

Measurement of ^{50}Cr and ^{53}Cr neutron capture cross sections for nuclear technology at CERN n_TOF and HiSPANoS

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A mi familia.
A Alma.

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Ponme, ponme esa cinta otra vez
Pónmela hasta que se arranquen
*Los cachitos de hierro y de **cromo***
Al cantar como tú sabes.
Kiko Veneno

Abstract

Nuclear energy has been recognized as crucial in terms of decreasing the CO₂ emissions due to carbon sources while satisfying the increasing energy demands worldwide. The associated development towards a more efficient and safer nuclear energy requires nuclear data of high quality, serving criticality safety benchmarks to identify and set priorities about those that need to be improved.

In this context, because of its significant abundance in stainless steel, chromium plays an important role in some benchmarks focused on structural materials. Indeed, it has been found that current $\sim 30\%$ discrepancies in the neutron capture cross sections of ^{50}Cr and ^{53}Cr have an impact of about 1% on k_{eff} . Such high discrepancy is related to the fact that the old chromium measurements could suffer from neutron-scattering effects, while the most recent one (Guber et al. from 2011) seems to have sample related issues. This was pointed out in the most recent evaluation (INDEN), which proposed an important increase of these cross sections but called for new data to confirm or deny it. For this reason, the Nuclear Energy Agency (NEA) opened an entry in its High Priority Request List (HPRL) to measure the $^{50,53}\text{Cr}$ (n, γ) cross sections with an accuracy of 8 to 10% between 1 and 100 keV, with emphasis on the region below 10 keV.

This work is dedicated to the measurements carried out as a response to the NEA request. First, a time-of-flight measurement was performed at the n_TOF facility of CERN (Switzerland), in which the capture yield of both isotopes was measured and a new set of resonance parameters has been proposed. In addition, for the first time a neutron activation measurement of ^{50}Cr was performed at the HiSPANoS facility of CNA (Spain) to determine its MACS at $kT = 30$ keV. The results from both experiments are in agreement, while the comparison to the evaluations indicates a clear cross section overestimation by 20-40% in the recent INDEN evaluation for both isotopes. When all evaluations are considered, our results are in better agreement with CENDL-3.2, which is the only one not considering the most recent data by Guber et al. (2011). With these findings, the new differential and integral cross sections reported herein should contribute to impulse new evaluation effort and solve the chromium puzzle.

Resumen

El papel de la energía nuclear se ha reconocido como crucial para reducir la emisión de CO_2 y, al mismo tiempo, satisfacer la creciente demanda de energía a nivel mundial. El desarrollo hacia una energía nuclear más eficiente y segura requiere de datos nucleares de alta calidad, que sirvan a los benchmarks de criticidad y seguridad para identificar y priorizar aquellos datos que necesitan ser mejorados.

En este contexto, debido a su abundancia en el acero inoxidable, el cromo desempeña un papel importante en algunos benchmarks centrados en materiales estructurales. De hecho, se ha constatado que las actuales discrepancias del $\sim 30\%$ en la sección eficaz de captura de neutrones del ^{50}Cr y ^{53}Cr tienen un impacto de alrededor del 1% en k_{eff} . Esta discrepancia tan elevada está relacionada con el hecho de que las medidas anteriores del cromo pudieron sufrir efectos de scattering de neutrones, mientras que la más reciente (Guber et al. de 2011) parece tener problemas relacionados con las muestras. Esto se señaló en la evaluación más reciente (INDEN), que propuso un incremento de dichas secciones eficaces, pero pidió nuevos datos para confirmarlo o desmentirlo. Por esta razón, la Agencia de Energía Nuclear (NEA) abrió una entrada en su Lista de Peticiones de Alta Prioridad (HPRL) para medir la sección eficaz de captura de $^{50,53}\text{Cr}$ entre 1 y 100 keV con una precisión del 8 al 10% , con énfasis en la región por debajo de 10 keV.

