle \Gamma_\gamma \rangle$ calculated in 250 eV wide energy intervals.

	This work	JEFF-3.1 JENDL-3.3 Bouland et al.	ENDF/B-VII Mughabghab Weston et al.	RIPL-2
$\langle\Gamma_\gamma\rangle$ (meV)	31.0 ± 1.5	31.8 ± 1.5	30.6 ± 2	30.7 ± 2.5

Table 7.8: Average radiation widths from this work and from the evaluations and previous works.

7.3.3.3 Neutron strength function

The neutron strength function S_0 is related to the average total cross section and is defined as the ratio of the average values of the reduced width and the level spacing:

$$S_0 = \frac{\langle g\Gamma_n^0 \rangle}{\langle D_0^{J=2,3} \rangle} = \frac{\Sigma_{E}^{E+\Delta E} g\Gamma_n^0}{\Delta E}, \quad (7.6)$$

where the reduced neutron width Γ_n^0 is defined as $\Gamma_n^0 = \Gamma_n/\sqrt{E}$, in the case of s -wave resonances. The definition indicates that S_0 can be calculated as the slope of the cumulative distribution of $g\Gamma_n^0$ as a function of neutron energy. This distribution is shown in black in Figure 7.29, where the red line corresponds to a linear fit between 500 eV and 2 keV that yields the value:

$$S_0 = (1.08 \pm 0.02) \cdot 10^{-4}. \quad (7.7)$$

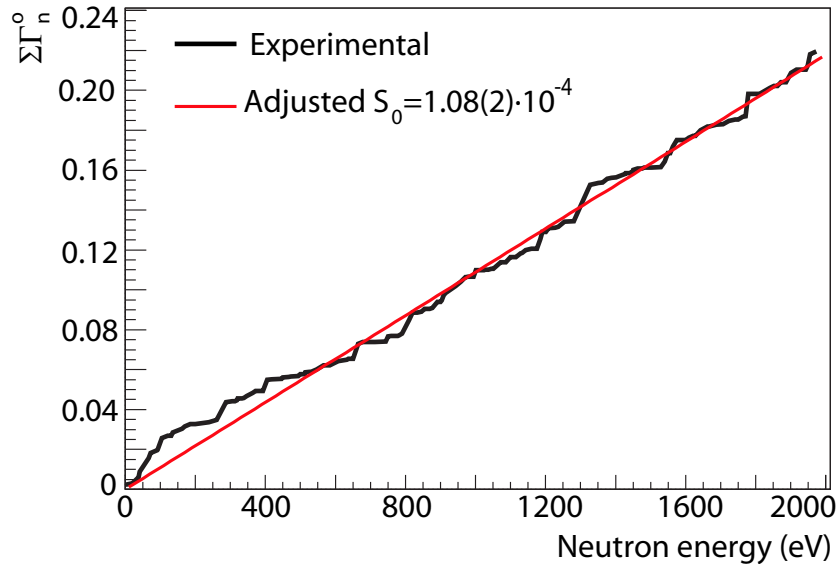


Figure 7.29: Cumulative sum of the reduced neutron widths as a function of neutron energy. The red line corresponds to a linear fit with a slope of $1.08(2) \cdot 10^{-4}$.

$E_n(keV)$	This work	JEFF-3.1	ENDF/B-VII	RIPL-2
		JENDL-3.3 Bouland et al.	Mughabghab Weston et al.	
0.0 - 0.5	1.35	1.09	1.10	1.05
0.5 - 1.0	1.06	1.05	1.03	
1.0 - 1.5	1.09	1.02	1.01	
1.5 - 2.0	1.08	1.22	1.17	
2.0 - 4.0	-	0.85	0.75	
4.0 - 5.7	-	1.15	0.97	
0.5 - 2.0	1.08±0.02	1.10±0.11	1.07±0.08	1.05±0.10

Table 7.9: Neutron strength functions from this work and from recent evaluations.

Table 7.9 summarizes the value of the neutron strength function of this work and of the recent evaluations in different energy intervals. Neglecting the lowest energy interval where the present results are affected by the non-uniform distribution of the mass in the sample, the strength function found in this work remains constant at all energies, as expected. The values calculated between 500 eV and 2 keV are all in agreement. However, the strength function values of the evaluations are not really constant. Therefore the corresponding average values suffer from uncertainties larger than the that of the present result. The more accurate values obtained in the analysis of the present capture data indicates that a new capture measurement above 2 keV could solve the discrepancies in the strength function at high neutron energies.

7.3.3.4 Neutron width distributions

The reduced neutron widths for a single J value should to follow the Porter-Thomas (P-T) distribution, i.e. a χ^2 distribution with only one degree of freedom ($\mu=1$):

$$P_{PT}(x) = \frac{1}{\sqrt{2\pi x}} e^{-x/2} \quad , \quad (7.8)$$

with $x = \Gamma_n^0 / \langle \Gamma_n^0 \rangle$. However, the comparison between the experimental data and the expected P-T distribution is usually illustrated from the integral of the P-T distribution as a function of x_i :

$$N(x_i) = N_0 \int_{x_i}^{\infty} P_{PT}(x) dx = N_0 (1 - \text{erf} \sqrt{x_i/2}) \quad (7.9)$$

where N_0 is the total number of resonances.

Figure 7.30 shows the experimental and calculated number of resonances with $\Gamma_n^0 > x_i \langle \Gamma_n^0 \rangle$ as a function of x_i . The number of resonances N_0^J has been calculated from the partial level spacing values summarized in Table 7.6. The excellent agreement between the experimental and expected distribution confirms the consistency of the resonance parameters in the energy region between 110 eV and 2 keV.

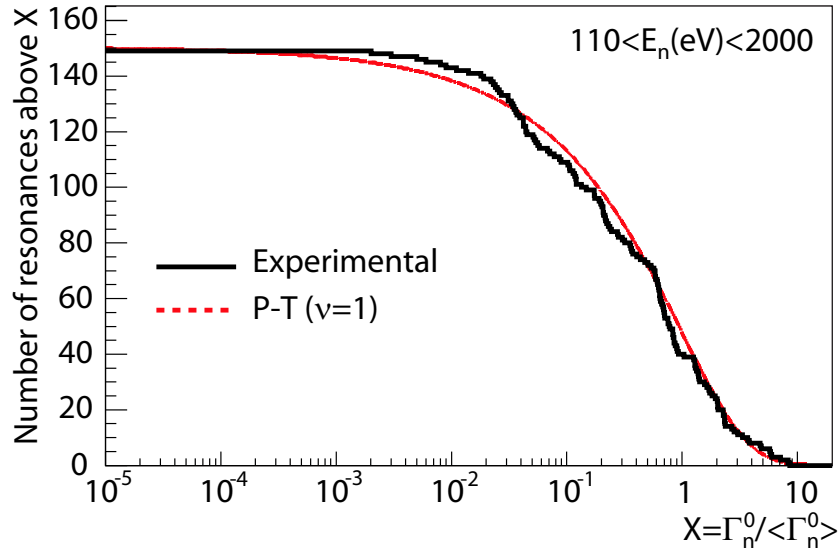


Figure 7.30: Comparison between the experimental and expected integral distributions of the neutron widths.

7.4 Comparison with previous measurements and the evaluated cross sections

The result of the resonance analysis between 110 eV and 2 keV is a complete set of resonance parameters for 158 resonances and the associated covariance matrix. An illustrative way of comparing the results of the present work with the evaluations is by means of average cross section values. During the discussion that follows it should be noted that in this energy range the JENDL-3.3 cross sections are equal to those given by JEFF-3.1 and Bouland et al., and the ENDF/B-VII evaluation corresponds to the data of Mughabghab and of Weston et al.

The capture cross section obtained in this work and those of the evaluated libraries have been averaged in the 14 energy intervals of Figures 7.11 to 7.24. The averaging has been performed with NJOY-99 [149] from the different sets of resonance parameters and the results are given in Table 7.10. The table also provides the ratios, that are shown in Figure 7.31, of the average capture cross sections of the evaluations with respect to the results of this work. Values in bold indicate that these values differ more than the uncertainties of the present results.

The results of this work are compatible within 5% with the JEFF-3.1 and JENDL-3.3 evaluations, except in the energy interval between 300 eV and 400 eV where these evaluations are 10% larger. On the contrary, the comparison with ENDF/B-VII, based on the analysis of Weston et al., reveals significant discrepancies. As in the case of the JEFF-3.1 and JENDL-3.3 evaluations, it is observed that the ENDF/B-VII evaluation is 10% larger than that of this work between 300 eV and 400 eV and the largest difference (22%) is found between 700 eV and 800 eV, where there is only one prominent resonance that has indeed large scattering and fission cross sections.

At high energies, between 1.4 keV and 2 keV, the ENDF/B-VII capture cross section is between 5% and 15% larger than that of this work, with an average difference of 10%. This difference is well beyond the uncertainty of the present results: 4.4% at those energies.

E_1-E_2 (eV)	$\frac{1}{\Delta E} \int \sigma_\gamma dE$	$\frac{JEFF-3.1^1}{This\ work}$	$\frac{ENDF/B-VII^2}{This\ work}$
110-200	17.7 ± 1.1	1.03	1.00
200-300	7.10 ± 0.47	1.02	1.04
300-400	7.22 ± 0.39	1.10	1.09
400-500	5.97 ± 0.30	1.01	1.00
500-600	6.03 ± 0.31	1.03	0.98
600-700	4.69 ± 0.24	0.95	0.99
700-800	2.11 ± 0.11	0.97	0.78
800-900	5.63 ± 0.29	1.01	0.93
900-1000	5.62 ± 0.28	1.02	0.97
1000-1200	3.68 ± 0.19	1.05	0.97
1200-1400	2.74 ± 0.14	1.03	1.00
1400-1600	2.63 ± 0.13	1.02	0.85
1600-1800	2.44 ± 0.12	1.04	0.91
1800-2000	2.21 ± 0.11	1.02	0.95

Table 7.10: Average capture cross section of ^{240}Pu from 110 eV to 2 keV calculated with NJOY-99 and compared to the values from previous measurements and evaluations. Values in bold indicate that the difference exceeds the uncertainty of the present results.

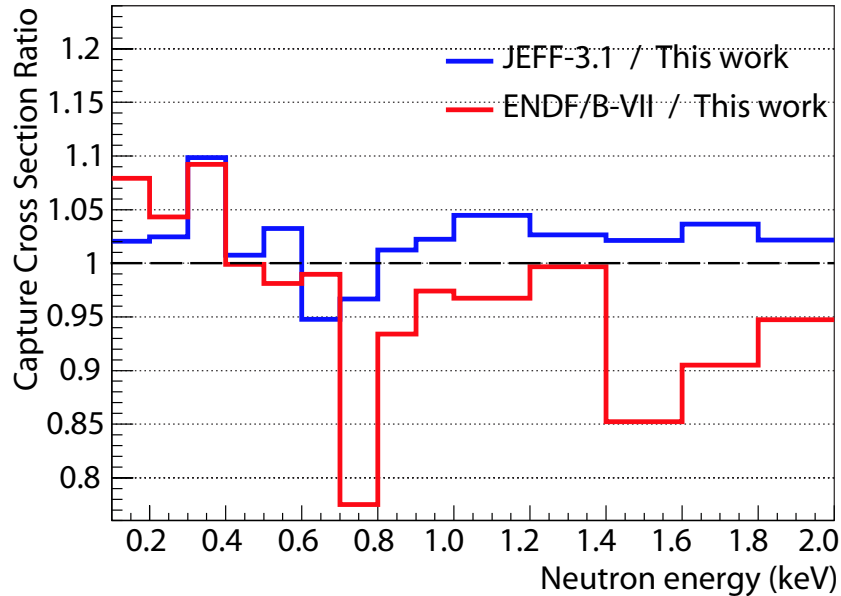


Figure 7.31: Average capture cross section ratios of the evaluations with respect to the present work. The exact values are given in Table 7.10.

7.5 Summary and conclusions

The neutron capture cross section of ^{240}Pu has been measured at the CERN/n_TOF facility in the energy range between 1 eV and 2 keV, which is within the resolved resonance region of ^{240}Pu .

The actinide material PuO_2 in the form of powder was sandwiched between two aluminium foils and then encapsulated in a titanium can to fulfil the *ISO-2919 norm* requested by the safety authorities at CERN. The isotopic composition of the sample was determined with an accuracy of 5% by means of γ spectroscopy at CERN. The results given by this analysis were fine tuned by a combined analysis of the capture data with the most reliable transmission measurement in the high energy region, where the experimental capture yield becomes directly proportional to the isotopic abundance of ^{240}Pu in the sample, yielding an improved isotopic abundance of ^{240}Pu of $84\pm 2\%$.

The capture yield was calculated from the measured counting rate with the main corrections calculated as follows:

- The detection efficiency and the effect of the dead-time losses have been determined from MC simulations. The accuracy in the resulting detection efficiency was better than 3% and the dead-time losses were found to be small at low energies and negligible above a few hundred eV.
- The normalization of the capture yield has been determined from the Saturated Resonance Method applied to the saturated resonances of ^{197}Au and ^{240}Pu at 4.9 eV and 1.06 eV, respectively. Both normalization values were in agreement within uncertainties, giving confidence to the applied data reduction techniques applied: calculation of the detection efficiency, determination of the neutron scattering background, etc. The average normalization factor, with an accuracy of better than 2%, was used for the analysis.

The present measurement describes the (n,γ) cross section of ^{240}Pu in the resolved resonance region with the highest resolution and accuracy to date. The accuracy of the capture yield was 6% below 300 eV and 4.6% at higher energies. Below 110 eV, the calculation of the capture yield was largely affected by self-shielding effects that could not be determined accurately due to the grain structure of the powder sample. For this reason the energy region between 1 eV and 110 eV was not included in the analysis.

The energy region between 110 eV and 2 keV, 158 ^{240}Pu resonances have been analyzed. The combined resonance analysis of the present capture data together with the transmission data of Kolar et al. showed that both data sets are compatible in the complete energy range. The analysis of the most prominent resonances provided accurate values of all resonance parameters, while the average radiative width was kept fixed for the weaker resonances.

The sensitivity of the TAC to fission reactions introduces some difficulties to the analysis of

those resonances with a sizable fission cross section. The limit for an accurate resonance analysis was found at $\sigma_f/\sigma_\gamma=0.15$, and therefore the eight prominent resonances with such a large fission cross section were not analyzed.

The consistency of the resonance parameters has been confirmed from their statistical analysis, which also allowed us to provide a set of average parameters for the calculation of high energy cross sections. These are given in Table 7.11, where the values recommended in the recent evaluations are included for comparison. The analysis revealed that:

- The large number of weak resonances that were proposed by Bouland et al. have been investigated in order to confirm or to reject them. Overall, among the 39 resonances proposed by Bouland et al., 18 have been confirmed, 14 have been rejected and 7 can neither be confirmed nor refused. In addition, a total of 6 new resonances have been observed for first time. The corresponding value of the level spacing is 12.6 ± 6 eV, which is in between the values of the recent evaluations: 12.06 eV (JEFF-3.1 and JENDL-3.3) and 13.6 eV (ENDF/B-VII).
- The radiative width was found to follow the expected constant behaviour except in a reduced energy interval where sub-threshold fission is large. The average value calculated from the 40 most prominent capture resonances $\langle\Gamma_\gamma\rangle=31.0$ meV is in between the values found in the different evaluations and is compatible within uncertainties.
- The strength function found in this work, $S_0=1.08(2)\cdot 10^{-4}$, is very close to those of previous evaluations and measurements. However, looking at the local values in energy intervals of 500 eV, the present strength function follows the expected constant behaviour while all previous values show large deviations between the values below and above 1.5 keV.

		JEFF-3.1	ENDF/B-VII	
	This work	JENDL-3.3 and Bouland et al.	Mughabghab Weston et al.	RIPL-2
Level spacing (eV)	12.6 ± 0.6	12.06 ± 0.06	13.6 ± 0.7	12.4 ± 0.7
$\langle\Gamma_\gamma\rangle$ (meV)	31.0 ± 1.5	31.8 ± 1.5	30.6 ± 2	30.7 ± 2.5
Strength function ($\cdot 10^{-4}$)	1.08 ± 0.02	1.03 ± 0.07	0.93 ± 0.08	1.05 ± 0.10

Table 7.11: *Average resonance parameters of ^{240}Pu determined in this work compared to previous values.*

The capture cross section measured and analyzed in this work has been averaged in 14 energy ranges using NJOY-99 for comparison with the evaluations. Below 700 eV, the present capture cross section is in good agreement with the JEFF-3.1, JENDL-3.3 and ENDF/B-VII evaluations. At higher energies, where there are significant differences between the recent evaluations, the present results are in good agreement with JEFF-3.1 and JENDL-3.3. The significant differences with respect to the ENDF/B-VII evaluation indicate that there might be a problem in the analysis of the transmission data of Kolar et al. by Weston et al., which is the basis of that evaluation.

The results of this work give confidence in the JEFF-3.1 and JENDL-3.3 evaluations and indicate that the ENDF/B-VII values should be revised. In view of the present results, the uncertainty of 10% in the capture cross section of ^{240}Pu seems overestimated and a more realistic uncertainty of only 5% can be safely assumed if the present data are included in future evaluations.

Part IV

SUMMARY AND CONCLUSIONS

Chapter 8

Summary and Conclusions

This manuscript has been devoted to the discussion of the first measurements of the capture cross section of minor actinides ^{237}Np and ^{240}Pu carried out at the CERN n_TOF facility in 2004. The aim of the measurement was to provide the nuclear data community a set of accurate experimental data with a good control of systematic uncertainties and correspondingly realistic estimates of their overall accuracy. This issue is crucial for the work of the evaluators that combine the available measurements by weighting each of them by their reliability and accuracy. The work presented in this manuscript is not restricted to the measurement and data reduction but reports careful analysis of the cross sections from the measured capture yields. The analysis include the reliable transmission data in order to derive the capture cross section of each isotope together with a set of parameters for individual resonances as well as the associated covariance matrices.

During the measurements, the data reduction and the cross section analysis, special attention has been paid to control and to determine all possible sources of systematic uncertainty.

8.1 Experimental set-up and data reduction techniques

The measurements have been performed at the CERN n_TOF facility using the Total Absorption Calorimeter, which offer an excellent combination of features for the accurate measurement of radioactive isotopes. The n_TOF facility has a large flight path (185 m) and the highest instantaneous neutron flux among the neutron time-of-flight facilities worldwide. In the past, the facility has proved its excellent capabilities concerning the measurement of both capture and fission cross sections.

The Total Absorption Calorimeter (TAC) is a 4π detector made of 40 BaF_2 crystals that provides an efficiency close to 100% for detecting capture cascades. Its segmentation and high total absorption efficiency are powerful tools for maximizing the capture to background ratio by

the selection of events based on their deposited energy and crystal multiplicity. The design of the TAC minimizes the probability of detecting scattered neutrons by placing a neutron absorber around the samples and by the use of crystal capsules enriched in ^{10}B .

The experimental data presented in this manuscript correspond to the first measurements performed with the TAC. Hence, an important part of the work consisted in its characterization and the full understanding of its performance and capabilities. Accordingly, this work contains a detailed study of the energy and time resolution of the detector, the identification and characterization of the different sources of background and the techniques that allow their minimization, and the determination of its efficiency for detecting both capture and scattering reactions.