Este trabajo se centra en las medidas llevadas a cabo como respuesta a dicha petición. En primer lugar, se realizó una medida de tiempo de vuelo en la instalación n_TOF del CERN (Suiza), en la que se midió el yield de captura de ambos isótopos, proponiendo un nuevo conjunto de parámetros de resonancia. Además, por primera vez se ha llevado a cabo una medida de activación con neutrones del ^{50}Cr en la instalación HiSPANoS del CNA (España), para determinar su MACS a $kT = 30$ keV. Los resultados de ambos concuerdan, mientras que la comparación con las evaluaciones indica una clara sobrestimación en la sección eficaz de ambos isótopos del 20-40% por parte de INDEN. Considerando todas ellas, nuestros resultados concuerdan mejor con CENDL-3.2, que es la única que no usa los datos de Guber et al. (2011). Con estos hallazgos, las nuevas secciones eficaces diferenciales e integrales aquí reportadas deberían contribuir a resolver el rompecabezas del cromo e impulsar nuevos esfuerzos en cuanto a su evaluación.

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List of Acronyms

AWF Averaged Weighting Factor.
BCT Beam Current Transformer.
BIF Beam Interception Factor.
CN Compound Nucleus.
CT Computed Tomography.
DAQ Data Acquisition System.
EAR Experimental Area.
FGM Free Gas Model.
HPRL High Priority Request List.
MACS Maxwellian Averaged Cross Section.
MC Monte Carlo.
MGAS Micromegas.
NEA Nuclear Energy Agency.
PHWT Pulse Height Weighting Technique.
PPAC Parallel Plate Avalanche Counters.
PS Proton-Synchrotron.
PSA Pulse Shape Analysis.
RF Resolution Function.
RM Reich-Moore approximation.
RMS Root Mean Square.
RRR Resolved Resonances Region.
RWF Resonance Weighting Factor.
SAEX Sample Exchanger.
SiMon Silicon Monitor.
SRM Saturated Resonance Method.
TAC Total Absorption Calorimeter.
TED Total Energy Detection.
TOF Time-of-Flight.
WF Weighting Function.

Part I

Introduction

1 | Motivations and goals

In this Chapter, a brief introduction about nuclear power and its roll in the energy-related problems that the world is facing is presented. In this context, the optimization of current and the design of future reactors relies on accurate nuclear data. Particularly, the neutron capture in the chromium isotopes ^{50}Cr and ^{53}Cr , major components of the stainless steel used in nuclear reactors, has been found to play a significant role in criticality safety calculations, and a call has been made by the Nuclear Energy Agency (NEA) to improve the corresponding cross sections. Two new measurements have been proposed and performed to address this issue, conforming the topic of this work.

1.1. Nuclear energy overview

According to the last report by the International Atomic Energy Agency (IAEA) [1], the energy consumption worldwide is expected to increase by about 30% by the end of 2050. In particular, the electricity production is expected to double. The dominant electricity production source has been coal over the last 40 years, even when the low-carbon sources have increased considerably over the same period. Currently, nuclear power contributes around 9% to the total electricity production, but its role in achieving net zero emissions by 2050 has been recognized on the recent 23th United Nations Climate Change conference (COP28). Accordingly, a declaration to triple nuclear energy capacity by that year has been signed by 22 countries. This demand requires nuclear energy to progress in terms of efficiency and in a better use of the uranium and thorium resources. In addition, the public concerns require also focusing on the improvement of the reactors safety performance. These should be achieved through a worldwide effort on *criticality safety* research programs (see Section 1.3).

The basic operation of a nuclear reactor is based on a self-sustained nuclear reaction. When some very heavy nuclei such as U or Pu are bombarded with neutrons there is a large probability that they undergo a *fission* reaction, in which they break apart into smaller nuclei, releasing energy and a few more neutrons (see Fig. 1.1). If only one of the emitted neutrons produces a fission in the next generation and the rest are captured

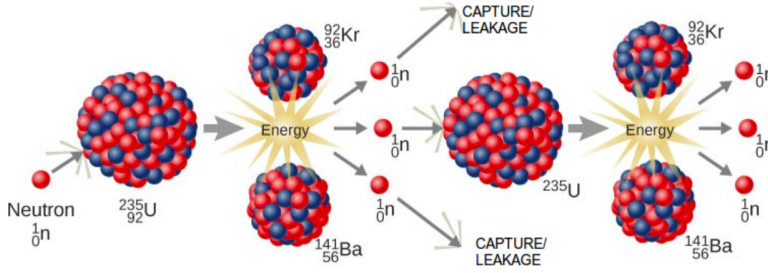


Figure 1.1: Schematic illustration of the neutron-induced fission chain reaction. From Ref. [2].

or escape the system, then a self-sustained and energy-releasing process is achieved.