One of the main innovations of this work concerning the data reduction refers to the calculation of the detection efficiency of the TAC and the dead-time losses by means of Monte Carlo simulations:

- The Monte Carlo simulations have been validated with experimental data using simple decay schemes from standard γ -ray sources and also neutron capture measurements of ^{197}Au . The results have shown that the detection efficiency of the TAC can be determined with an accuracy of better than 3% for any combination of conditions on the deposited energy and multiplicity of the detected events.
- The model that has been designed and implemented for the calculation of the dead-time losses, related to the slow scintillation component of BaF_2 , considers the experimentally determined dead-time of each BaF_2 module in the simulation of events. Since the primary events for the simulation are generated with measured time-of-flight distributions it was possible to determine the detection efficiency accurately, including the neutron energy dependence introduced by the counting rate density. The results obtained with this model have been validated successfully with the measurement of the $^{197}\text{Au}(n,\gamma)$ cross section, in particular by using a set of resonances which covered a wide range of counting rates.

Furthermore, the comparison of the simulation with experimental data have been shown to be a powerful tool for the investigation of Photon Strength Functions, which are the main input of the model for the generation of realistic capture cascades as primary events of the simulation.

8.2 Measurement and analysis of the capture cross section of minor actinides

The neutron capture cross section of minor actinides have been measured using low mass oxide samples of 43.3 mg and 51.2 mg for ^{237}Np and ^{240}Pu , respectively. In both samples, the actinide material was deposited between aluminium foils and then encapsulated in sealed titanium to satisfy the *ISO-2919 norm* requested by the Safety Authorities at CERN.

The measurements on ^{237}Np and ^{240}Pu provided spectra with high statistics, well suited for the accurate analysis of the corresponding capture cross sections. In both cases, the measurements represent the highest resolution capture measurements to date.

The analysis of the experimental capture yields has been performed with the SAMMY code, adopting the Reich-Moore approximation in both the resolved and unresolved resonance region.

8.2.1 Measurement and analysis of ^{237}Np

The neutron capture cross section of ^{237}Np has been measured between 1 eV and 2 keV, which includes the complete region of resolved resonances and part of the unresolved resonance region. In this measurement the counting rates was kept low enough (<0.5 counts/ μs) so that the dead-time losses were negligible. Concerning the background related to scattered neutrons, the capture to scattering ratio is very large and therefore this background was also negligible.

The capture yield has been calculated using the detection efficiency determined by Monte Carlo simulations and the neutron intensity recorded by the ^6Li based neutron monitor *SiMon*. Two different normalizations have been studied: (i) using the Saturated Resonance Method applied to the 4.9 eV resonance in ^{197}Au , which was measured under exactly the same experimental conditions, (ii) normalizing the capture yield to the most recent and reliable transmission data. The results presented in this work correspond to the second option, which provides a more accurate capture yield and - most importantly- allows one to minimize the correlations between the resonance parameters by means of the combined analysis of both, capture and transmission data. However, the resulting capture cross section with one or the other normalization method have been shown to be compatible within $\pm 3\%$, giving confidence in the various techniques developed for the calculation of the capture yield.

In total, 557 resonances have been analyzed between 1 eV and 500 eV. The combined analysis of capture and transmission data has shown that both data sets are compatible in the complete RRR. The analysis of the energy region below 43 eV provided the values of the scattering radius R' and the average radiative width $\langle\Gamma_\gamma\rangle$ that are kept constant for the analysis of resonances at higher energies. The resulting covariance matrix indicates that a sizable correlation exists between the parameters of neighbouring resonances, which is in most cases limited to next neighbours. On the other hand, the correction parameters of normalization and background show a clear negative correlation, on average -0.25, with the capture and neutron width of most resonances. The consistency of the large number of resonance parameters was confirmed from their statistical analysis. This analysis provided a set of average resonance parameters and showed that the radiative width follows a constant behaviour within uncertainties, that the distribution of next neighbour resonance distances is in agreement with the expected Wigner distribution -even when resonances of different spins are analyzed separately-, and that the neutron strength function S_0 , as expected, does not show any dependence with the neutron energy.

The URR has been analyzed between 500 eV and 2 keV. Capture yield and transmission

were converted to capture and total cross section, respectively. The cross sections were analyzed with the FITACS code implemented in SAMMY. This code uses the Hauser-Feshbach theory with width fluctuations to calculate the total and partial cross sections from a set of average resonance parameters that can be fitted to the experimental data. The analysis has shown that the capture and transmission data are compatible and hence it was possible to find a set of average parameters $\langle\Gamma\rangle$, R' and S_0 , which agree within uncertainties with those obtained from the statistical analysis in the RRR.

The combination of the results from the resolved and unresolved resonance regions provide a set of recommended values for the statistical description of the cross section that is compared in Table 8.1 to the values of different evaluated libraries. The value of R' has been derived from the analysis of transmission data in the energy region between resonances.

	This work	JEFF-3.1 JENDL-3.3 ENDF/B-VII	RIPL-2
D_0 (eV)	0.56 ± 0.02	0.58	0.57 ± 0.03
$\langle\Gamma_\gamma\rangle$ (meV)	40.8 ± 1.8	40.0	40.8 ± 1.2
S_0 ($^{-4}$)	1.03 ± 0.08	1.0	0.97 ± 0.07
R' (fm)	9.8 ± 0.6	10.5	-

Table 8.1: Average resonance parameters of ^{237}Np determined in this work compared to previous values.

The capture cross section of ^{237}Np obtained in this work is compared in Figure 8.1 with previous measurements and evaluated data libraries. The left panel shows the ratio of average capture cross sections calculated with NJOY-99 from the resonance parameters. The right panel shows the capture cross section in the URR.

Overall, the results of this work are compatible within uncertainties with the most recent evaluated cross sections. There is also reasonable agreement with the data of Scherbakov et al. below 100 eV and Weston et al. in the complete energy range. The comparison with other published results reveals significant differences beyond their quoted uncertainties. This is the case for the data of Kobayashi et al., which show large differences with respect to the present results in both, the resolved and unresolved resonance region, and also for the data of Esch et al., which are agree with the present results around 500 eV but show differences as large as -25% at higher neutron energies.

To summarize, this work has provided a set experimental data for the capture cross section of ^{237}Np in the energy region between 1 eV and 2 keV. The capture yield was calculated paying special attention to the identification, reduction and estimation of the different sources of uncertainty, resulting in a capture cross section with an overall accuracy of better than 4%. The cross section analysis of the capture data includes the most recent and reliable transmission data. The result of the analysis is a complete set of resonance parameters, the associated covariance

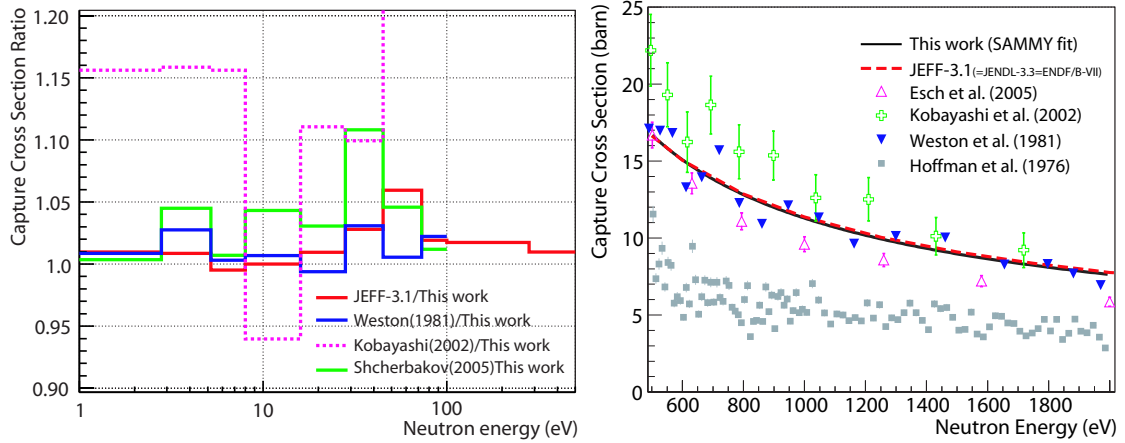


Figure 8.1: Comparison of the ^{237}Np capture cross section measured and analyzed in this work with respect to previous measurements and the evaluated data libraries.

matrix and a set of statistical parameters, suitable for the calculation of cross section in both the resolved and unresolved resonance regions. The compatibility between the present capture data with the transmission data as well as with the most recent evaluations indicates that, if the present data are included in future evaluations, the overall cross section uncertainty of 15% should be reduced safely down to 4%.

8.2.2 Measurement and analysis of ^{240}Pu

The neutron capture cross section of ^{240}Pu has been measured between 110 eV and 2 keV, which is within the resolved resonance region of ^{240}Pu , that ranges up to 2.7 keV or 5.7 keV depending on the evaluated data library.

The information on the sample of ^{240}Pu was not very accurate concerning its isotopic composition. Since the counting rate of the measurement revealed that there was a significant fraction of ^{239}Pu impurities that were not declared by the manufacturer, the isotopic composition was determined by means of γ spectroscopy at CERN with an accuracy of 5%. The isotopic composition in ^{240}Pu was fine tuned by means of a combined analysis of the capture data with the most reliable transmission data. The isotopic content of ^{240}Pu was fitted from the analysis in the high energy region, where the thin target approximation is valid and the capture yield is directly proportional to the surface atomic density: $Y_\gamma = n(\text{atoms/barn}) \cdot \sigma_\gamma(\text{barn})$.

The capture yield has been determined using the detection efficiency calculated by means of Monte Carlo simulations. The normalization factor corresponds to that obtained by the Saturated Resonance Method applied to the saturated resonances of ^{197}Au and ^{240}Pu at 4.9 eV and 1.06 eV, respectively. Both normalization values were compatible within uncertainties. This compatibility between the two independent normalization factors gives confidence in the

various data reduction techniques applied for the calculation of the detection efficiency, for the determination of the neutron scattering background, etc.

The capture cross section of ^{240}Pu has been analyzed between 110 eV and 2 keV from the experimental capture yield measured in this work and the transmission data included in the most recent evaluations. The region below 110 eV is strongly affected by self-shielding and multiple scattering effects which are difficult to calculate due to the grain structure of the powder sample, which results in a non-uniform mass distribution within the sample. The analysis of the energy region between 110 eV and 2 keV, described by 158 ^{240}Pu resonances, has shown that the capture and transmission data are compatible. The analysis of the most prominent resonances provided accurate resonance parameters. In the analysis of the weaker resonances, the radiative width was kept constant using the average value $\langle\Gamma_\gamma\rangle=29.7$ eV.

A large number of weak resonances has been proposed by Bouland et al. Among these 39 resonances, 18 could be confirmed, 14 had to be rejected, while 7 could neither be confirmed nor rejected, because they are either very small or they fall very close to a strong ^{239}Pu resonance. In addition, a total of 6 new resonances have been observed for first time.

The fact that the TAC is sensitive not only to capture cascades but also to the radiation emitted in fission reactions implies that resonances with a sizable fission to capture ratio can not be analyzed accurately. It has been shown that the analysis of such resonances is accurate only if the fission to capture ratio is lower than ~ 0.15 .

		JEFF-3.1	ENDF/B-VII	
	This work	JENDL-3.3 and Bouland et al.	Mughabghab Weston et al.	RIPL-2
D_0 (eV)	12.0 ± 0.6	12.06 ± 0.06	13.6 ± 0.7	12.4 ± 0.7
$\langle\Gamma_\gamma\rangle$ (meV)	29.7 ± 1.9	31.8 ± 1.5	30.6 ± 2	30.7 ± 2.5
S_0 (10^{-4})	1.08 ± 0.02	1.03 ± 0.07	0.93 ± 0.08	1.05 ± 0.10

Table 8.2: *Average resonance parameters of ^{240}Pu determined in this work compared to previous values.*

The consistency of the resonance parameters has been validated by a statistical analysis. The average level spacing found in this work is 12.6 ± 6 eV, which is in between the values of the recent evaluations: 12.06 eV (JEFF-3.1 and JENDL-3.3) and 13.6 eV (ENDF/B-VII). It was confirmed that the radiative width follows the expected constant behaviour except in a reduced energy interval where subthreshold fission is large. The value of $\langle\Gamma_\gamma\rangle=29.7$ meV is 7% lower than that of the JEFF-3.1 and JENDL-3.3 evaluations and just 3% lower than the corresponding ENDF/B-VII value. Indeed, the average radiative width found in this work is in better agreement with respect to the well known width of the first resonance at 1.05 eV (29.15 ± 0.6 meV [76, 21]) than the evaluations. Concerning the neutron strength function, the combined analysis of capture and transmission data provides a value that remains constant with neutron energy, even above 1.5 keV where previous works show significant variations. The

recommended average resonance parameters from this work are summarized in Table 8.2, where the value from previous measurements and from the evaluations are given for comparison.

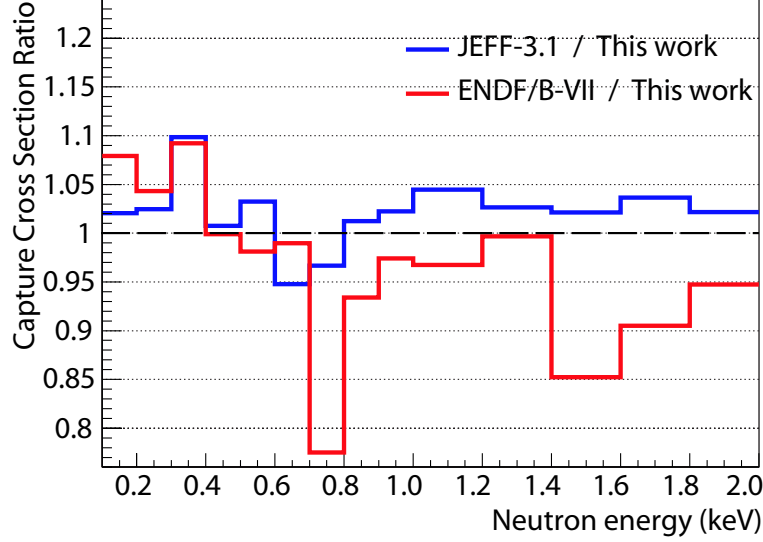


Figure 8.2: Comparison of the ^{240}Pu capture cross section measured at n_TOF and that of previous measurements and the evaluated data libraries

The capture cross section measured at n_TOF has been averaged in 14 energy intervals and compared to recent evaluations in Figure 8.2. Below 700 eV, the comparison indicates that the present results are in good agreement with all the evaluations: JEFF-3.1, JENDL-3.3 and ENDF/B-VII. At higher neutron energies, where significant differences are found between the evaluations, the n_TOF data are in better agreement with the JEFF-3.1 and JENDL-3.3 suggesting that there might be a problem in the analysis of the transmission data of Kolar et al. which are the basis of the ENDF/B-VII evaluation.

The present results give confidence in the capture cross section of the JEFF-3.1 and JENDL-3.3 evaluations, while they indicate that the ENDF/B-VII should be revised. The estimated uncertainty of 10% in the capture cross section of ^{240}Pu seems overestimated, indeed a more realistic uncertainty of only 5% can be safely assumed if the present data are included in future evaluations.

8.3 Improvements for future measurements

The high quality of the data obtained in the measurements on $^{237}\text{Np}(n,\gamma)$ and $^{240}\text{Pu}(n,\gamma)$ will play an important role in upcoming evaluations. So far, none of the evaluations of these isotopes include any neutron capture data in the resolved resonance region because the energy resolution or the reliability of the available data was not comparable to that of the transmission

measurements. This situation has changed with the availability of the present experimental data.

Future n_TOF experiments on actinides with the TAC setup will benefit from improvements based on the experience acquired in this work. The main sources of uncertainty in the measurements presented in this work will be eliminated or, at least, reduced significantly by means of the following upgrades:

- The replacement of the titanium canning of the actinide samples by an optimized *long-aluminium-canning* that will eliminate the background due to the Ti canning, therefore providing high quality data up to several tens of keV.
- The design of a new neutron absorber based on borated polyethylene will reduce the neutron sensitivity of the detection set-up further.
- By the use of the long canning for the sample, the vacuum windows inside the TAC will be eliminated, with the subsequent reduction of the background caused by neutrons scattered along the beam line.
- Active backings for the actinide samples will be used for tagging fission reactions, allowing the measurement of isotopes with a large subthreshold fission and even of fissile nuclei.
- The direct supervision of the manufacturing of the samples will avoid the problems associated to a poor characterization of the samples: mass, contaminants, mass distribution, etc.
- The use of metallic samples will avoid the non-uniform mass distributions associated to the grain structure of the powder samples.

Part V

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Part VI

APPENDIX

Appendix A

Resumen en español

A.1 Introducción y Motivación

Esta tesis doctoral trata sobre la medida y el análisis de la sección eficaz de captura neutrónica del ^{237}Np y el ^{240}Pu . Ambos isótopos son de origen humano y forman parte de los residuos radioactivos de las centrales nucleares.

En esta introducción se presenta la situación mundial en relación con el consumo y la demanda de energía haciendo especial énfasis en la energía nuclear. Se discuten algunas de las principales ventajas y desventajas de la energía nuclear y se presenta la tecnología de separación y transmutación como una solución a la gestión de los residuos nucleares de alta actividad. Por último, se demuestra la importancia que tiene medir con suficiente precisión las secciones eficaces neutrónicas de actínidos minoritarios y su papel en el diseño de sistemas de transmutación.

A.1.1 Producción de energía en el mundo

Desde la revolución industrial en el siglo XIX al surgimiento de las nuevas economías asiáticas a comienzos del siglo XXI, el crecimiento económico de todos los países está correlacionado con un aumento en la demanda de energía. Durante estos dos siglos se han explotado una gran variedad de fuentes de energía, desde combustibles fósiles como el carbón, petróleo y gas natural, a la energía nuclear, hidroeléctrica y otras fuentes renovables como la solar y la eólica.

La contribución de cada fuente de energía en el consumo total de electricidad no es homogénea y tiene una fuerte dependencia geográfica, incluso dentro de la Unión Europea (UE-25). La Figura A.1 muestra la contribución de las diferentes fuentes de energía en el mundo y en la UE-25 [4]. Se observa que el carbón (40,3%) y el gas (19,7%) suponen más de la mitad de la producción mundial de electricidad, mientras que en la UE-25 las principales contribuciones

corresponden a la energía nuclear (31%) y al carbón (29%). En cuanto a las tecnologías renovables, la mayor contribución proviene de la energía hidroeléctrica, mientras que el resto (solar, eólica, biocombustibles, etc.) contribuyen con menos del 5% al total.

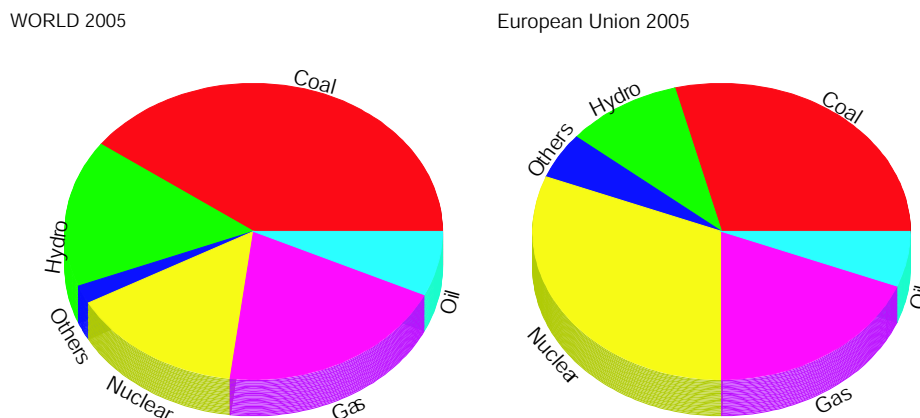


Figure A.1: Contribución de las diferentes fuentes de energía al consumo de electricidad en el mundo (izquierda) y en la UE-25 (derecha). Otros: solar, eólica, biocombustibles, etc.

La predicciones de la Agencia Internacional de Energía (IAEA - World Energy Outlook 2007 [4]) auguran para las próximas décadas un incremento anual en la demanda de electricidad del 2,6%. Este aumento estará dominado por las nuevas economías no pertenecientes a la OCDE de modo que su demanda de electricidad superará la de los países de la OCDE en el año 2015. Este hecho, junto con el aumento de los precios del petróleo, los riesgos geopolíticos asociados a la energía y la preocupación por el calentamiento global, han provocado en todo el mundo el inicio de un intenso debate sobre el futuro de la producción de energía mundial.

En vista de este escenario, la UE-25 propone en su informe *A European strategic energy technology plan: Towards a low carbon future* [5], una reducción del 20% en el consumo de energía primaria y de las emisiones de CO₂ con una contribución de 20% de las energías renovables para el año 2020. Estos objetivos siguen las líneas principales del informe *On Conventional energy sources and energy technology* [6] en el que se establece claramente que la energía nuclear es indispensable para suplir las necesidades básicas de energía en Europa a media plazo. Esta conclusión lleva a la necesidad de aumentar los esfuerzos en el diseño de una tecnología nuclear sostenible, tanto respecto a la producción de energía como a la gestión de los residuos radioactivos.

A.1.2 La Energía Nuclear

El desarrollo de la energía nuclear se produjo sólo unos años después del descubrimiento del neutrón por James Chadwick en 1932 [7]. Seis años después Otto Hahn y Fritz Strassmann [8]

observaron, en sus estudios sobre la interacción de neutrones con la materia, que los átomos de uranio se rompían en dos fragmentos bajo el bombardeo de neutrones emitiendo en el proceso entre dos y tres neutrones además de una cantidad importante de energía. El proceso, hasta entonces desconocido, fue explicado por Meitner y Frisch en su trabajo *Desintegration of Uranium by Neutrons: a New Type of Nuclear Reaction* [9] que fue publicado en la revista *Nature* en 1939 y en cual se le otorgó su nombre actual: *fisión nuclear*.

Históricamente, Leó Szilárd fue el primero en darse cuenta del amplio espectro de aplicaciones que ofrecía este descubrimiento y patentó la reacción en cadena en 1934 [10], aunque fue la importante contribución de Enrico Fermi la que superó el desafío de utilizar la reacción en cadena como proceso fundamental para la generación de energía. La Figura A.2 muestra un esquema de la reacción en cadena en la que los neutrones emitidos en una reacción de fisión son los encargados de producir nuevas reacciones de fisión. Fermi y Szilárd patentaron conjuntamente el reactor de neutrones [11] y, como parte del Proyecto Manhattan en la Universidad de Chicago, construyeron el primer reactor nuclear (Chicago Pile-1) en 1944, sólo doce años después del descubrimiento del neutrón.

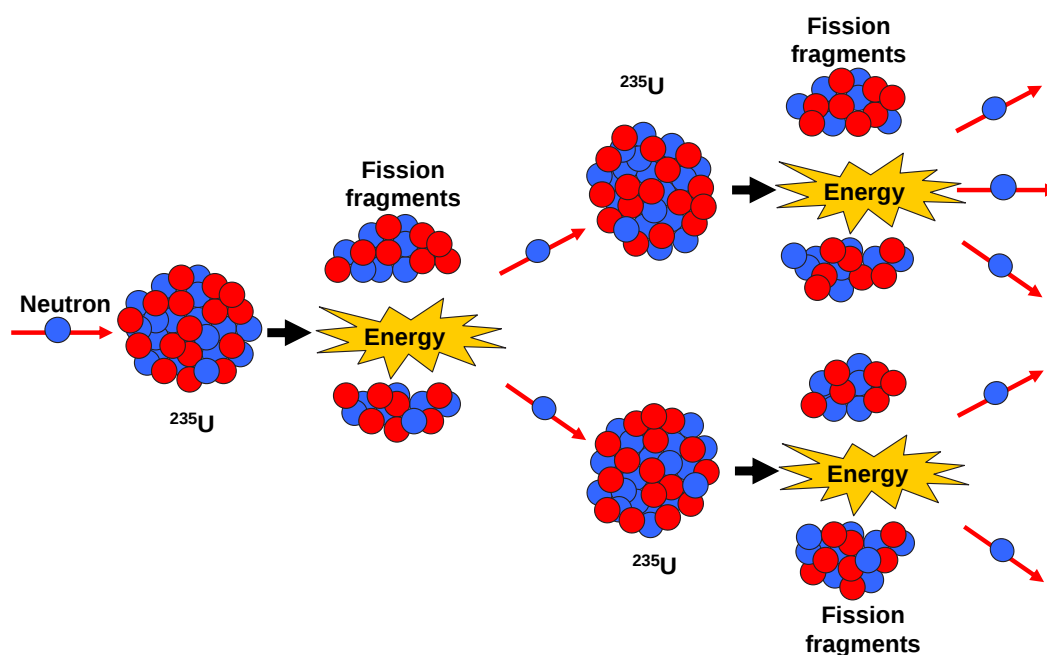


Figure A.2: Esquema de la reacción de fisión en cadena en la que las reacciones de fisión son inducidas por neutrones emitidos en las anteriores reacciones de fisión.

La Energía Nuclear en el mundo

En 1951, el reactor experimental EBR-I en Idaho (EE.UU.) se convirtió en la primera planta nuclear capaz de generar electricidad, tanta como para iluminar ocho bombillas de 100 vatios. Unos años más tarde, en 1958, se inauguró la primera planta de energía nuclear en Gran Bretaña y

desde entonces muchos países han desarrollado sus propios programas de energía nuclear. Alentados por la crisis del petróleo en los primeros años setenta, algunos países industrializados como Alemania, Canadá, España, Francia, Italia y Japón iniciaron sus programas nucleares, seguidos por México, Brasil, Taiwán y Corea. Pero en la décadas siguientes se redujo sustancialmente la construcción de nuevas centrales nucleares debido principalmente a preocupaciones por la seguridad de esta tecnología tras el accidente de Chernobyl en 1986.

A la fecha de Abril de 2008 operaban un total de 439 reactores nucleares en 31 países, con una potencia instalada de 372 GWe. En la UE-25 el número ascendía a 197 centrales nucleares con una potencia instalada de 170 GWe, y 13 nuevos reactores estaban en construcción en cinco países diferentes. La Tabla A.1 contienen el número de reactores nucleares en operación y en construcción en la UE-25, junto con la potencia asociada.

País	En operación		En construcción	
	#	MWe	#	MWe
Bélgica	7	5824	-	-
Bulgaria	2	1906	2	1906
República Checa	6	3523	-	-
Finlandia	4	2696	1	1600
Francia	59	63260	1	1600
Alemania	17	20470	-	-
Hungría	4	1829	-	-
Lituania	1	1185	-	-
Países Bajos	1	482	-	-
Rumania	1	1300	-	-
Federación de Rusia.	31	21743	7	4789
Rep eslovaco	5	2034	-	-
Y Eslovenia	1	666	-	-
España	8	7450	-	-
Suecia	10	8974	-	-
Suiza	5	3220	-	-
Ucrania	15	13107	2	1900
Reino Unido	19	10222	-	-
UE-25	197	169901	13	11795

Table A.1: *Reactores nucleares en la UE-25, en funcionamiento y en construcción, en Abril de 2008 [2].*

Respecto a la contribución de la energía nuclear al total de la producción de energía en cada país, Francia ocupa la primera posición con una cuota del 79%, seguida de Lituania con el 70%, Bélgica y la República de Eslovaquia con el 56%, y Suecia con el 47%. En España [1] hay un total de 6 plantas nucleares con 8 reactores que generan el 20% de la electricidad consumida en el país, mientras que más de la mitad se produce mediante el uso de fuentes emisoras de CO₂ como el carbón, el gas y el petróleo.

SPAIN 2007

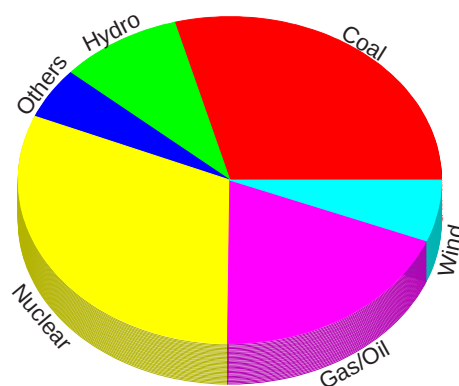


Figure A.3: *Distribución de la producción de electricidad en España por los diferentes fuentes de energía [1].*

Los reactores nucleares

El mecanismo de producción de energía en la mayoría de las centrales nucleares es similar al de las plantas térmicas convencionales (gas, carbón y petróleo), en el sentido de que el agua se calienta con el fin de producir vapor e impulsar una turbina que genera electricidad. La principal diferencia, que de hecho es muy importante, es que en un reactor nuclear el calor es producido por una reacción de fisión en cadena autosostenida.

La Figura A.4 muestra un esquema simplificado del funcionamiento de un reactor nuclear de agua a presión (PWR), donde los números entre paréntesis se corresponden con las diferentes partes del reactor. El núcleo del reactor (12) consiste en uranio enriquecido (3-5% de ^{235}U) en el que se producen las reacciones de fisión, liberando una gran cantidad de energía y algunos neutrones (en promedio 2,3 neutrones/fisión) que mantienen la reacción en cadena. La tasa de reacción se controla mediante el uso de barras de control (1) que pueden absorber una gran cantidad de neutrones. La energía liberada en las reacciones de fisión se utiliza para calentar el líquido que refrigera en núcleo (13) y el calor se transfiere a un circuito secundario (11) a través de un generador de vapor (3) el cual finalmente se pasa por una turbina (4) produciendo electricidad (5,6). El agua del circuito secundario se enfría por intercambio de calor con un foco frío (8,15), generalmente un río o un lago, produciendo vapor de agua (7) que se suele emitir a través de las torres de refrigeración.

La mayoría de los reactores nucleares en uso utilizan combustible de uranio enriquecido al 4% en ^{235}U y son refrigerados con agua ligera que también se utiliza como moderador de neutrones. Estos reactores se conocen como reactores de agua ligera (LWR) y pueden trabajar con agua a presión (PWR) o en ebullición (BWR). El siguiente tipo de reactores más abundante es el reactor nuclear canadiense de agua pesada a presión (PHWR), que trabaja con agua deuterada y con un combustible de uranio natural o ligeramente enriquecido. Otro tipos de reactores son los de grafito de agua ligera (LWGR) diseñado por la Unión Soviética, o el reactor avanzado

refrigerado por gas (AGCR) diseñado por el Reino Unido.

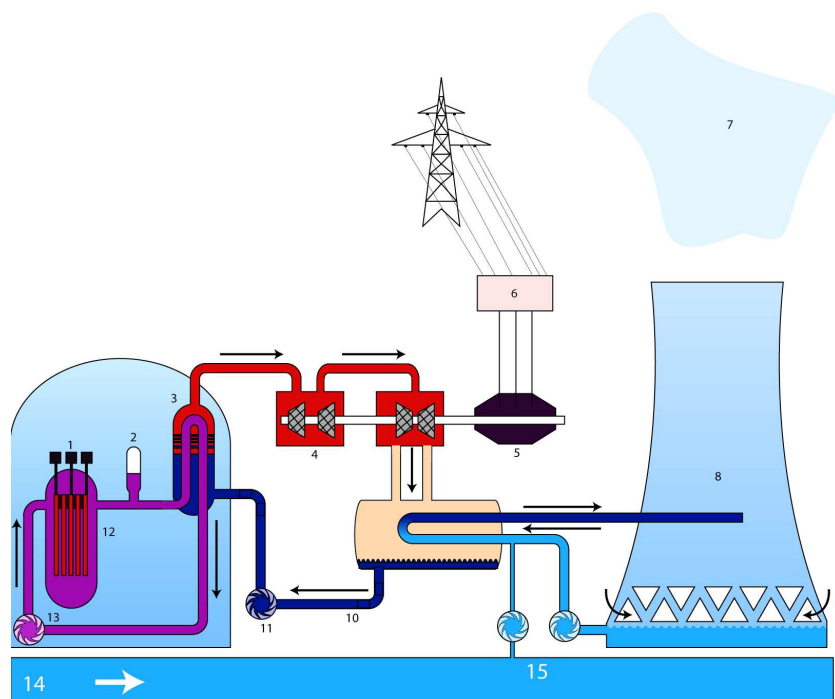


Figure A.4: Esquema de una planta de energía nuclear con un reactor de agua ligera a presión. (1) Barras de control. (2) Presurizador. (3) Generador de vapor. (4) Turbina. (5) Generador de energía eléctrica. (6) Transformador de alta tensión. (7) Vapor de agua. (8) Torre de refrigeración. (9) Entrada de aire para al refrigeración. (10) Entrada de agua. (11) Bomba del circuito secundario. (12) Núcleo del reactor. (13) Bomba del circuito primario. (14) Foco frío para la refrigeración. (15) Suministro de agua.

Generación y gestión de residuos radioactivos

A lo largo de la operación de una planta de energía nuclear los residuos nucleares radioactivos se generan mediante: (i) reacciones de fisión produciendo núcleos radioactivos, (ii) reacciones inducidas por neutrones en uranio -tales como (n, γ) o (n, α) - que dan lugar a elementos transuránicos, o (iii) activación del agua y los materiales estructurales. Los residuos producidos por fisión y activación son generalmente emisores γ , mientras que algunos fragmentos de fisión y los elementos transuránicos emiten radiación β y α y suelen tener vidas medias largas, del orden de centenas, miles o incluso millones de años.

Una de las soluciones para la gestión de los residuos de alta actividad consiste en su almacenamiento bajo tierra en un lugar especialmente diseñado, situado a varios cientos de metros bajo la superficie, que utiliza la roca circundante como una barrera para evitar las llegadas de isótopos radioactivos al medio ambiente. Sin embargo, entre las políticas energéticas de la UE y de muchos otros países existe la voluntad de lograr sistemas de producción de energía sostenibles, a través de la optimización de los recursos naturales y la minimización de las consecuencias a

largo plazo para las generaciones futuras.

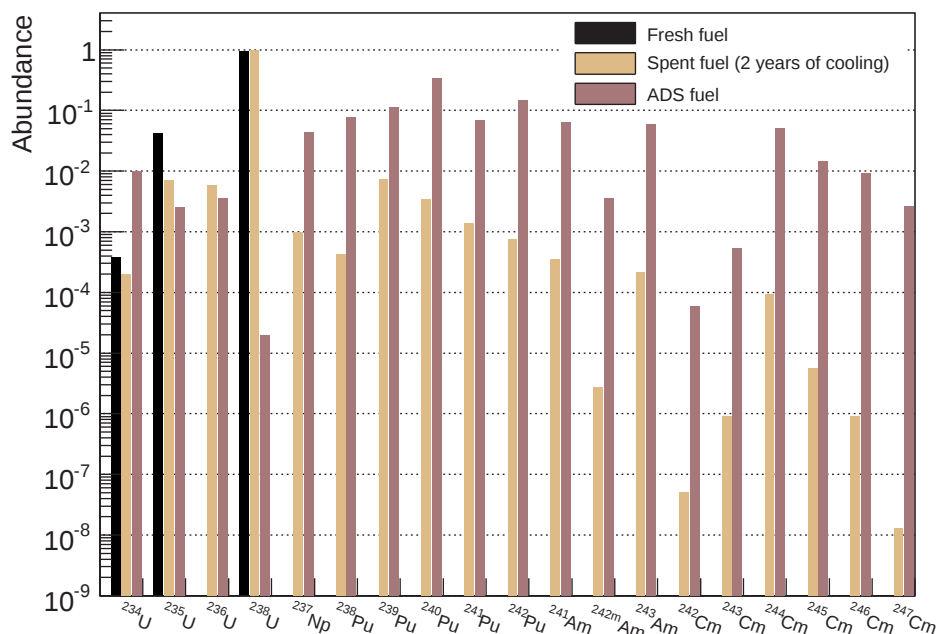


Figure A.5: *Composición isotópica del combustible nuclear en diferentes escenarios: combustible inicial de un PWR (negro), combustible gastado de un PWR después de 4 años de enfriamiento (marrón), combustible de un transmutador ADS (marrón oscuro).*

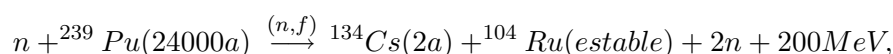
El principio de sostenibilidad aplicado a la energía nuclear implica, principalmente, la reducción al mínimo de la radiotoxicidad (volumen y dosis) de los residuos de alta actividad antes de su almacenamiento geológico. Este objetivo puede alcanzarse por medio de la técnica de *Separación y Transmutación* S&T que permite reducir significativamente el volumen y la radiotoxicidad de los residuos almacenados. ésta es la línea de investigación de uno de los proyectos de la Comunidad Europea denominado RED-IMPACT [13], que se encarga de evaluar las repercusiones que la tecnología de S&T tendrían respecto a la sostenibilidad de la energía nuclear.

La Figura A.5 ilustra la variedad de elementos transuránicos que se producen durante la operación de un reactor nuclear. En la figura se muestra la composición isotópica del combustible de un reactor PWR antes de ser irradiado (negro) y del combustible gastado tras cuatro años de enfriamiento (marrón claro). Se observa claramente que los actínidos minoritarios (Np, Pu, Am y Cm), que no formaban parte del combustible inicial, se producen después de la irradiación: por cada 100 kg de combustible irradiado aparece 1 kg de plutonio y 100 g de actínidos minoritarios. La tercera columna asociada a cada isótopos (marrón oscuro) corresponde a la composición del combustible nuclear que se espera utilizar en un sistema transmutador.