The first self-sustained fission chain was accomplished by E. Fermi in 1942 using natural uranium and graphite in the so-called Chicago Pile-1 (CP-1) [3]. After World War II, the rapid development of nuclear technology led to the first commercial nuclear reactors by the end of the 50s, and as of 2022, there were 411 nuclear reactors in operation and 85 under construction around the world [1].

As in the rest of thermal power plants, the electricity in a nuclear reactor is produced by turbines moved by water steam, which is created by heating up water using the energy released, in this case, by the fission process. Nuclear reactors can be classified into thermal reactors, if the fissions are produced by thermalized or moderated neutrons (~ 25 meV), or fast reactors, if the neutron energies are higher (> 10 keV). Most of the current nuclear reactors are thermal, being fueled with ^{235}U and using light water as moderator. These are called Light Water Reactors (LWR), and depending on their design can be classified as Boiling Water Reactors (BWR) (see Fig. 1.3) or Pressurized Water Reactors (PWR). The next generation of reactors (Gen-IV) aims to offer new solutions to the current limitations of these systems, mainly in terms of safety and nuclear waste. Several design concepts have been selected as the future Gen-IV reactors (the detailed list can be found in [4]), and recently, there has also been an increasing interest in small modular reactors [5], which provide energy in a more local area and can also be used in non-electrical applications.

1.2. Role of nuclear data: neutron cross sections

Nuclear Data refers to every quantity or observable related to any specific property of a nucleus and the interaction of other particles with the nuclear system [6]. Needs for nuclear data can be found in a large variety of applications, such as the following [7]:

- Nuclear technology, design of nuclear (fission and fusion) reactors, fuel cycle, waste management,
- Production of radioisotopes, for instance for medical applications,
- Astrophysics, mainly related to the nucleosynthesis of elements inside the stars,
- Nuclear structure and reaction models.

Regarding nuclear technology, nuclear data is crucial for describing all the nuclear phenomena happening inside a reactor. For example, the number of neutrons inside the reactor is given by the *neutron transport equation*, which requires of reaction rates derived from the *cross sections* of all reactions that could occur:

- Neutron absorption reactions such a fission (n,f), capture (n, γ) or He production (n, α) on the fuel and structural materials,
- Scattering reactions like (n,n), (n,n' γ) or (n,xn),
- Emission probability of secondary particles like neutrons, γ -rays or fission fragments, and their spectra,
- Mass distribution of the fission fragments and details of their decay properties.

The usual steps followed by nuclear data constitute the so-called *Nuclear Data Cycle*, outlined in Fig. 1.2. According to the demands of the different fields, nuclear data measurements are designed to fulfill their necessities in terms of measured quantities and uncertainties. Measurements are then planned, performed and analyzed, and the results are collected in the Experimental Nuclear Reaction Data (EXFOR) [8] database. The data can be differential (like cross sections or energy and angular distributions) or integral, that can be use to validate differential data if measured with enough accuracy.

The next step in the nuclear data path is its *evaluation*, so it can be suitable for applications. An evaluation consists on the careful analysis and selection of the measured data with its uncertainty, and through a combination of theoretical predictions and validation a "recommended" value is given. The result are the Evaluated Nuclear Data Files (ENDF) [9]. There are plenty of evaluated libraries that compiles every evaluated quantity in a common format so it is easily available for any researcher. Some of the current main libraries are JEFF-3.3 [10], ENDF/B-VIII.0 [11], JENDL-5 [12], CENDL-3.2 [13] or BROND-3.1 [14].

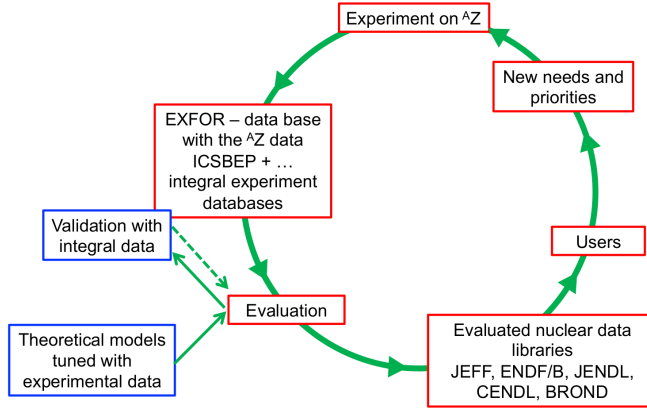


Figure 1.2: Schematic of the so-called Nuclear Data Cycle. More details about each step is given throughout the text. From Ref. [7].