A.1.3 Transmutación de residuos radioactivos

La idea de la Separación y Transmutación S&T es la separación química de los diferentes elementos del combustible gastado y su quemado en un dispositivo nuclear que utilice un combustible con un alto contenido de actínidos minoritarios. De este modo podría reducirse sustancialmente la radiotoxicidad y el volumen (en un factor 100) de los residuos de alta actividad. En los últimos años se han desarrollado varios proyectos de investigación en diferentes países y organismos internacionales relacionados con el diseño de la futura generación de reactores de transmutación. La referencia [14] ofrece un resumen muy completo de los proyectos financiados por la Unión Europea

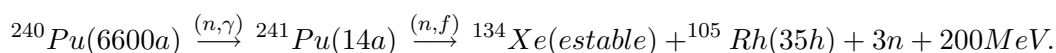
El proceso fundamental en la transmutación es, como en los reactores convencionales, la fisión inducida por neutrones que puede transformar un isótopo radiactivo en un par de núcleos con vidas medias más cortas o incluso núcleos estables. La transmutación tendría lugar en un sistema nuclear con un espectro de neutrones rápidos, ya que la probabilidad de fisión en actínidos crece a altas energías. Un ejemplo de transmutación por reacciones de fisión es



donde la vida media media de cada isótopo se indica entre paréntesis. La transmutación también puede llevarse a cabo por otros mecanismos como la captura neutrónica



o una combinación de varias reacciones



La Figura A.6 muestra la contribución de los isótopos transuránicos a la radiotoxicidad total de los residuos nucleares en mSv por cada tonelada inicial de uranio. Esta información da idea de la importancia de la transmutación de los diferentes isótopos para la reducción global de radiotoxicidad. En la figura se observa que en los primeros años los isótopos ${}^{239-241}\text{Pu}$ y ${}^{244}\text{Cm}$ son los principales contribuyentes a la radiotoxicidad, mientras que en el intervalo entre el 50 y el 2000 años tras la descarga la radiotoxicidad es dominada por el ${}^{241}\text{Am}$. Finalmente, el ${}^{240}\text{Pu}$, el ${}^{239}\text{Pu}$ y el ${}^{243}\text{Am}$ dominan a tiempos de 10^5 años, cuando la radiotoxicidad del combustible gastado alcanza el valor correspondiente a la del combustible inicial.

A primera vista, el neptunio parece poco importante en un esquema de transmutación, sin embargo el neptunio es motivo de preocupación en la gestión de los residuos a largo plazo debido a su larga vida media ($2,1 \cdot 10^6$ años), a que se va generando a lo largo del tiempo debido a la emisión α del ${}^{241}\text{Am}$, y a su alto grado de movilidad en aguas subterráneas. Por esto motivos, la transmutación del neptunio se considera de gran importancia [15] en la evaluación general del riesgo y las consecuencias a largo plazo del almacenamiento de residuos de vida larga.

A día de hoy se barajan dos opciones para la transmutación de residuos:

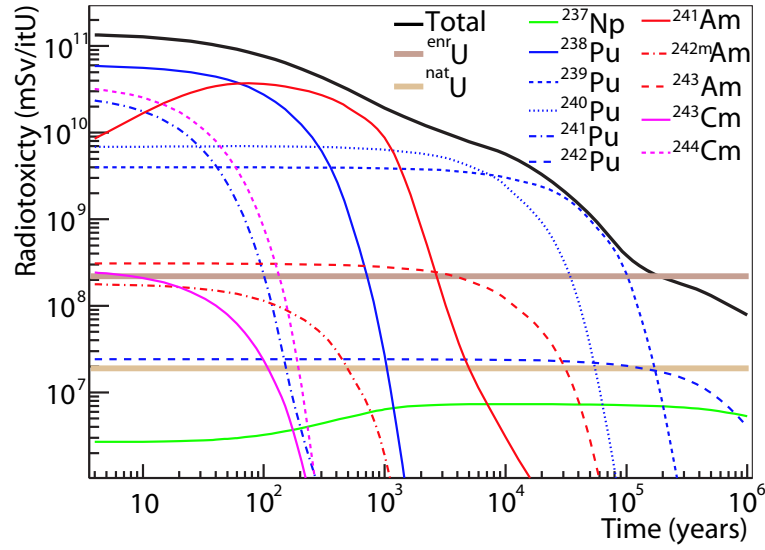


Figure A.6: Contribución de cada uno de los actínidos minoritarios a la radiotoxicidad de los residuos nucleares.

- La primera opción consiste en el uso de reactores rápidos críticos de tipo Generación-IV. El objetivo principal de este tipo de reactores es mejorar la seguridad nuclear y la resistencia a la proliferación, reduciendo al mínimo la generación de residuos y optimizando la utilización de los recursos naturales porque utilizarían el ^{238}U como combustible. Además se espera que sea posible añadir al combustible una cierta fracción de actínidos minoritarios producidos en reactores convencionales, con el fin de reducir poco a poco el inventario de residuos de larga vida.
- La segunda opción consiste en el uso de sistemas nucleares subcríticos con un espectro de neutrones rápido. En un reactor subcrítico, la tasa de reacción se sostiene mediante una fuente de neutrones externa, consistente en un acelerador de protones acoplado a un blanco de espalación. Este sistema se conoce como un transmutador asistido por acelerador (Accelerator Driver System - ADS) y ofrece varias ventajas a la hora de su control: siempre que la fuente de neutrones se apaga, la tasa de reacción se detiene. La operación subcrítica permite aumentar la flexibilidad en la composición del combustible de modo que se puede utilizar un combustible con una alta concentración de actínidos minoritarios en comparación con el combustible de los reactores de Generación-IV.

El diseño conceptual del ADS [16], que se muestra en la Figura A.7, ha sido ampliamente estudiado en los últimos años pero todavía no se ha construido ningún prototipo. En principio, la tecnología actual permitiría la construcción de un ADS a escala experimental [17], pero aún falta profundizar en una serie de aspectos fundamentales del diseño para la construcción de un ADS a escala industrial. Dichos aspectos fundamentales son los siguientes [18, 19]:

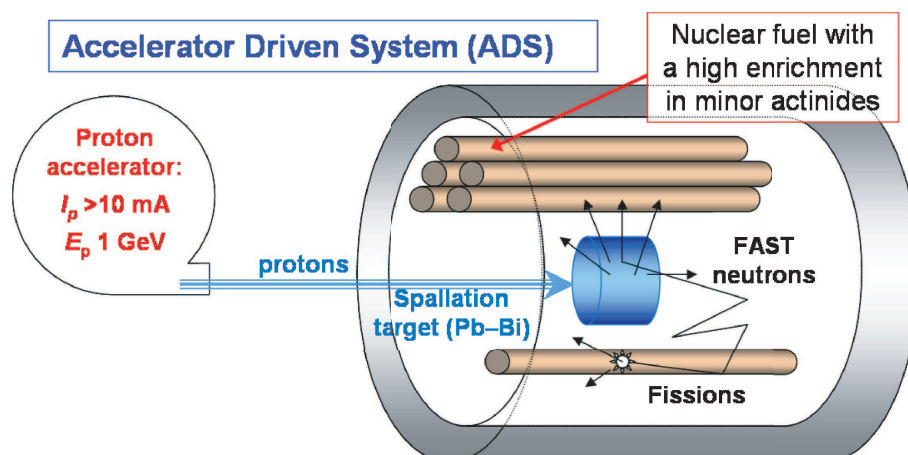


Figure A.7: Esquema de los diferentes componentes de un transmutador asistido por acelerador (Accelerator Driver System - ADS).

- Aumentar la precisión de las bases de datos nucleares que se utilizan en los cálculos de cinética y dinámica de reactores a fin de reducir las incertidumbres en el diseño.
- Investigar la respuesta de los materiales estructurales y del propio combustible a una alta irradiación con neutrones rápidos y protones.
- Mejorar los rendimientos de la tecnología de separación de los diferentes isótopos del combustible gastado.
- Evaluar de forma precisa el coste/beneficio de la tecnología de separación, transmutación y almacenamiento en profundidad de los residuos de alta actividad.

A.1.4 Necesidad de datos nucleares

El diseño de cualquier dispositivo nuclear, incluido los futuros reactores ADS y de Generación-IV, se basa en la validez de los resultados de los códigos de transporte de neutrones, transferencia de calor, termohidráulica, daño de materiales, etc.

El principal ingrediente de los códigos de transporte de neutrones, que determinan entre otros el grado de quemado de los diferentes isótopos en el combustible, son las secciones eficaces nucleares, probabilidades de emisión de secundarios (multiplicidades, energía y distribución angular), vidas medias y otros datos nucleares. Por lo tanto, la incertidumbre de los cálculos de transporte neutrónico está directamente relacionada con la precisión de estos datos nucleares básicos.

La obtención de secciones eficaces neutrónicas pasa por un complejo proceso de evaluación que retrasa el acceso de la comunidad de usuarios a los datos nucleares, tal como se ilustra en la

Figura A.8. Para cada isótopo, los evaluadores contrastan los datos experimentales disponibles con modelos teóricos y producen un conjunto coherente de secciones eficaces para cada canal de reacción. El proceso de evaluación prosigue con la validación de las secciones eficaces mediante la comparación de cálculos de reactores de referencia con los correspondientes parámetros experimentales. El proceso de evaluación sufre varias iteraciones, hasta que finalmente las secciones eficaces se distribuyen en forma de *Evaluated Nuclear Data Files* (ENDF) [20]. Las bases de datos más utilizadas son JEFF (UE), ENDF (EE.UU.), JENDL (Japón), BROND (Rusia) y CENDL (China), que se pueden consultar en [21].

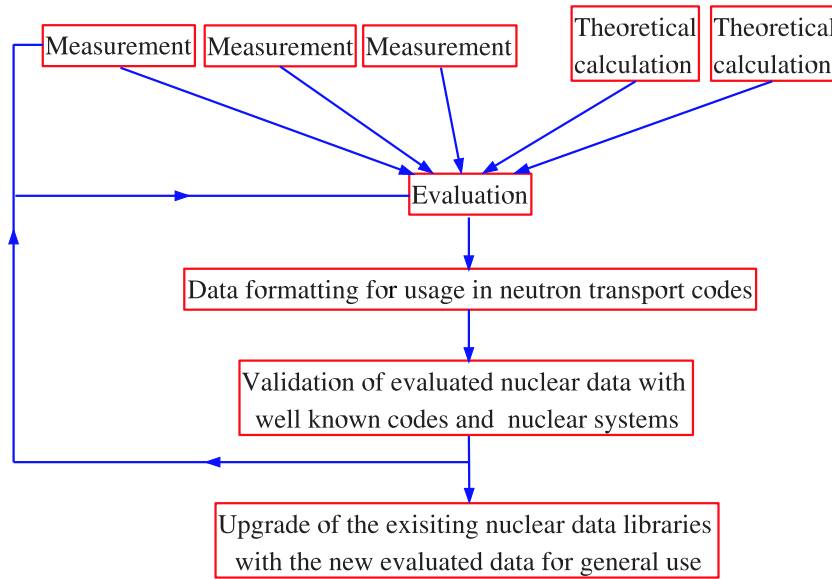


Figure A.8: Esquema conceptual del proceso de evaluación de datos nucleares que va desde las medidas hasta la distribución de las secciones eficaces evaluadas.

Actualmente, la precisión de las secciones eficaces evaluadas es muy diferente entre unos y otros isótopos. Estas diferencias están relacionadas con el hecho de que las centrales nucleares convencionales funcionan con un flujo de neutrones de baja energía (térmicos) y con un combustible de uranio. Por ello la precisión de la sección eficaz de fisión de ^{235}U es mejor del 1%, mientras que las de secciones eficaces de actínidos minoritarios poseen una incertidumbre que varía entre el 5% y el 50%, dependiendo de: el isótopo en cuestión, el rango de energía y el tipo de reacción [22]. Dichas incertidumbres se deben principalmente a que el conjunto de datos experimentales disponibles presenta incompatibilidades entre unas medidas y otras, no cubre todo el rango de energías necesario o poseen una resolución insuficiente. Además, las incertidumbres sistemáticas que declaran los autores de las medidas están generalmente subestimadas, lo cual lleva a los evaluadores a tener que decidir qué datos son más confiables sin demasiada información al respecto.

Ya que las medidas de secciones eficaces requieren un dispositivo experimental complejo y que su análisis y evaluación requiere un proceso largo, es necesario identificar las medidas que son prioritarias para el diseño de un ADS. Este es el objetivo de los *análisis de sensibilidad y propagación de incertidumbres*, en los que se determina el impacto de la incertidumbre en cada

las diferentes secciones eficaces en el cálculo de las propiedades macroscópicas de los reactores: κ_{eff} , β_{eff} , $\Delta\rho^{Doppler}$, $\Delta\rho^{Void}$, etc. Atendiendo a este tipo de estudios, la Agencia de Energía Nuclear (NEA) [23] publica cada cierto tiempo una lista de prioridades (HPRL) [23] con el fin de proporcionar una guía para la planificación de nuevas medidas y evaluaciones.

Varios análisis de sensibilidad recientes [24, 22] han evidenciado la necesidad de determinar las secciones eficaces de captura neutrónica de un conjunto de actínidos minoritarios (^{237}Np , $^{238,239,240,241,242}\text{Pu}$, $^{241,243}\text{Am}$ y ^{245}Cm) con una precisión mejor o igual al 5% en el rango de energías hasta 1 MeV. Teniendo en cuenta los datos disponibles a día de hoy y las dificultades asociadas a las diferentes medidas, la lista de medidas prioritarias se reduce al ^{237}Np , $^{240,242}\text{Pu}$, $^{241,243}\text{Am}$ y ^{245}Cm .

Dichas medidas han sido propuestas [28] para su realización en el experimento n_TOF del CERN [27], una instalación especialmente diseñada a tal efecto:

1. Proporciona un haz de neutrones de muy alta intensidad, en un amplio rango de energías y con una alta resolución.
2. Dispone de avanzados dispositivos de detección que permiten medir con buena precisión las secciones eficaces de captura y fisión utilizando muestras de poca masa y/o con una tasa de actividad alta.

El proyecto que contempla estas medidas forma parte del 5º Programa Marco de la UE (FIKW-CT-2000-00107) y del acuerdo CIEMAT-ENRESA para la *Transmutación de Residuos de Alta Actividad*, y ha sido cofinanciado por el Ministerio de Educación y Ciencia (actualmente Ministerio de Ciencia e Innovación) a través del *Plan Nacional de Física de Partículas*.

En este trabajo se presentan las primeras medidas de actínidos que se han realizado dentro del proyecto n_TOF y que se corresponden con las secciones eficaces de captura neutrónica del ^{237}Np y el ^{240}Pu en el rango entre 1 eV y 2 keV. Las medidas se realizaron en 2004 y ambas han proporcionado un conjunto de datos con una precisión mejor del 4%. Para llegar a tal precisión se ha caracterizado de forma detallada el funcionamiento del dispositivo experimental y se han desarrollado varias técnicas que permiten calcular de forma precisa las diferentes fuentes de fondo, la eficiencia de detección, los efectos de pérdidas por tiempo-muerto, etc. Los datos experimentales se han combinado con los medidas de transmisión más recientes y se han analizado utilizando el formalismo de la matriz-R. Los resultados son unas secciones eficaces precisas y con un buen control de errores sistemáticos, que han permitido reducir la incertidumbres que existían en las bases de datos disponibles.

A.2 Resumen y Conclusiones

Esta tesis doctoral esta dedicada a las primeras medidas por tiempo de vuelo de la sección eficaz de captura neutrónica de actínidos minoritarios llevadas a cabo en la instalación n_TOF del

CERN en 2004, correspondientes al ^{237}Np y el ^{240}Pu . El objetivo de las medidas [28] es proveer a la comunidad de datos nucleares de un conjunto de datos experimentales de alta precisión con un buen control de las incertidumbres sistemáticas y, por tanto, con una estimación correcta de su incertidumbre global. Este punto es crucial para la labor de los evaluadores que necesitan contrastar los datos experimentales disponibles y por ello se ha hecho un esfuerzo en determinar la naturaleza y magnitud de la incertidumbres sistemáticas.

El trabajo ha cubierto todos los aspectos relevantes: se ha participado en todo el proceso experimental, desde la puesta a punto de los detectores hasta la medida de datos, el desarrollo de software para el análisis de los datos y el diseño de técnicas específicas para el tratamiento de los diferentes fuentes de incertidumbre. Una parte importante del trabajo, incluyendo las propias medidas, se llevó a cabo durante una estancia de seis meses en el CERN. Además, parte de la metodología utilizada para el análisis de las secciones eficaces de desarrolló durante una visita a ORNL en USA, donde se encuentra una de los grupos de investigación líder a nivel mundial en la evaluación de datos nucleares. Finalmente, los resultados de este trabajo han sido presentados en foros internacionales y formarán parte de la nueva librería evaluada JEFF.

A.2.1 Dispositivo experimental y técnicas de reducción de los datos

Las medidas se han realizado en la instalación n_TOF del CERN utilizando el Calorímetro de Absorción Total (Total Absorption Calorimeter - TAC). La combinación de ambos ofrece unas características excelentes para la medida de muestras radioactivos. La instalación n_TOF posee una línea de tiempo de vuelo muy larga (185 m) y produce el flujo instantáneo de neutrones más intenso a nivel mundial de entre este tipo de instalaciones. Desde el año 2001, la instalación n_TOF ha demostrado sus excelentes características para la medida de secciones eficaces de fisión y captura neutrónica.

El Calorímetro de Absorción Total es un detector 4π formado por 40 cristales de BaF_2 . La puesta a punto del TAC se realizó principios de 2004 como parte de este trabajo se llevó a cabo una caracterización completa del detector y de sus respuesta a medidas de fuentes de calibración y también medidas de captura neutrónica. Además, se desarrollaron una serie de herramientas informáticas para realizar las calibración semanales del sistema de detección, la monitorización de la ganancia de los 40 cristales, las sincronización temporal de todos los canales de adquisición digital, y el software de histogramación de los datos almacenados en el sistema CASTOR del CERN. Durante los trabajos de caracterización del TAC se identificaron los límites que afectan las medidas: el rango de energía que se puede analizar de forma precisa, la contribución de los diferentes fondos y las tasas de conteo aceptables. Se validó experimentalmente que la eficiencia de detección del TAC a procesos de dispersión neutrónica se puede reducir por debajo del 1% mediante el uso combinado de un moderador/absorbente de neutrones esférico colocado alrededor de la muestra y el uso de cápsulas de fibra de carbono enriquecida con ^{10}B para cada cristal.

Se estudiaron la diferentes fuentes de fondo y su efecto en el análisis de los datos de cada medida realizada y se encontraron las condiciones de energía depositada y multiplicidad que

minimizan su contribución. Se determinó el fondo punto a punto en función de la energía del neutrón para su posterior inclusión en el análisis de la sección eficaz. Para este propósito, N. Larsson desarrolló un nuevo paquete para el código de análisis de resonancias SAMMY.

Una de las principales innovaciones de este trabajo respecto de otros anteriores y que contribuye de forma muy significativa a la precisión de los resultados finales es la de utilizar herramientas de simulación Monte Carlo para el cálculo de la eficiencia de detección del TAC y de las pérdidas de señales relacionadas con el tiempo muerto en cada cristal.

Se ha desarrollado un código de simulación basado en GEANT4, que contiene la geometría detallada del detector y del área experimental así como dos generadores de eventos primarios para desintegración β y para reacciones de captura neutrónica. Ambos generadores de eventos se han validado mediante la comparación con datos experimentales, demostrando la excelente capacidad de la simulación para reproducir los datos experimentales en medidas con y sin absorbente neutrónico. La simulación Monte Carlo realizada permite calcular la eficiencia de detección del TAC con una precisión inferior al 3%. Esta precisión es excelente y ha sido determinada mediante el estudio de la desviación sistemática en el valor de la eficiencia de detección para un gran número de simulaciones en las que se varían los parámetros del generador de cascadas y de la geometría del detector dentro de unos intervalos razonables. Este trabajo fue presentado para la obtención del Diploma de Estudios Avanzados por la UCM en Junio de 2005. Por otro lado, el generador de eventos de captura acoplado al código de simulación ha demostrado ser una herramienta muy potente para el estudio sistemático de *Funciones de intensidad fotónica* asociadas a las probabilidades de transición nucleares, como ha sido discutido en diferentes conferencias internacionales [114, 119].

Otra de las principales fuentes de errores sistemáticos en este tipo de medidas está relacionado con la pérdida de señales en el detector debido al tiempo muerto de cada cristal. Tras un extenso estudio bibliográfico, se verificaron diferentes modelos de corrección (analíticos y Monte Carlo) y se comprobó que ninguno era aplicable a los datos del TAC debido a las particularidades intrínsecas del sistema de adquisición de datos digital. La utilización de correcciones estándar para sistemas paralizables y no paralizables a tasas de conteo superiores a $1\ \mu\text{s}^{-1}$ puede dar lugar a desviaciones sistemáticas superiores al 30%. Por ello, se ha desarrollado un modelo original, basado en técnicas Monte Carlo en el cual el tiempo muerto característico de cada cristal se incluye en la reconstrucción de eventos que deben ser generados con una distribución de tiempo de vuelo similar a la obtenida durante las propias medidas. De este modo puede obtenerse una eficiencia de detección en función del tiempo de vuelo para cada medida particular. El modelo se validó mediante medidas de captura neutrónica con resonancias ofreciendo un amplio rango de tasas de conteo.

El conjunto de técnicas desarrolladas para el cálculo de la tasa de reacción a partir de las medidas experimentales ha sido validado a través de la comparación de la tasa de reacción experimental de la medida del ^{197}Au con los valores esperados a partir de las secciones eficaces evaluadas. Los resultados de dicha comparación confirman las correcciones aplicadas al cálculo de la tasa de reacción a partir de los datos experimentales ya que son compatibles con las librerías evaluadas.

A.2.2 Medida y análisis de la sección eficaz de captura neutrónica de actínidos minoritarios

La sección eficaz de captura neutrónica de los actínidos minoritarios se ha medido utilizando muestras en forma de polvo de óxido XO_2 con una masa de 43.3 mg y 51.2 mg de ^{237}Np y ^{240}Pu , respectivamente. Las muestras se fabricaron en el IPPE de Obnisk y su montaje consiste en una cápsula de titanio dentro del cual se encuentra el material radioactivo depositado entre dos láminas de aluminio. Este montaje cumple la normativa ISO-2919 requerida por las autoridades de protección radiológica del CERN.

Las medidas de ^{237}Np y ^{240}Pu se realizaron entre Junio y Septiembre de 2004 y los primeros resultados fueron presentados el mismo año en la conferencia ND2004 celebrada en Notre Dame (USA) [161]. Estas medidas son, a día de hoy, las que presentan una mayor resolución y estadística de entre todas las medidas disponibles en la base de datos EXFOR [162].

Medida y análisis del ^{237}Np

Se realizó un estudio bibliográfico completo de las medidas de la sección eficaz de captura neutrónica y de las evaluaciones más recientes. Dicho estudio reveló que existen diferencias significativas entre las medidas disponibles, incluso entre las más recientes. Estas diferencias son mayores en la región de energía de las resonancias no resueltas, por encima de 500 eV. Además, se observó que todas las secciones eficaces evaluadas en la zona de resonancias resueltas están basadas en el análisis de datos de transmisión y fisión pero no de captura. Se puede concluir que estos resultados subrayan la importancia de realizar una medida precisa de la sección eficaz de captura del ^{237}Np en las regiones de resonancias resueltas y no resueltas para resolver las discrepancias encontradas hasta la fecha.

La sección eficaz de captura neutrónica del ^{237}Np se ha medido entre el 1 eV y 2 keV, intervalo que incluye la zona de resonancias resueltas y parte de la zona de resonancias no resueltas. Para reducir la magnitud de las correcciones debidas al tiempo muerto de los cristales, la tasa de conteo se mantuvo durante la medida siempre por debajo de $< 0,5$ cuentas/ μs . Los datos destacan por su excelente resolución en energía y la alta estadística, lo que ha permitido observar estructuras resonantes a energías por encima de 500 eV.

La tasa de captura se ha calculado utilizando dos métodos diferentes. El primer método consiste en el cálculo de la tasa de reacción sólo a partir de las medidas realizadas en n-TOF, es decir determinando la eficiencia de detección mediante simulaciones Monte Carlo y normalizando la tasa de captura observada a la medida de referencia de ^{197}Au en la resonancia saturada a 4.9 eV. El segundo método consiste en normalizar la tasa de reacción observada a los datos de transmisión correspondiente a las medidas realizadas en GELINA en 1999 [132]. Este segundo método reduce las incertidumbres asociadas a los factores que influyen en la normalización (eficiencia de detección, intensidad del haz de neutrones y masa de la muestra) y permite obtener la tasa de captura con un 4% de incertidumbre.

Los resultados obtenidos con el primer método son compatibles con los obtenidos con el

segundo, lo cual demuestra que la técnica de reducción de datos desarrollada en este trabajo produce resultados correctos, incluso en el caso de que no haya medidas de referencia disponibles. Sin embargo, el segundo método ha sido utilizado para el análisis de la sección eficaz ya que el uso de información adicional proporciona una tasa de captura más precisa y, lo que es más importante, permite reducir las correlaciones entre los parámetros de las resonancias Γ_γ y Γ_n mediante el análisis combinado de los datos de captura y transmisión.

El análisis de la sección eficaz a partir de la tasa de reacción se ha realizado con el código SAMMY mediante un análisis combinado de los datos de captura y transmisión. Dicho análisis es un proceso iterativo en el que se van refinando los parámetros que describen la sección eficaz hasta que se consigue un conjunto de los mismos que reproducen simultáneamente los datos de captura y transmisión. Para ello se ha desarrollado el software necesario para realizar un gran número de análisis simultáneos y comparar los resultados de los mismos.

Se han observado un total de 557 resonancias durante el análisis de la zona de resonancias resueltas, por debajo de 500 eV. El análisis por debajo de 43 eV ha proporcionado el valor del radio de dispersión R' y la anchura radiativa promedio $\langle\Gamma_\gamma\rangle$, que se han mantenido constantes a mayores energías del neutrón. Se ha observado que existe una correlación importante entre los parámetros de resonancias vecinas, aunque dicha correlación se restringe en la mayoría de los casos a los vecinos más próximos. Por otro lado, se ha observado que la normalización y el parámetro que describe el nivel de fondo muestran una correlación negativa (-0.25 en promedio) con los parámetros de la mayoría de las resonancias. El análisis combinado de los datos de captura y transmisión ha demostrado que ambos datos son compatibles en todo el intervalo de energía.

Por encima de 500 eV el solapamiento entre resonancias vecinas es demasiado grande como para realizar un análisis de resonancias individuales. Por tanto se ha analizado intervalo de energía hasta 2 keV mediante el formalismo de Hauser-Feshbach mediante el código FITACS implementado en SAMMY. Para ello se han calculado las secciones eficaces promediadas a partir de la tasa de reacción y los datos de transmisión experimentales asumiendo en ambos casos la aproximación de muestra delgada. El resultado del análisis muestra de nuevo que los datos de captura y transmisión son compatibles y más aún, que la normalización y el nivel de fondo obtenido a bajas energías son compatibles con los obtenidos a altas energías. El conjunto de parámetros $\langle\gamma\rangle$, R' y S_0 que permite reproducir los datos experimentales en este rango de energías está de acuerdo con los resultados obtenidos entre 1 eV y 500 eV, de modo que al análisis de los resultados en el rango de energías completo proporciona un conjunto de valores para la descripción estadística de la sección eficaz promedio. Estos valores se muestran en la Tabla A.2, en la que se han incluido además los valores encontrados en las diferentes evaluaciones.

La sección eficaz de captura neutrónica obtenida en n_TOF se ha comparado con los valores de medidas anteriores y las evaluaciones más recientes. En general, la sección eficaz de captura neutrónica obtenida en este trabajo es compatible con las evaluaciones JEEF-3.1, JENDL-3.3 y ENDF/B-VII. También hay un acuerdo dentro de las incertidumbres estimadas con los datos de Scherbakov et al. [136] por debajo de los 100 eV y Weston et al. [124] en el rango completo de energías analizado. La comparación con otras medidas muestra diferencias significativas más allá de las incertidumbres estimadas. Este es el caso de los datos de Kobayashi et al. [133],

	n_TOF	JEFF-3.1 JENDL-3.3 ENDF / B-VII	RIPL-2
$D_0(\text{eV})$	0.56 ± 0.02	0.58	0.57 ± 0.03
$\langle \Gamma_\gamma \rangle (\text{meV})$	40.8 ± 1.8	40.0	40.8 ± 1.2
$S_0(10^{-4})$	1.03 ± 0.08	0.97	1.0 ± 0.07
$R'(\text{fm})$	9.8 ± 0.6	10.5	

Las evaluaciones no dan las incertidumbres asociadas a estos parámetros.

Table A.2: *Parámetros estadísticos de la sección eficaz del ^{237}Np de este trabajo comparados con los valores de las evaluaciones.*

que muestran grandes diferencias con los datos de n_TOF en el todo el rango entre 1 eV y 2 keV, y los datos de Esch et al. [134], que está de acuerdo con los datos de n_TOF a 500 eV pero muestran diferencias significativas a mayores energías. La comparación entre las diferentes secciones eficaces de captura se ilustra en la Figura A.9. La figura de la izquierda muestra la comparación de la secciones eficaces en la zona de resonancias resueltas comparadas con las de n_TOF, todas calculadas con NJOY-99, y la figura de la derecha muestra las diferentes secciones eficaces de captura en la zona de resonancias no resueltas.

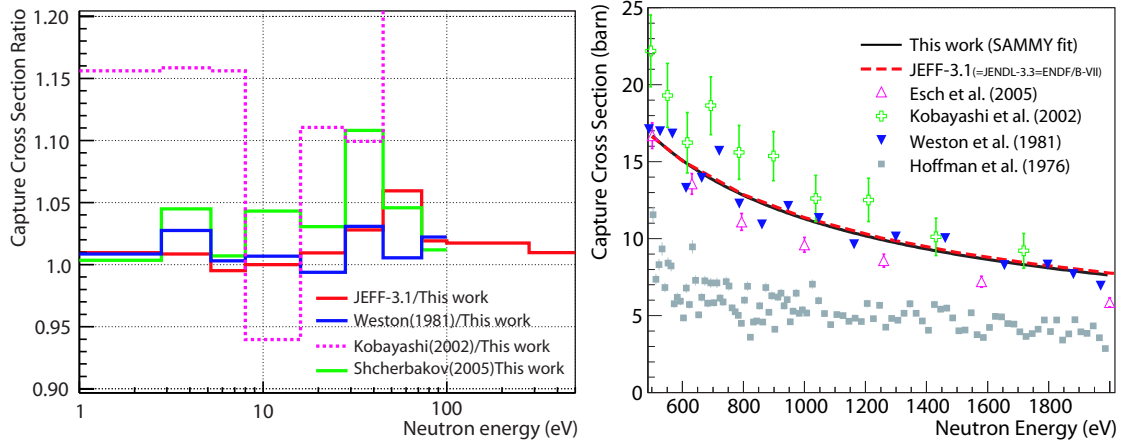


Figure A.9: *Comparación de la sección eficaz de captura neutrónica del ^{237}Np medida en n_TOF con los valores de medidas previas y de las secciones eficaces evaluadas.*

Resumiendo, este trabajo ha proporcionado un conjunto de datos experimentales de la sección eficaz de captura neutrónica del ^{237}Np en el rango de energías entre el 1 eV y 2 keV con una precisión del 4%. Todas las fuentes de incertidumbre han sido cuidadosamente investigadas y su magnitud cuantificada. Se ha demostrado que los datos experimentales son compatibles con los datos de transmisión más recientes y ambos se han analizado de manera conjunta para proporcionar un conjunto completo de parámetros de las resonancia y las matrices de covarianza. La compatibilidad con los datos de transmisión y las secciones eficaces evaluadas indican que la

incertidumbre actual del 15% puede reducirse al $\sim 4\%$ en el intervalo de energías analizado en este trabajo. En cuanto a los parámetros de las resonancias obtenidos en este trabajo, serán la base de la nueva evaluación JEFF.

Medida y análisis del ^{240}Pu

La revisión bibliográfica de las medidas de la sección eficaz de captura neutrónica del ^{240}Pu indican que no hay ninguna medida que haya producido datos experimentales fiables para el análisis de resonancias. De hecho, la medida referencia es todavía la de Weston et al. [150] de 1977 en la que sólo se determinó la sección eficaz de captura promedio en diferentes intervalos de energía. Debido a la falta de datos precisos, las evaluaciones más recientes están basadas exclusivamente en el análisis de medidas de transmisión y fisión. La comparación entre las diferentes evaluaciones muestra que existen diferencias superiores al 10% entre unas y otras en el rango de energías por encima de 700 eV. Las medidas llevadas a cabo en este trabajo deben dilucidar las diferencias encontradas a altas energías y producir los primeros datos experimentales de captura con la resolución y precisión suficiente para ser incluidas en la próximas evaluaciones.

La sección eficaz de captura neutrónica de ^{240}Pu se ha medido en el rango de energías entre 110 eV y 2 keV, que están dentro de la zona de resonancias resueltas que llega hasta 2,7 keV o 5,7 keV dependiendo de la evaluación. En esta medida se observan claramente resonancias resueltas hasta el límite de 2 keV, a partir del cual es imposible realizar análisis fiable alguno debido al fondo causado por la dispersión de neutrones en el titanio de la muestra.

Los análisis preliminares de la medida mostraron algunas características inesperadas:

En primer lugar, se observó un gran número de pequeñas resonancias que no correspondían al ^{240}Pu . Un análisis exhaustivo de estas resonancias reveló que éstas pertenecen al ^{239}Pu , que no estaba declarado en la composición isotópica de la muestra. El análisis por espectroscopía γ de la muestra indicó que había al menos un 12% de ^{239}Pu en la muestra. La cantidad de ^{240}Pu ha sido determinada mediante en análisis combinado de los datos de captura de este trabajo junto con los datos de transmisión de Kolar et al. [154] medidos en GELINA en 1968 y que son hasta hoy la base todas la evaluaciones.

En segundo lugar se observó que no era posible reproducir la forma de las resonancias a baja energía mediante el uso de parámetros de las resonancias razonables. La explicación más plausible es que la muestra es inhomogénea y que las diferencias entre las formas de las resonancias medidas y calculadas está relacionada con el cálculo del coeficiente de autoabsorción en la muestra. La inhomogeneidad de la muestra puede estar relacionada con el tamaño de los granos de polvo que la forman, que es comparable al espesor del mismo [160]. Desafortunadamente, el código SAMMY no permite tratar este tipo de inhomogeneidades en la densidad superficial de la muestra. Por tanto, el análisis de la sección eficaz del ^{240}Pu se realizó sólo en el rango de energías por encima de 110 eV, donde la corrección de los efectos de autoabsorción son despreciables.

A pesar de las dificultades en el análisis asociadas a la fabricación de la muestra, el análisis combinado de captura y transmisión ha permitido obtener la sección eficaz de captura neutrónica de ^{240}Pu de forma precisa entre 110 eV y 2 keV. En este rango de energía, se observan un total

de 158 resonancias que han sido analizadas una a una. El análisis de las resonancias más prominentes ha proporcionado los valores de los parámetros de la resonancias, mientras que para el análisis de la resonancias más débiles fue necesario fijar la anchura radiativa a un valor constante de $\langle\Gamma_\gamma\rangle=29.7$ eV, resultante del análisis de las resonancias prominentes. Dado que el TAC es capaz de detectar no sólo las cascadas emitidas en reacciones de captura sino también en las reacciones de fisión los datos tiene un fondo resonante. Esta medida y con la metodología aplicada ha determinado que el valor límite de la sección eficaz de fisión para el cual el análisis de las resonancias no es preciso es de $\sigma_f/\sigma_\gamma = 0,15$. En el caso del ^{240}Pu sólo hay 15 de estas resonancias en el intervalo de energías analizado, de la cuales sólo 8 son realmente prominentes.

El estudio estadístico de los parámetros del resonancias indica que los parámetros de las resonancias en diferentes rangos de energía son coherentes, ya que no se ha observado dependencia alguna de la anchura radiativa promedio $\langle\Gamma_\gamma\rangle$ o de la *strength function* S_0 con la energía del neutrón. Por otro lado, el valor $\langle\Gamma_\gamma\rangle= 29,7$ meV encontrado en este trabajo es 7% inferior al de las evaluaciones JEFF-3.1 y JENDL-3.3 (31,8 meV) y un 3 % inferior al de la evaluación ENDF/B-VII (30,6 meV). Sin embargo, este valor están en mejor acuerdo que las evaluaciones con respecto a la anchura radiativa de la primera resonancia a 1.05 eV (29.15 ± 0.6 MeV [76, 21] que es la más estudiada y mejor conocida de todas. La Tabla A.3 contiene los valores recomendados para la descripción estadística de la sección eficaz y también muestra los valores de las evaluaciones y las medidas anteriores más relevantes.

	n_TOF	JEFF-3.1	ENDF/B-VII	
		JENDL-3.3 Bouland et al. [76]	Mughabghab [121] Weston et al. [124]	RIPL-2
$D_0(\text{eV})$	12.0 ± 0.6	12.06 ± 0.60	13.6 ± 0.7	12.4 ± 0.7
$\langle\Gamma_\gamma\rangle(\text{meV})$	29.7 ± 1.9	31.8 ± 1.5	30.6 ± 2	30.7 ± 2.5
$S_0(10^{-4})$	1.08 ± 0.02	1.03 ± 0.07	0.93 ± 0.08	1.05 ± 0.10

Table A.3: *Parámetros estadísticos de la sección eficaz del ^{240}Pu de este trabajo comparados con los valores de las evaluaciones y las medidas más relevantes.*

La sección eficaz de captura neutrónica obtenida en este trabajo se ha promediado en 14 rangos de energía utilizando NJOY-99 y se ha comparado con las secciones eficaces evaluadas. La comparación se muestra en la Figura A.10. Los resultados indican que a energías inferiores a 700 eV la sección eficaz de n_TOF y las evaluaciones están todas de acuerdo dentro de las incertidumbres. Sin embargo, para el rango de energías de 700 eV a 2 keV, donde hay diferencias significativas entre las diferentes evaluaciones, los datos de este trabajo están de acuerdo las evaluaciones JEFF-3.1 y JENDL-3.3 pero no con la evaluación ENDF/B-VII. Como todas las evaluaciones están basadas en los datos de transmisión de Kolar et al. [154], estos resultados sugieren que hay un problema en la evaluación ENDF/B-VII.