1.3. Criticality safety benchmarks

Nowadays, the nuclear data accuracy required for some fields, specially for nuclear technology, is very demanding. A more accurate nuclear data increases the efficiency and the safety of a nuclear reactor. One of the most important parameters regarding reactor safety is the effective neutron multiplication factor k_{eff} , defined as the ratio between the number of neutrons produced in a generation with respect to the previous one. A similar factor is k_{∞} , equivalent to k_{eff} but for an infinite system, so neutrons can only be lost by capture because there is no leakage. Therefore, a nuclear reactor (or any neutron multiplication system) can be classified according to its *criticality* depending on its working principle:

- If $k_{eff} < 1$, the system is *subcritical*, so the number of neutrons is reduced with each generation.
- If $k_{eff} = 1$, the system is *critical*, and the reaction chain is controlled and self-sustained. A nuclear reactor must work in sub- or critical conditions.
- If $k_{eff} > 1$, the system is *supercritical*, so the number of neutrons in each generation increases exponentially. Nuclear weapons work in this kind of regime.

A major effort is periodically carried out by the scientific community when new experimental data or evaluation is available to study how it affects the criticality calculations, and to determine which particular data set needs to be measured or re-evaluated as a priority. The identification of the priority data needed is done by studying which one contributes more to the uncertainty of some parameter calculated through the so-called

sensitivity analysis (SA). The goal of a sensitivity analysis is to detect the isotopes, reaction cross sections and energy ranges that mostly affect the calculation of a certain quantity k , in this case the criticality. This is done by studying the sensitivity coefficient S_α , defined as [15]

$$S_\alpha = \frac{\partial k}{\partial \alpha}, \quad (1.1)$$

where α represents the input data, for example a cross section within an energy range. In this way, the impact in the results of a calculation when varying the input data can be assessed. The sensitivity coefficients depends on the system studied; e. g., the same cross section can led to a positive S_α in one nuclear system and to a negative value in another. The latest Monte Carlo neutron transport codes allow to perform very accurate SA for criticality safety.

The uncertainty of each nuclear data is propagated and influences the overall uncertainty of the result. This is assessed through *uncertainty analysis* (UA), and it is not a trivial subject since usually the uncertainties of different energy ranges, different cross sections of the same isotopes or even different isotopes are correlated. The information about the nuclear data uncertainty and its correlations is given by the *covariance* and *correlation matrices*.

The quality of some nuclear data library and its impact on criticality calculations are determined with SA/UA analysis in the so-called *integral criticality benchmarks*. A criticality benchmark can be understood as an "evaluated experiment", in which the performance of some nuclear data is tested by simulating the integral experiments that use it, and comparing the results against the very well known experimental values. For example, if an evaluated cross section of some isotope is updated, to test its new performance some benchmarks specially sensitive to that particular material or reaction channel can be performed. Often, the results from this kind of exercise are not straightforward to interpret, because of the large number of materials and reactions taken place in a nuclear reactor and their correlations.

During the development of the nuclear energy industry, thousands of criticality safety experiments have been performed all over the world. The International Criticality Safety Benchmark Evaluation Project [16] (ICSBEP) aims to compile and standardize all critical and subcritical benchmark experimental data, so a common and accessible format is implemented in the different analysis codes and libraries. This has facilitated the access to nuclear data qualification to the nuclear physics community.

1.4. Status and performance of $^{50,53}\text{Cr}$ capture cross sections.

Chromium is a metal characterized by its very high melting point, which is commonly used in metallurgy. Natural chromium is formed by four isotopes with the abundances displayed in Table 1.1. ^{50}Cr is considered unstable but with an extremely long half-life ($T_{1/2} > 1.3 \cdot 10^{18}$ y). Because of their proximity to the neutron shell closure at $N = 28$, with ^{52}Cr being "magic" with exactly $N = 28$, these isotopes are characterized by relatively low level densities.

Table 1.1: Isotopic composition of natural chromium.