La sección eficaz de captura del ^{240}Pu medida en este trabajo es, a pesar de los problemas asociados a la fabricación de la muestra, la medida con mayor resolución, estadística y precisión realizada hasta la fecha. Los resultados del análisis de los datos otorgan una mayor confianza

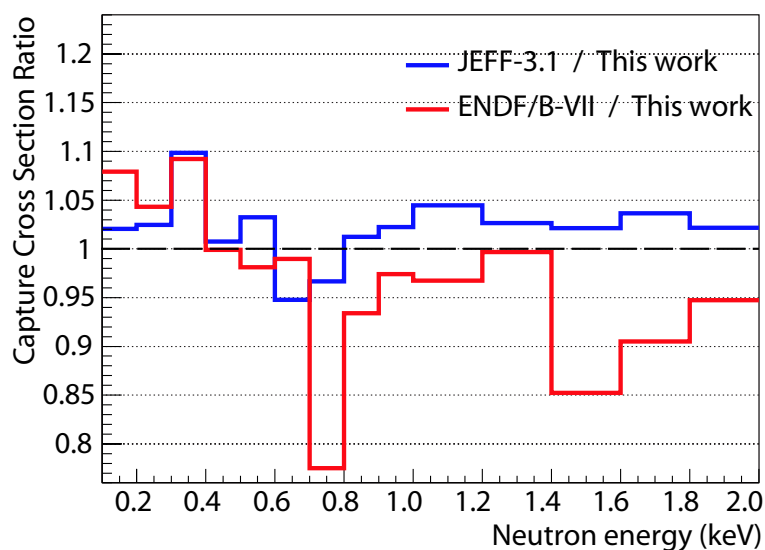


Figure A.10: Comparación de la sección eficaz de captura neutrónica del ^{240}Pu medida en n_TOF con las secciones eficaces evaluadas.

a las evaluaciones JEFF-3.1 y JENDL-3.3. De hecho, el grado de acuerdo entre los datos de n_TOF y estas evaluaciones sugiere que la incertidumbre del 10% estimada para la sección eficaz captura del ^{240}Pu , y que está relacionada con la incompatibilidad entre las diferentes evaluaciones, debería ser revisada y que una precisión del 5% parece más realista. Un de las conclusiones del análisis, es que sería necesario realizar una nueva medida de transmisión que confirme o rectifique las medidas de Kolar et al., que datan del año 1968. El resultado de este análisis formará parte de la base de datos EXFOR [162] será tenido en cuenta en las próximas evaluaciones.

A.2.3 Mejoras para futuras medidas

La calidad de las medidas presentadas en este trabajo ha permitido que los resultados del análisis sean considerados para las próximas evaluaciones. Hasta hoy ninguna de las evaluaciones recientes relativas de estos dos isótopos incluía datos de captura en la zona de resonancias resueltas debido a la baja resolución y el rango de energía limitado de los datos experimentales disponibles. Esta situación ha cambiado con la disponibilidad de las medidas de n_TOF.

Una parte muy importante de los resultados de este trabajo, es que la experiencia adquirida y las técnicas de análisis desarrolladas permitirán realizar en el futuro medidas en la instalación n_TOF más precisas. Atendiendo a las principales fuentes de incertidumbre que se han encontrado en este trabajo, se proponen una serie de modificaciones que permitirán eliminarlas, o al menos se reducirán de forma significativa:

- La sustitución de las muestras encapsuladas en titanio por un nuevo tipo de muestra en el que el actínido se deposita sobre una lámina de aluminio delgada dentro de un tubo en vacío y que también cumple la normativa ISO-2919. Este nuevo tipo de muestras permitirán realizar medidas hasta energías de al menos 20 keV, y posiblemente hasta 100 keV. las nuevas muestras han sido diseñadas por el CIEMAT y su construcción está siendo negociada con el VNIIE-Sarov.
- La utilización de un nuevo absorbente de neutrones fabricado en polietileno borado que reducirán aún más la eficiencia de detección para neutrones dispersados en la muestra. El nuevo absorbente ya ha sido diseñado [123] y construido en los talleres del CIEMAT.
- La utilización de un blindaje para neutrones fabricado en polietileno borado rodeando todas las ventanas de vacío de la línea del haz para minimizar el fondo asociado a la dispersión de neutrones en las mismas. Este sistema de blindaje ya ha sido diseñado [123] y está siendo construido en los talleres del CIEMAT.
- El uso de soportes activos para las muestras que permitan realizar medidas combinadas de fisión y captura. Este tipo de soportes se encuentra en fase de diseño.
- La utilización de muestras metálicas en lugar de muestras en forma de óxido con el fin de eliminar las inhomogeneidades relacionadas con el tamaño de los granos del polvo, que suelen ser comparables con el espesor de las muestras.
- La supervisión directa del proceso de fabricación de las muestras con el fin de garantizar su composición isotópica.

Además, gracias a las técnica y el software desarrollados será posible realizar el análisis de las nuevas medidas en un tiempo inferior. La fiabilidad de los resultados estará asegurada gracias al exhaustivo proceso de validación de la técnica de análisis.

Appendix B

Resonance parameters of ^{237}Np between 1 eV and 500 eV

The following resonance parameters are the result of the analysis with SAMMY of the capture and transmission data of ^{237}Np in the energy interval between 1 eV and 500 eV. The resonance parameters correspond to the Reich-Moore formalism and are given in the ENDF format [21]. All the resonances are considered s -waves ($\ell=0$).

<i>Energy(eV)</i>	<i>Spin</i>	$\Gamma_n(\text{eV})$	$\Gamma_\gamma(\text{eV})$	$\Gamma_{FA}(\text{eV})$	<i>MAT MF/T #</i>
1.320868079	3.000000000	3.148437-5	3.901911-2	8.670400-6	9346 2151 9
1.477576351	2.000000000	1.824787-4	4.253700-2	1.328500-6	9346 2151 10
1.968559118	3.000000000	1.418707-5	3.937686-2	4.186700-6	9346 2151 11
2.996260856	3.000000000	1.685350-9	4.048619-2	2.000000-6	9346 2151 12
3.865051320	3.000000000	2.102649-4	3.987437-2	5.800000-6	9346 2151 13
4.264291742	2.000000000	3.300655-5	4.170892-2	6.400000-6	9346 2151 14
4.862922785	2.000000000	3.949670-5	3.739017-2	4.400000-6	9346 2151 15
5.777446079	3.000000000	5.309797-4	4.216232-2	1.050000-5	9346 2151 16
6.376475856	3.000000000	7.885382-5	3.879866-2	3.388000-6	9346 2151 17
6.676754052	2.000000000	1.444167-5	5.370555-2	2.178200-5	9346 2151 18
7.189804875	2.000000000	8.769190-6	4.346541-2	2.487700-5	9346 2151 19
7.422121444	3.000000000	1.223663-4	3.773907-2	1.000000-5	9346 2151 20
7.653673835	2.000000000	2.178014-6	5.350766-2	1.477500-4	9346 2151 21
8.306050832	3.000000000	8.972431-5	3.645467-2	3.600000-6	9346 2151 22
8.976719250	3.000000000	1.011676-4	3.577488-2	2.270000-5	9346 2151 23
9.298520977	2.000000000	6.058009-4	4.135578-2	1.503600-6	9346 2151 24
10.23206253	2.000000000	2.910743-5	4.606073-2	1.324700-6	9346 2151 25
10.68051139	3.000000000	4.192856-4	3.631993-2	4.100000-6	9346 2151 26
10.84390392	3.000000000	6.925342-4	3.955148-2	1.300000-6	9346 2151 27
11.09673971	2.000000000	1.026156-3	4.464030-2	1.200000-6	9346 2151 28

12.20234017 3.00000000 4.887246-5 4.350354-2 2.273700-6 9346 2151 29
 12.61794379 2.00000000 9.158578-4 4.040229-2 2.819000-8 9346 2151 30
 13.13850398 3.00000000 1.747366-5 4.261205-2 1.365700-5 9346 2151 31
 14.33560187 2.00000000 1.153091-6 4.044719-2 6.826300-6 9346 2151 32
 15.78744622 3.00000000 6.037308-5 2.303880-2 3.863800-6 9346 2151 33
 15.91987571 3.00000000 3.171494-5 3.189423-2 2.603200-6 9346 2151 34
 16.08755620 2.00000000 1.070625-3 4.094250-2 1.203400-6 9346 2151 35
 16.86005074 2.00000000 2.977731-4 3.822529-2 4.307800-6 9346 2151 36
 17.59617898 3.00000000 1.540611-4 3.689619-2 8.205300-6 9346 2151 37
 17.89697964 2.00000000 1.715552-5 3.980408-2 1.309200-5 9346 2151 38
 17.96396429 3.00000000 3.065742-6 3.935202-2 4.347000-5 9346 2151 39
 18.89542374 2.00000000 4.561984-5 3.973204-2 6.375800-9 9346 2151 40
 19.12814777 3.00000000 8.366809-5 3.143968-2 1.031800-5 9346 2151 41
 19.92739681 3.00000000 6.060549-5 2.245038-2 1.361100-5 9346 2151 42
 20.40240880 2.00000000 1.351153-3 4.012348-2 2.257300-7 9346 2151 43
 21.10068106 3.00000000 4.316070-4 3.461373-2 3.500000-6 9346 2151 44
 21.32406991 2.00000000 3.165535-5 5.041999-2 1.747100-5 9346 2151 45
 22.01971374 2.00000000 1.501871-3 4.251682-2 3.292400-6 9346 2151 46
 22.86956192 3.00000000 3.779551-4 4.067666-2 6.600000-6 9346 2151 47
 23.67960179 3.00000000 1.435865-3 4.587502-2 3.725900-7 9346 2151 48
 23.99269160 2.00000000 1.707199-4 4.065856-2 9.708300-6 9346 2151 49
 24.79897020 3.00000000 2.587079-5 4.347694-2 4.036800-6 9346 2151 50
 24.98803354 3.00000000 3.534962-3 4.605348-2 9.100000-6 9346 2151 51
 26.19654140 3.00000000 1.969159-4 3.984505-2 1.190000-4 9346 2151 52
 26.56621592 3.00000000 2.322991-3 4.297714-2 5.900000-5 9346 2151 53
 27.09358178 2.00000000 3.958220-5 4.737564-2 5.649700-7 9346 2151 54
 28.46274096 2.00000000 1.045721-4 7.231799-2 1.055200-7 9346 2151 55
 28.62066380 3.00000000 2.853343-5 4.563256-2 6.877700-9 9346 2151 56
 28.93871317 2.00000000 1.399775-4 5.055878-2 .0876410-9 9346 2151 57
 29.48557998 2.00000000 8.453654-5 4.839434-2 3.000000-4 9346 2151 58
 30.42481552 3.00000000 3.116209-3 4.160381-2 2.490000-4 9346 2151 59
 30.75267792 2.00000000 3.608099-4 5.207496-2 1.800000-5 9346 2151 60
 31.31136302 3.00000000 2.407330-4 4.078807-2 1.500000-5 9346 2151 61
 31.67198776 3.00000000 4.740275-5 6.423486-2 5.200000-5 9346 2151 62
 32.41454872 2.00000000 1.429242-5 4.867882-2 9.322900-9 9346 2151 63
 33.43135820 3.00000000 3.988100-4 4.849124-2 8.815800-6 9346 2151 64
 33.91284116 2.00000000 4.462921-4 2.836575-2 9.190500-7 9346 2151 65
 34.07476751 3.00000000 4.101190-5 3.318798-2 9.872200-6 9346 2151 66
 34.69126741 3.00000000 1.534747-4 2.948677-2 6.115800-6 9346 2151 67
 35.21150603 2.00000000 3.785816-4 2.670710-2 2.020600-7 9346 2151 68
 36.39256497 3.00000000 1.160411-4 3.042213-2 9.385500-8 9346 2151 69
 36.84608638 2.00000000 9.528999-5 3.665572-2 1.131100-7 9346 2151 70
 37.16623922 3.00000000 1.105081-3 3.114671-2 3.640000-4 9346 2151 71
 37.82701104 2.00000000 3.208587-5 4.097756-2 1.071900-4 9346 2151 72
 38.04525274 2.00000000 1.689047-4 3.377440-2 5.539000-5 9346 2151 73
 38.20338497 3.00000000 1.212019-3 4.073617-2 3.800000-5 9346 2151 74

38.93026235	3.00000000	8.782655-4	3.111461-2	8.340000-4	9346	2151	75
39.06163862	2.00000000	3.390800-4	3.525683-2	1.048100-4	9346	2151	76
39.26246995	3.00000000	4.708637-4	1.649459-2	7.910000-4	9346	2151	77
39.80332412	2.00000000	1.059697-4	3.601275-2	1.500000-3	9346	2151	78
39.94584238	3.00000000	4.404435-4	2.649972-2	7.015000-3	9346	2151	79
41.37033092	3.00000000	1.899602-3	3.071711-2	6.340000-4	9346	2151	80
42.40613372	3.00000000	6.520385-5	2.199386-2	1.275200-6	9346	2151	81
42.82904288	3.00000000	3.709453-6	4.080000-2	2.056700-6	9346	2151	82
42.83657299	3.00000000	7.663816-5	3.374430-2	1.057000-3	9346	2151	83
43.65375125	2.00000000	3.155893-4	4.080000-2	1.090500-8	9346	2151	84
44.19975954	2.00000000	1.326494-5	4.080000-2	1.095000-3	9346	2151	85
45.35916844	2.00000000	5.743324-6	4.080000-2	2.567300-4	9346	2151	86
45.73174966	2.00000000	5.007425-4	4.080000-2	1.841300-5	9346	2151	87
46.04958229	3.00000000	5.737501-4	4.080000-2	7.900000-4	9346	2151	88
46.36892894	3.00000000	2.522264-3	4.080000-2	1.900000-5	9346	2151	89
47.33770838	2.00000000	2.781857-3	4.080000-2	6.595000-7	9346	2151	90
48.45196290	2.00000000	1.018501-4	4.080000-2	1.400000-4	9346	2151	91
48.78353089	3.00000000	3.384533-4	4.080000-2	2.452300-5	9346	2151	92
48.91605828	2.00000000	1.669416-4	4.080000-2	1.122200-5	9346	2151	93
49.33586874	2.00000000	1.043493-5	4.080000-2	2.916200-5	9346	2151	94
49.82933587	3.00000000	4.113154-3	4.080000-2	8.000000-6	9346	2151	95
50.33011747	2.00000000	1.254814-3	4.080000-2	5.724700-5	9346	2151	96
50.42514757	3.00000000	6.129989-3	4.080000-2	6.600000-5	9346	2151	97
51.70650514	3.00000000	9.584911-5	4.080000-2	2.787700-5	9346	2151	98
52.22348491	2.00000000	4.028051-4	4.080000-2	7.921700-8	9346	2151	99
52.65701162	2.00000000	8.562207-4	4.080000-2	1.799200-5	9346	2151	100
53.06688091	3.00000000	6.793236-5	4.080000-2	2.478500-5	9346	2151	101
53.89720953	2.00000000	4.800233-4	4.080000-2	4.452600-6	9346	2151	102
54.27664903	2.00000000	1.742761-4	4.080000-2	1.638800-5	9346	2151	103
55.05495787	3.00000000	2.576890-4	4.080000-2	1.981300-7	9346	2151	104
56.02707500	2.00000000	1.219955-3	4.080000-2	1.804100-9	9346	2151	105
56.17031283	3.00000000	6.751086-4	4.080000-2	1.131300-5	9346	2151	106
56.60427815	2.00000000	3.375965-5	4.080000-2	1.561600-6	9346	2151	107
56.73942532	3.00000000	1.247794-5	4.080000-2	8.242500-6	9346	2151	108
57.45413541	2.00000000	5.020740-6	4.080000-2	1.970500-6	9346	2151	109
58.40491487	3.00000000	3.818492-4	4.080000-2	1.720100-5	9346	2151	110
58.65451821	3.00000000	2.157772-4	4.080000-2	5.283600-5	9346	2151	111
59.52621220	2.00000000	2.339197-3	4.080000-2	4.127000-6	9346	2151	112
60.07082860	3.00000000	2.233702-3	4.080000-2	1.316600-5	9346	2151	113
60.97875688	3.00000000	1.516844-3	4.080000-2	2.770500-6	9346	2151	114
61.27375243	3.00000000	1.639470-5	4.080000-2	0.00000000	9346	2151	115
61.66659368	3.00000000	1.147137-4	4.080000-2	2.960700-5	9346	2151	116
61.68146126	3.00000000	3.265734-4	4.080000-2	1.238900-7	9346	2151	117
62.39772376	2.00000000	3.965381-4	4.080000-2	3.529900-6	9346	2151	118
62.51643336	3.00000000	1.375566-3	4.080000-2	1.038300-5	9346	2151	119
62.93028775	3.00000000	1.441554-3	4.080000-2	8.488800-7	9346	2151	120

63.43850634	2.00000000	6.915229-5	4.080000-2	8.670800-5	9346	2151	121
63.98220738	3.00000000	2.331346-4	4.080000-2	5.338100-6	9346	2151	122
64.98681664	3.00000000	8.464323-4	4.080000-2	3.005300-6	9346	2151	123
65.72374413	3.00000000	3.727336-3	4.080000-2	4.241600-6	9346	2151	124
66.51782418	2.00000000	2.877898-5	4.080000-2	3.257900-7	9346	2151	125
67.05645381	2.00000000	1.661991-5	4.080000-2	1.704900-7	9346	2151	126
67.20641963	2.00000000	1.309826-5	4.080000-2	5.795700-7	9346	2151	127
67.49821964	3.00000000	4.754684-3	4.080000-2	7.132800-6	9346	2151	128
67.98921924	2.00000000	2.800743-3	4.080000-2	4.182000-6	9346	2151	129
68.83741799	3.00000000	3.257312-4	4.080000-2	1.194500-5	9346	2151	130
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71.45628897	2.00000000	2.441416-3	4.080000-2	6.330000-7	9346	2151	134
71.56584739	3.00000000	5.309162-4	4.080000-2	0.00000000	9346	2151	135
71.93162577	2.00000000	4.636752-6	4.080000-2	1.745300-7	9346	2151	136
72.60145211	2.00000000	9.781103-6	4.080000-2	8.113500-8	9346	2151	137
73.90529848	3.00000000	2.773996-4	4.080000-2	1.490200-7	9346	2151	138
74.31686878	2.00000000	1.683972-3	4.080000-2	2.656700-6	9346	2151	139
74.61484224	3.00000000	4.170448-4	4.080000-2	1.038500-5	9346	2151	140
75.14393919	2.00000000	1.413737-4	4.080000-2	5.128400-6	9346	2151	141
75.30408591	3.00000000	1.008386-5	4.080000-2	2.351800-7	9346	2151	142
76.23928063	3.00000000	3.370916-5	4.080000-2	8.135000-6	9346	2151	143
76.60432325	2.00000000	1.732197-4	4.080000-2	2.009300-5	9346	2151	144
77.02267544	3.00000000	2.765464-4	4.080000-2	3.843300-8	9346	2151	145
77.58646874	2.00000000	4.244220-5	4.080000-2	1.231300-5	9346	2151	146
78.08948479	3.00000000	1.888731-5	4.080000-2	3.772600-7	9346	2151	147
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79.29583559	2.00000000	2.857021-3	4.080000-2	1.075500-7	9346	2151	150
79.99337586	3.00000000	9.925970-6	4.080000-2	9.963600-8	9346	2151	151
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81.64890941	2.00000000	4.533046-4	4.080000-2	7.733000-8	9346	2151	154
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83.42295798	2.00000000	3.230002-3	4.080000-2	1.348200-6	9346	2151	157
83.69307766	3.00000000	3.164262-3	4.080000-2	5.931600-6	9346	2151	158
83.83390707	2.00000000	2.463119-3	4.080000-2	7.066600-6	9346	2151	159
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86.13005446	2.00000000	1.022879-3	4.080000-2	2.379700-7	9346	2151	161
86.55387698	3.00000000	4.733952-3	4.080000-2	5.855200-6	9346	2151	162
87.61605707	2.00000000	1.663486-3	4.080000-2	3.026200-6	9346	2151	163
87.77561013	3.00000000	1.789499-3	4.080000-2	9.332300-7	9346	2151	164
88.19937106	3.00000000	8.761169-4	4.080000-2	9.689200-6	9346	2151	165
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115.8753267	2.000000000	2.317180-3	4.080000-2	6.368200-5	9346	2151	214
116.8246981	3.000000000	3.435240-4	4.080000-2	3.445700-5	9346	2151	215
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119.1415268	2.000000000	1.347875-3	4.080000-2	9.341300-4	9346	2151	217
119.5404057	3.000000000	6.964913-4	4.080000-2	1.027400-2	9346	2151	218
120.1931263	3.000000000	3.186433-4	4.080000-2	2.563800-4	9346	2151	219
121.6550967	2.000000000	1.196980-4	4.080000-2	1.172700-4	9346	2151	220
122.0796349	3.000000000	2.746831-4	4.080000-2	8.573000-6	9346	2151	221
122.4064312	2.000000000	4.506457-5	4.080000-2	1.376200-4	9346	2151	222
123.5550057	3.000000000	7.245390-5	4.080000-2	1.543400-4	9346	2151	223
123.9084985	3.000000000	3.384981-4	4.080000-2	1.361300-4	9346	2151	224
124.5524938	2.000000000	7.356540-5	4.080000-2	1.548200-4	9346	2151	225
125.0942677	3.000000000	1.028281-3	4.080000-2	1.403400-4	9346	2151	226
125.7268607	3.000000000	1.759994-3	4.080000-2	2.040700-6	9346	2151	227
125.9721335	2.000000000	1.006007-3	4.080000-2	1.011800-4	9346	2151	228
126.3385299	3.000000000	1.252785-3	4.080000-2	4.923200-9	9346	2151	229
127.0726532	3.000000000	2.372405-4	4.080000-2	7.893200-8	9346	2151	230
127.4041956	2.000000000	6.214823-4	4.080000-2	5.587400-7	9346	2151	231
127.8227198	2.000000000	3.131374-4	4.080000-2	3.546300-5	9346	2151	232
128.6466428	3.000000000	5.892794-5	4.080000-2	1.172100-6	9346	2151	233
129.4687942	2.000000000	6.341443-4	4.080000-2	2.375000-5	9346	2151	234
129.7173799	3.000000000	6.186692-4	4.080000-2	1.212100-5	9346	2151	235
130.4660484	2.000000000	2.576044-5	4.059700-2	3.620200-7	9346	2151	236
131.4622101	2.000000000	7.987915-4	4.080000-2	4.172500-7	9346	2151	237
131.6218164	2.000000000	2.041664-5	4.059900-2	2.784700-7	9346	2151	238
131.9360932	3.000000000	8.820758-4	4.080000-2	4.175000-5	9346	2151	239
132.6205127	2.000000000	1.384197-4	4.080000-2	1.046500-5	9346	2151	240
133.0044291	2.000000000	1.515495-4	4.080000-2	6.733300-7	9346	2151	241
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134.1759047	3.000000000	2.625937-4	4.080000-2	8.800300-6	9346	2151	243
134.3649617	3.000000000	4.465194-5	4.080000-2	5.466000-7	9346	2151	244
135.4849944	2.000000000	1.193843-4	4.080000-2	4.680700-6	9346	2151	245
136.1963947	2.000000000	5.172744-5	4.080000-2	1.487800-5	9346	2151	246
136.7538879	2.000000000	6.766787-5	4.080000-2	2.617900-6	9346	2151	247
136.8006850	3.000000000	1.415349-4	4.080000-2	7.627900-6	9346	2151	248
137.4749036	3.000000000	2.261038-3	4.080000-2	2.759000-5	9346	2151	249
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138.2257590	2.000000000	8.110916-4	4.080000-2	3.389300-7	9346	2151	251
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139.0444459	2.000000000	2.843975-3	4.080000-2	1.281500-5	9346	2151	253
139.5781940	3.000000000	4.383676-4	4.080000-2	1.295600-5	9346	2151	254
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141.2966829	2.000000000	1.768582-3	4.080000-2	1.886800-6	9346	2151	257
141.5638255	3.000000000	3.430853-4	4.080000-2	3.285800-5	9346	2151	258

142.3146123	3.000000000	4.772394-4	4.080000-2	2.583900-5	9346	2151	259
143.3391475	2.000000000	6.058726-5	4.080000-2	1.389100-4	9346	2151	260
144.3864852	3.000000000	3.873990-3	4.080000-2	2.968000-6	9346	2151	261
144.7412612	2.000000000	3.973909-4	4.080000-2	3.280700-7	9346	2151	262
145.6193712	2.000000000	8.339844-4	4.080000-2	9.991900-6	9346	2151	263
146.5881043	2.000000000	1.446957-3	4.080000-2	2.362200-5	9346	2151	264
146.8803949	3.000000000	4.295735-4	4.080000-2	2.587400-6	9346	2151	265
147.3540271	3.000000000	1.578078-3	4.080000-2	2.594600-6	9346	2151	266
148.5955849	3.000000000	9.511462-4	4.080000-2	1.571600-5	9346	2151	267
149.6588588	2.000000000	1.425517-3	4.080000-2	2.071700-8	9346	2151	268
150.0226031	2.000000000	1.746003-3	4.080000-2	7.319700-6	9346	2151	269
150.5136444	3.000000000	4.053718-3	4.080000-2	1.269100-5	9346	2151	270
150.8475944	3.000000000	8.422107-4	4.080000-2	5.765400-7	9346	2151	271
152.0166387	2.000000000	4.964520-4	4.080000-2	4.547900-8	9346	2151	272
152.3448766	3.000000000	2.168959-3	4.080000-2	4.310200-6	9346	2151	273
154.2244758	3.000000000	1.776934-3	4.080000-2	1.489500-6	9346	2151	274
154.5046556	2.000000000	7.953169-4	4.080000-2	2.966200-5	9346	2151	275
155.4353421	3.000000000	9.967185-4	4.080000-2	1.996000-5	9346	2151	276
156.4730103	3.000000000	1.542685-3	4.080000-2	1.134600-7	9346	2151	277
156.9065891	3.000000000	7.456202-4	4.080000-2	5.938600-8	9346	2151	278
158.0845888	3.000000000	1.187187-3	4.080000-2	2.020200-5	9346	2151	279
158.4872250	3.000000000	3.442614-3	4.080000-2	4.953700-7	9346	2151	280
159.4996814	3.000000000	8.652457-5	4.080000-2	8.029300-5	9346	2151	281
160.1830955	2.000000000	4.038228-5	4.080000-2	1.740700-5	9346	2151	282
160.7176941	3.000000000	4.397172-3	4.080000-2	3.244000-6	9346	2151	283
161.1717764	2.000000000	1.940922-4	4.080000-2	7.171000-7	9346	2151	284
161.9321693	3.000000000	8.921297-4	4.080000-2	1.962700-7	9346	2151	285
163.0238965	3.000000000	3.337656-3	4.080000-2	1.448400-5	9346	2151	286
164.2470294	2.000000000	5.058424-4	4.080000-2	7.787100-6	9346	2151	287
164.3878162	3.000000000	1.646342-3	4.080000-2	3.167900-6	9346	2151	288
165.2684591	3.000000000	4.682558-4	4.080000-2	1.407900-6	9346	2151	289
166.8410937	3.000000000	2.669123-3	4.080000-2	1.616500-5	9346	2151	290
166.9965952	2.000000000	2.121340-3	4.080000-2	0.00000000	9346	2151	291
168.7316336	3.000000000	4.436969-4	4.080000-2	2.996900-6	9346	2151	292
169.0699854	3.000000000	4.082815-3	4.080000-2	2.101700-6	9346	2151	293
169.9878811	3.000000000	1.305708-3	4.080000-2	8.490200-6	9346	2151	294
170.3935382	3.000000000	6.763719-4	4.080000-2	1.084700-5	9346	2151	295
171.0696717	2.000000000	4.674541-3	4.080000-2	3.282900-5	9346	2151	296
171.2516335	2.000000000	9.767991-5	4.000000-2	4.365200-8	9346	2151	297
172.3252729	2.000000000	1.302350-3	4.080000-2	2.533300-6	9346	2151	298
173.0303586	3.000000000	1.615214-3	4.080000-2	9.191100-6	9346	2151	299
173.3669415	3.000000000	7.762012-4	4.080000-2	3.453800-6	9346	2151	300
173.7078701	3.000000000	1.324222-4	4.080000-2	2.760800-5	9346	2151	301
174.8720636	3.000000000	1.087139-3	4.080000-2	1.776900-5	9346	2151	302
175.3320437	2.000000000	3.696431-3	4.080000-2	3.828200-5	9346	2151	303
176.3029321	3.000000000	3.816012-3	4.080000-2	7.361300-6	9346	2151	304

176.9512283	3.000000000	5.247431-3	4.080000-2	1.461300-5	9346	2151	305
177.2613180	3.000000000	4.660099-4	4.080000-2	6.577500-5	9346	2151	306
180.1859546	3.000000000	5.563923-3	4.080000-2	1.700900-5	9346	2151	307
181.0842347	2.000000000	3.321220-3	4.080000-2	1.430300-5	9346	2151	308
181.5567431	2.000000000	5.523828-4	4.080000-2	7.453600-6	9346	2151	309
182.4592624	3.000000000	1.795512-3	4.080000-2	5.983100-6	9346	2151	310
182.7772770	2.000000000	3.982843-4	4.080000-2	1.156100-5	9346	2151	311
183.3681277	2.000000000	2.367140-3	4.080000-2	4.895800-5	9346	2151	312
184.1548049	2.000000000	6.897718-3	4.080000-2	3.858500-5	9346	2151	313
184.5247986	2.000000000	3.875255-4	4.080000-2	1.502500-4	9346	2151	314
185.5783910	2.000000000	4.051391-4	4.080000-2	9.631500-6	9346	2151	315
186.0856661	2.000000000	3.412965-4	4.080000-2	2.060000-5	9346	2151	316
186.3761303	2.000000000	8.364150-4	4.080000-2	5.150300-5	9346	2151	317
188.0571690	2.000000000	1.428106-2	4.080000-2	1.708000-4	9346	2151	318
189.1998652	2.000000000	7.357122-3	4.080000-2	3.857100-5	9346	2151	319
190.6851486	3.000000000	3.888085-3	4.080000-2	3.976400-5	9346	2151	320
190.9715717	2.000000000	1.933660-3	4.080000-2	9.423400-5	9346	2151	321
191.8003901	3.000000000	1.035298-3	4.080000-2	1.414700-6	9346	2151	322
193.6542202	2.000000000	4.011863-3	4.080000-2	1.343500-4	9346	2151	323
194.5769355	3.000000000	1.103131-3	4.080000-2	3.359300-4	9346	2151	324
195.0987843	2.000000000	3.169173-4	4.080000-2	5.511900-3	9346	2151	325
195.8091923	2.000000000	3.391410-3	4.080000-2	7.680700-4	9346	2151	326
196.6562957	3.000000000	9.899619-3	4.080000-2	1.659800-4	9346	2151	327
196.8413222	3.000000000	2.907365-3	4.080000-2	1.026700-4	9346	2151	328
197.9341395	2.000000000	3.037132-4	4.080000-2	2.401000-3	9346	2151	329
198.6942913	2.000000000	7.196668-4	4.080000-2	1.193200-4	9346	2151	330
199.8639068	2.000000000	1.176015-3	4.080000-2	1.264800-5	9346	2151	331
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 392.2977493 3.00000000 1.747861-3 4.080000-2 2.272900-5 9346 2151 556
 394.7776189 2.00000000 8.893029-4 4.080000-2 2.054300-5 9346 2151 557
 397.1650811 2.00000000 1.481579-3 4.080000-2 2.310000-5 9346 2151 558
 398.6357891 3.00000000 1.195808-3 4.080000-2 2.054600-5 9346 2151 559
 399.8812624 2.00000000 5.195050-3 4.080000-2 1.038300-5 9346 2151 560
 401.3921013 2.00000000 1.742894-3 4.080000-2 2.000600-5 9346 2151 561
 401.9233709 3.00000000 2.471528-3 4.080000-2 2.126300-5 9346 2151 562
 402.8518400 2.00000000 2.731028-3 4.080000-2 1.959400-5 9346 2151 563
 403.3105857 2.00000000 4.845445-3 4.080000-2 2.075900-5 9346 2151 564
 404.3082427 3.00000000 5.203308-4 4.080000-2 2.045300-5 9346 2151 565
 405.1428977 2.00000000 7.684469-3 4.080000-2 2.143700-5 9346 2151 566
 405.9700623 2.00000000 3.754727-4 4.080000-2 1.994900-5 9346 2151 567
 407.2337851 2.00000000 2.417850-3 4.080000-2 2.080300-5 9346 2151 568
 407.9177408 2.00000000 2.482175-3 4.080000-2 1.994900-5 9346 2151 569
 408.4057355 3.00000000 1.353648-3 4.080000-2 2.123600-5 9346 2151 570
 410.2317817 3.00000000 1.112016-3 4.080000-2 1.954200-5 9346 2151 571
 410.8049776 2.00000000 2.837211-3 4.080000-2 2.088600-5 9346 2151 572
 412.2255583 3.00000000 1.486401-4 4.080000-2 1.977600-5 9346 2151 573
 413.1990215 2.00000000 4.054959-3 4.080000-2 2.132500-5 9346 2151 574
 413.9552917 2.00000000 2.627040-3 4.080000-2 1.975500-5 9346 2151 575
 414.5293784 3.00000000 3.779394-3 4.080000-2 2.112200-5 9346 2151 576
 415.0436639 2.00000000 4.484930-3 4.080000-2 2.085800-5 9346 2151 577
 415.8622902 2.00000000 7.211337-3 4.080000-2 1.214800-4 9346 2151 578
 417.2869991 3.00000000 4.966696-4 4.080000-2 2.860800-6 9346 2151 579
 418.2578797 3.00000000 3.375858-3 4.080000-2 2.336700-4 9346 2151 580

418.8227639	3.00000000	2.308256-3	4.080000-2	1.121000-5	9346	2151	581
420.5748026	3.00000000	6.076123-3	4.080000-2	2.527900-4	9346	2151	582
421.0604838	2.00000000	8.557596-3	4.080000-2	1.507600-8	9346	2151	583
421.6107964	2.00000000	5.007380-3	4.080000-2	3.039900-7	9346	2151	584
422.8341753	3.00000000	6.721834-3	4.080000-2	2.471700-4	9346	2151	585
423.8381052	3.00000000	6.452050-4	4.080000-2	3.191900-6	9346	2151	586
424.9878822	2.00000000	5.121492-4	4.080000-2	1.648600-6	9346	2151	587
426.6746280	2.00000000	5.281171-4	4.080000-2	1.012000-5	9346	2151	588
426.7085196	3.00000000	5.734220-3	4.080000-2	1.961400-3	9346	2151	589
427.5091066	3.00000000	1.463992-3	4.080000-2	3.461000-3	9346	2151	590
428.2844224	3.00000000	1.409714-4	4.080000-2	1.610200-4	9346	2151	591
429.6764969	2.00000000	5.861050-4	4.080000-2	1.442400-3	9346	2151	592
430.3527605	2.00000000	1.887506-6	4.080000-2	1.975700-5	9346	2151	593
430.3696836	2.00000000	1.890717-6	4.080000-2	1.975700-5	9346	2151	594
431.5505356	3.00000000	1.468334-3	4.080000-2	2.492300-5	9346	2151	595
432.2352872	2.00000000	2.459431-3	4.080000-2	1.835800-5	9346	2151	596
432.5741948	2.00000000	3.128353-4	4.080000-2	2.144400-5	9346	2151	597
433.2004112	2.00000000	3.970266-4	4.080000-2	1.785100-5	9346	2151	598
434.2235840	2.00000000	3.245026-4	4.080000-2	6.273500-7	9346	2151	599
435.7565241	2.00000000	1.188152-3	4.080000-2	2.536300-5	9346	2151	600
436.3611917	3.00000000	3.882856-3	4.080000-2	6.902300-6	9346	2151	601
437.5651241	3.00000000	1.469358-3	4.080000-2	2.214400-5	9346	2151	602
438.1829336	2.00000000	6.780556-3	4.080000-2	1.420900-5	9346	2151	603
438.9599878	3.00000000	2.582864-3	4.080000-2	2.137900-5	9346	2151	604
439.9776710	2.00000000	7.217607-3	4.080000-2	9.043700-6	9346	2151	605
441.2169562	3.00000000	7.763081-4	4.080000-2	2.042900-5	9346	2151	606
442.7176264	2.00000000	5.374895-4	4.080000-2	1.940700-5	9346	2151	607
443.6873545	2.00000000	2.448677-4	4.080000-2	2.024200-5	9346	2151	608
444.9388173	2.00000000	1.807107-4	4.080000-2	2.005300-5	9346	2151	609
444.9803743	2.00000000	1.008454-2	4.080000-2	1.809900-5	9346	2151	610
447.2168706	2.00000000	1.566845-3	4.080000-2	2.068200-5	9346	2151	611
447.8784142	3.00000000	1.442326-3	4.080000-2	2.074100-5	9346	2151	612
448.7138943	2.00000000	1.217530-2	4.080000-2	1.818800-8	9346	2151	613
449.4303701	3.00000000	4.294492-3	4.080000-2	2.022400-5	9346	2151	614
450.1847378	3.00000000	1.098380-3	4.080000-2	1.546800-5	9346	2151	615
451.5482736	2.00000000	4.110138-3	4.080000-2	1.959900-5	9346	2151	616
452.2090553	3.00000000	9.443455-3	4.080000-2	1.468000-5	9346	2151	617
452.9003513	2.00000000	9.598310-3	4.080000-2	2.009300-5	9346	2151	618
453.2271741	2.00000000	3.308191-3	4.080000-2	1.864200-5	9346	2151	619
453.9246272	3.00000000	4.001865-3	4.080000-2	1.889300-5	9346	2151	620
455.0033828	2.00000000	3.298048-3	4.080000-2	2.031700-5	9346	2151	621
456.6666760	3.00000000	9.468762-3	4.080000-2	9.096000-5	9346	2151	622
457.7074121	3.00000000	4.222335-3	4.080000-2	2.072000-5	9346	2151	623
458.2974542	2.00000000	1.381101-3	4.080000-2	1.934300-5	9346	2151	624
459.1233048	3.00000000	5.370367-4	4.080000-2	2.012500-5	9346	2151	625
460.5274368	2.00000000	3.169846-3	4.080000-2	1.925500-5	9346	2151	626

461.5409197 2.00000000 8.831534-3 4.080000-2 5.613300-7 9346 2151 627
 461.8846607 2.00000000 7.001804-4 4.080000-2 3.615500-5 9346 2151 628
 463.0760586 2.00000000 6.559827-3 4.080000-2 3.154800-5 9346 2151 629
 463.8217434 3.00000000 9.143743-4 4.080000-2 2.753100-8 9346 2151 630
 465.2684459 3.00000000 2.491143-3 4.080000-2 2.198100-4 9346 2151 631
 465.8129766 2.00000000 3.014069-3 4.080000-2 9.255200-6 9346 2151 632
 467.0738382 3.00000000 2.781239-3 4.080000-2 3.856600-6 9346 2151 633
 468.6634533 2.00000000 8.009683-3 4.080000-2 .1565900-9 9346 2151 634
 469.9924945 3.00000000 2.172845-3 4.080000-2 7.461500-5 9346 2151 635
 470.9746252 3.00000000 1.418055-2 4.080000-2 3.108200-4 9346 2151 636
 472.0696707 2.00000000 5.043021-4 4.080000-2 2.071600-6 9346 2151 637
 473.2379748 3.00000000 3.023570-4 4.080000-2 4.781300-7 9346 2151 638
 475.1376392 2.00000000 5.104804-3 4.080000-2 1.330100-3 9346 2151 639
 476.1414784 2.00000000 2.092521-3 4.080000-2 1.489400-3 9346 2151 640
 478.0615688 3.00000000 9.689889-4 4.080000-2 1.657900-4 9346 2151 641
 479.1233053 3.00000000 7.223748-3 4.080000-2 2.127900-7 9346 2151 642
 480.4523070 3.00000000 2.683178-4 4.080000-2 8.265000-6 9346 2151 643
 480.8330573 2.00000000 8.915304-4 4.080000-2 2.575000-4 9346 2151 644
 481.6127535 3.00000000 2.015290-3 4.080000-2 1.036500-4 9346 2151 645
 482.6363621 3.00000000 8.267430-3 4.080000-2 1.117300-5 9346 2151 646
 483.2670213 2.00000000 1.461713-3 4.080000-2 4.730700-6 9346 2151 647
 484.0408759 2.00000000 2.688399-3 4.080000-2 7.717000-5 9346 2151 648
 485.0572028 2.00000000 8.377701-4 4.080000-2 1.878000-5 9346 2151 649
 486.1253993 2.00000000 5.959359-3 4.080000-2 8.209000-8 9346 2151 650
 487.0472249 2.00000000 1.154783-3 4.080000-2 1.250700-4 9346 2151 651
 487.7960648 3.00000000 1.053510-2 4.080000-2 1.692900-5 9346 2151 652
 488.3495486 2.00000000 1.048212-3 4.080000-2 3.328100-5 9346 2151 653
 489.5319578 2.00000000 1.377987-4 4.080000-2 1.984500-5 9346 2151 654
 490.1909051 3.00000000 6.714093-3 4.080000-2 3.837200-5 9346 2151 655
 490.2233821 2.00000000 3.144824-3 4.080000-2 5.332700-6 9346 2151 656
 491.2799136 3.00000000 5.505543-4 4.080000-2 1.226000-4 9346 2151 657
 492.3540837 2.00000000 1.113274-3 4.080000-2 0.00000000 9346 2151 658
 493.6359572 3.00000000 2.515043-3 4.080000-2 1.014800-5 9346 2151 659
 494.8212900 2.00000000 6.858772-3 4.080000-2 2.234400-8 9346 2151 660
 495.1545353 2.00000000 6.883205-4 4.080000-2 5.296100-5 9346 2151 661
 496.0036165 3.00000000 1.357355-2 4.080000-2 2.067600-5 9346 2151 662
 496.8010028 2.00000000 3.467288-3 4.080000-2 0.00000000 9346 2151 663
 497.4602383 2.00000000 1.544864-3 4.080000-2 9.748500-6 9346 2151 664
 498.1578587 3.00000000 1.584536-3 4.080000-2 3.702000-6 9346 2151 665
 499.1441343 2.00000000 2.739318-4 4.080000-2 2.324400-5 9346 2151 666
 499.7411570 2.00000000 2.377910-2 4.080000-2 2.100800-5 9346 2151 667

Appendix C

Resonance parameters of ^{240}Pu between 110 eV and 2 keV

The following resonance parameters are the result of the analysis with SAMMY of the capture and transmission data of ^{240}Pu in the energy interval between 110 eV and 2 keV. The resonance parameters correspond to the Reich-Moore formalism and are given in the ENDF format [21]. All the resonances are considered s -waves ($\ell=0$).

<i>Energy(eV)</i>	<i>Spin</i>	$\Gamma_n(\text{eV})$	$\Gamma_\gamma(\text{eV})$	$\Gamma_{FA}(\text{eV})$	<i>MAT MF/T #</i>
121.6569920	0.50000000	1.179252-2	4.914995-2	4.300300-5	9440 2151 20
125.7880434	0.50000000	1.937889-5	3.100000-2	2.000000-5	9440 2151 21
130.8636862	0.50000000	1.954123-4	4.774834-2	2.413000-4	9440 2151 22
135.3794496	0.50000000	1.392215-2	5.795896-2	2.636400-5	9440 2151 23
151.9916852	0.50000000	1.198501-2	4.442067-2	3.567700-4	9440 2151 24
162.7609097	0.50000000	8.305390-3	3.594397-2	5.208000-6	9440 2151 25
170.1224252	0.50000000	1.158933-2	5.077146-2	1.349500-4	9440 2151 26
185.8893728	0.50000000	1.358900-2	4.978350-2	8.954000-6	9440 2151 27
192.1471443	0.50000000	1.884698-4	3.065000-2	1.284000-4	9440 2151 28
195.6721642	0.50000000	4.161521-4	3.100000-2	1.200000-4	9440 2151 29
197.2657699	0.50000000	4.356884-5	3.228902-2	1.200000-4	9440 2151 30
199.6802822	0.50000000	1.011987-3	5.586198-2	1.373000-4	9440 2151 31
239.2069013	0.50000000	1.271367-2	2.649324-2	4.086000-5	9440 2151 32
260.4442321	0.50000000	1.628364-2	7.265950-2	9.113300-5	9440 2151 33
287.0881385	0.50000000	1.560334-1	2.899495-2	3.771900-4	9440 2151 34
304.9317401	0.50000000	6.817071-3	3.587269-2	6.923900-5	9440 2151 35
313.3659539	0.50000000	1.243082-4	3.100000-2	3.000000-4	9440 2151 36
318.2694644	0.50000000	5.056867-3	4.456433-2	1.400000-4	9440 2151 37
320.6476364	0.50000000	1.475739-2	4.176476-2	5.570100-5	9440 2151 38
333.1392731	0.50000000	9.455846-5	3.100000-2	2.000000-5	9440 2151 39

338.3816023	0.50000000	5.404537-3	2.444914-2	5.842000-6	9440	2151	40
345.9470415	0.50000000	1.468282-2	2.600241-2	1.385900-4	9440	2151	41
363.7273026	0.50000000	3.486501-2	2.698944-2	2.446800-5	9440	2151	42
371.9729949	0.50000000	1.408826-2	2.721684-2	4.024100-5	9440	2151	43
393.0413675	0.50000000	1.931182-4	3.100000-2	2.000000-5	9440	2151	44
404.9447190	0.50000000	1.233621-1	2.940304-2	4.717500-4	9440	2151	45
418.9295144	0.50000000	6.202750-3	3.116276-2	7.077000-5	9440	2151	46
445.8407729	0.50000000	1.923492-3	2.677474-2	2.210000-4	9440	2151	47
449.7953633	0.50000000	1.702201-2	2.777301-2	4.316000-5	9440	2151	48
466.5388778	0.50000000	3.349216-3	3.183223-2	3.438000-4	9440	2151	49
473.3421151	0.50000000	4.113583-3	3.070500-2	0.00000000	9440	2151	50
493.8798606	0.50000000	5.629859-3	3.605275-2	1.218000-4	9440	2151	51
499.2709486	0.50000000	1.829557-2	3.264259-2	1.218600-4	9440	2151	52
509.8745943	0.50000000	1.194410-3	3.100000-2	5.000000-5	9440	2151	53
512.5815015	0.50000000	5.420974-4	3.100000-2	5.000000-5	9440	2151	54
514.3205849	0.50000000	2.026193-2	3.368819-2	7.429100-5	9440	2151	55
526.2788127	0.50000000	9.607572-4	3.100000-2	0.00000000	9440	2151	56
530.6130226	0.50000000	7.914874-4	3.100000-2	4.800000-4	9440	2151	57
546.4561614	0.50000000	2.564256-2	4.098736-2	3.872600-5	9440	2151	58
553.1882550	0.50000000	1.627888-2	4.248152-2	2.168300-4	9440	2151	59
566.2637911	0.50000000	2.672327-2	3.558244-2	1.830200-4	9440	2151	60
584.2013772	0.50000000	1.783655-3	3.100000-2	7.700000-4	9440	2151	61
596.7826976	0.50000000	5.438668-2	3.230701-2	4.478100-5	9440	2151	62
607.9264029	0.50000000	2.109854-2	3.632343-2	2.107400-5	9440	2151	63
632.3250585	0.50000000	1.535134-2	3.100574-2	1.836000-4	9440	2151	64
637.4243836	0.50000000	1.159037-2	3.717866-2	5.091300-5	9440	2151	65
650.0877990	0.50000000	2.746413-4	3.100000-2	1.700000-3	9440	2151	66
664.9945995	0.50000000	1.984581-1	3.096511-2	3.856200-4	9440	2151	67
678.3703808	0.50000000	2.281247-2	3.876292-2	8.988500-4	9440	2151	68
711.8507324	0.50000000	1.316745-3	3.100000-2	7.806000-4	9440	2151	69
743.4474341	0.50000000	1.537843-3	3.100000-2	1.370000-3	9440	2151	70
749.9097055	0.50000000	6.835064-2	3.852591-2	1.131900-2	9440	2151	71
758.6953184	0.50000000	7.608648-3	3.588985-2	5.764000-4	9440	2151	72
777.9877481	0.50000000	6.427202-4	3.100000-2	5.700000-4	9440	2151	73
781.8485101	0.50000000	6.256604-3	3.661189-2	-1.8588000	9440	2151	74
790.7197824	0.50000000	3.032442-2	4.185971-2	1.398800-2	9440	2151	75
810.3688941	0.50000000	2.134895-1	3.911811-2	1.397600-2	9440	2151	76
819.7231064	0.50000000	1.118129-1	3.089659-2	1.007900-3	9440	2151	77
845.4921572	0.50000000	1.042767-2	3.305785-2	2.999000-4	9440	2151	78
854.6836213	0.50000000	4.828365-2	3.272541-2	2.401000-4	9440	2151	79
876.1870158	0.50000000	1.380035-2	3.212273-2	8.688100-4	9440	2151	80
891.3496207	0.50000000	8.964075-2	3.273255-2	1.274500-3	9440	2151	81
900.1356813	0.50000000	8.649685-4	3.100000-2	1.200000-2	9440	2151	82
903.8208108	0.50000000	2.343577-2	2.774471-2	7.322000-4	9440	2151	83
908.7157948	0.50000000	7.927710-2	2.914710-2	3.237000-5	9440	2151	84
915.0326063	0.50000000	3.308494-2	3.738130-2	3.398000-4	9440	2151	85

943.2934408	0.50000000	1.064974-1	3.237838-2	2.976600-4	9440	2151	86
958.2060670	0.50000000	6.803713-2	3.088769-2	7.036000-5	9440	2151	87
969.9647446	0.50000000	9.751752-4	3.100000-2	5.000000-3	9440	2151	88
971.1476340	0.50000000	6.545746-2	3.438742-2	6.005000-5	9440	2151	89
979.1764981	0.50000000	6.931281-3	3.098848-2	4.366000-4	9440	2151	90
997.3176245	0.50000000	1.673629-3	3.079137-2	0.00000000	9440	2151	91
1001.648465	0.50000000	1.148855-1	3.029267-2	1.078900-3	9440	2151	92
1012.115772	0.50000000	8.737632-4	3.100000-2	7.322100-3	9440	2151	93
1023.848014	0.50000000	4.296562-3	3.100000-2	4.555000-4	9440	2151	94
1028.163856	0.50000000	1.040452-3	3.100000-2	6.534000-3	9440	2151	95
1041.472865	0.50000000	1.141691-2	3.048124-2	1.599200-4	9440	2151	96
1045.084086	0.50000000	4.689846-3	3.100000-2	1.085100-3	9440	2151	97
1072.277077	0.50000000	9.589050-2	3.089841-2	1.840000-4	9440	2151	98
1099.504360	0.50000000	8.646252-2	2.422482-2	2.769900-4	9440	2151	99
1114.859809	0.50000000	2.281702-3	3.100000-2	3.400000-4	9440	2151	100
1128.575209	0.50000000	5.923049-2	2.643350-2	6.977000-4	9440	2151	101
1133.550993	0.50000000	7.462298-3	3.100000-2	2.397000-4	9440	2151	102
1138.902374	0.50000000	1.905523-3	3.100000-2	7.322100-3	9440	2151	103
1142.516069	0.50000000	4.523621-2	2.752604-2	1.214400-4	9440	2151	104
1159.151473	0.50000000	2.561732-2	2.429229-2	3.075900-4	9440	2151	105
1174.897794	0.50000000	1.351351-3	3.100000-2	2.770000-3	9440	2151	106
1185.246217	0.50000000	1.634889-1	3.080988-2	1.236800-4	9440	2151	107
1190.469023	0.50000000	1.040507-1	3.238797-2	1.785900-4	9440	2151	108
1208.573851	0.50000000	6.196078-2	3.066493-2	1.159900-4	9440	2151	109
1227.628662	0.50000000	1.104017-2	3.100000-2	4.000000-4	9440	2151	110
1236.207602	0.50000000	1.436071-2	3.100000-2	4.150000-4	9440	2151	111
1254.390567	0.50000000	7.106165-2	3.332753-2	1.624000-3	9440	2151	112
1281.149225	0.50000000	8.732072-3	3.100000-2	4.100000-4	9440	2151	113
1283.845794	0.50000000	9.938767-3	3.100000-2	7.322100-3	9440	2151	114
1292.229661	0.50000000	3.971856-3	3.100000-2	7.322100-3	9440	2151	115
1299.942227	0.50000000	2.673976-1	2.981155-2	9.150400-4	9440	2151	116
1327.755154	0.50000000	3.894822-1	3.018770-2	6.805000-4	9440	2151	117
1344.726406	0.50000000	2.746796-2	3.100000-2	4.534000-4	9440	2151	118
1350.813040	0.50000000	5.676075-3	3.100000-2	3.000000-5	9440	2151	119
1362.425605	0.50000000	5.552644-3	3.100000-2	1.262100-3	9440	2151	120
1376.579417	0.50000000	6.673175-2	3.189166-2	9.682800-4	9440	2151	121
1388.412422	0.50000000	1.684502-2	3.100000-2	6.296000-3	9440	2151	122
1401.166531	0.50000000	1.001740-2	3.100000-2	-2.0855000	9440	2151	123
1408.378509	0.50000000	1.804430-2	3.100000-2	8.520300-2	9440	2151	124
1425.707723	0.50000000	4.403713-2	2.985389-2	5.485200-3	9440	2151	125
1428.059453	0.50000000	9.751337-3	3.100000-2	2.225700-3	9440	2151	126
1449.017630	0.50000000	3.421886-2	3.100000-2	2.028200-3	9440	2151	127
1450.378693	0.50000000	2.883607-2	3.531583-2	2.181100-3	9440	2151	128
1462.582405	0.50000000	2.138173-2	3.100000-2	1.069800-3	9440	2151	129
1465.987141	0.50000000	1.642743-3	3.100000-2	1.170000-2	9440	2151	130
1481.082648	0.50000000	1.018309-2	3.100000-2	1.241200-3	9440	2151	131

1492.087337	0.50000000	2.199089-3	3.100000-2	2.500000-3	9440	2151	132
1496.734160	0.50000000	4.395036-3	3.100000-2	1.000000-4	9440	2151	133
1528.972953	0.50000000	5.124819-3	3.100000-2	6.476700-3	9440	2151	134
1540.129440	0.50000000	1.014317-1	3.031797-2	2.799700-4	9440	2151	135
1549.006799	0.50000000	1.685295-1	2.980356-2	1.509400-4	9440	2151	136
1563.255928	0.50000000	1.204025-1	2.964171-2	9.999700-5	9440	2151	137
1575.016343	0.50000000	1.157059-1	2.989739-2	3.459800-4	9440	2151	138
1602.432730	0.50000000	1.834097-3	3.100000-2	1.000000-4	9440	2151	139
1609.212215	0.50000000	4.280686-2	3.100000-2	6.809400-4	9440	2151	140
1620.989986	0.50000000	3.365088-2	3.100000-2	2.900000-4	9440	2151	141
1628.088015	0.50000000	5.500588-3	3.100000-2	9.018500-4	9440	2151	142
1642.585538	0.50000000	1.057025-1	3.241717-2	1.609900-4	9440	2151	143
1662.278228	0.50000000	6.579760-2	3.028531-2	3.778800-4	9440	2151	144
1666.813704	0.50000000	6.327771-3	3.100000-2	1.000000-4	9440	2151	145
1687.469527	0.50000000	3.970574-2	3.100000-2	6.552600-4	9440	2151	146
1707.121575	0.50000000	1.613136-3	3.100000-2	1.000000-3	9440	2151	147
1718.256777	0.50000000	2.233328-3	3.100000-2	7.322100-3	9440	2151	148
1723.627225	0.50000000	6.899609-2	3.260213-2	2.140400-3	9440	2151	149
1741.352059	0.50000000	2.165681-2	3.100000-2	6.094600-4	9440	2151	150
1763.189895	0.50000000	4.850169-2	3.100000-2	5.780000-4	9440	2151	151
1771.144714	0.50000000	1.115366-2	3.245581-2	1.100000-4	9440	2151	152
1778.392857	0.50000000	4.656629-1	2.973099-2	8.528500-5	9440	2151	153
1787.777917	0.50000000	2.721234-3	3.100000-2	1.657300-3	9440	2151	154
1792.885191	0.50000000	1.982381-3	3.100000-2	7.322100-3	9440	2151	155
1808.718348	0.50000000	3.677143-3	3.100000-2	1.754500-3	9440	2151	156
1834.254641	0.50000000	2.843522-3	3.100000-2	7.322100-3	9440	2151	157
1840.842105	0.50000000	1.282071-1	3.306300-2	1.034500-2	9440	2151	158
1852.420968	0.50000000	3.916815-2	3.100000-2	3.639700-3	9440	2151	159
1872.488651	0.50000000	7.257044-2	3.598809-2	4.163200-3	9440	2151	160
1886.180475	0.50000000	4.313698-3	3.100000-2	1.396400-3	9440	2151	161
1901.139593	0.50000000	2.137014-1	3.037339-2	3.296100-3	9440	2151	162
1916.055627	0.50000000	3.518600-2	3.100000-2	9.561600-2	9440	2151	163
1935.880002	0.50000000	1.955837-3	3.100000-2	1.81040000	9440	2151	164
1942.922255	0.50000000	7.560565-3	3.100000-2	6.095800-3	9440	2151	165
1948.597446	0.50000000	1.030425-1	3.115559-2	8.834700-3	9440	2151	166
1956.400000	0.50000000	2.762500-1	3.083600-2	2.436800-2	9440	2151	167
1972.517760	0.50000000	7.623504-2	3.100000-2	1.898100-3	9440	2151	168
1990.813720	0.50000000	1.188974-1	2.680351-2	5.575500-5	9440	2151	169
1998.160988	0.50000000	6.073553-3	3.100000-2	4.982800-5	9440	2151	170