Isotope	Abundance (%)
^{50}Cr	4.345(13)
^{52}Cr	83.789(18)
^{53}Cr	9.501(17)
^{54}Cr	2.365(7)

Chromium is an important component of stainless steel (SS), with abundances between 11 to 26% depending on the kind of steel. Because of its properties, SS is used as a structural material in a large number of applications, including nuclear technology. Indeed, as shown in Fig. 1.3, SS is found in many parts of the nuclear reactor assembly, mostly in the vessel. More recently, chromium has been proposed to be used in new Enhanced-Accident Tolerant Fuel (E-ATF) to improve the performance of the nuclear fuel in normal operation and to decrease the damage during accident conditions, by doping the UO_2 fuel pellets with Cr_2O_3 and by chromium-coating the fuel cladding [17]. For this reason, chromium has to be taken into account when performing criticality benchmarks. As detailed in the following, it has been observed that the effects of chromium in selected benchmarks with large amounts of this material is dominated by neutron capture of ^{50}Cr and ^{53}Cr .

Regarding neutron reactions, as in most light isotopes, the probability that it captures an incident neutron is small compared with the one of producing a neutron scattering. This is illustrated in Fig. 1.4, where the capture and total cross sections of $^{50,53}\text{Cr}$ are displayed, showing that the elastic cross section is around 100 to 1000 times larger than the capture cross section. This has important implications in the chromium cross section measurements carried up to date and also in this work.

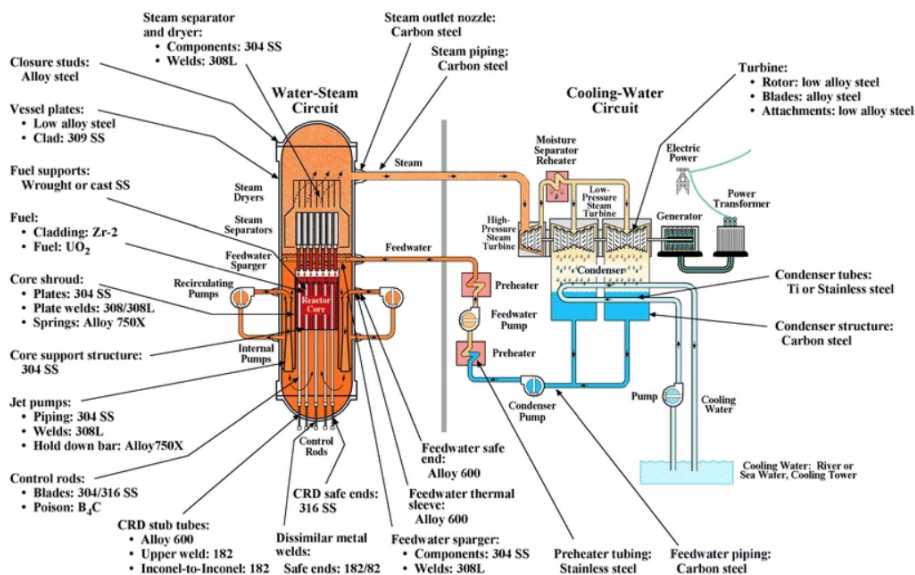


Figure 1.3: Typical design and materials of a Boiling Water Reactor (BWR), illustrating the important presence of stainless steel, and therefore chromium, in the vessel and the rest of the reactor. From Ref. [18].

1.4.1. Previous measurements

Because of the special relevance of chromium as a structural material, it has received the attention of the nuclear data community for a long time. In fact, neutron capture measurements of chromium isotopes have been performed in the main facilities worldwide, as summarized in Table 1.2.

The first high-resolution measurement in the keV region was performed by Stieglitz et al. (1971) [19] at RPI [20] (USA). Capture and transmission measurements were performed with enriched chromium oxide, and resonance parameters up to 356 keV for ^{50}Cr and 176 keV for ^{53}Cr were obtained. As the goal of the measurement was to achieve good statistics even at high neutron energies to identify as many resonances as possible, the samples employed were very thick, thus suffering from important neutron-scattering effects. These occur when the neutrons undergo one or more scatterings before being captured (multiple-scattering), so the measured data has to be corrected to take into account this issue. Another factor that affects the measurements is when scattered neutrons are captured by the detectors, which in this case was a liquid organic scintillator tank, rich in hydrogen and thus very sensitive to neutrons. These aspects were also an issue in the rest of measurements of the 70s: Beer et al. (1975) [21] and Kenny et al. (1977) [22], in which capture and transmission measurements of enriched

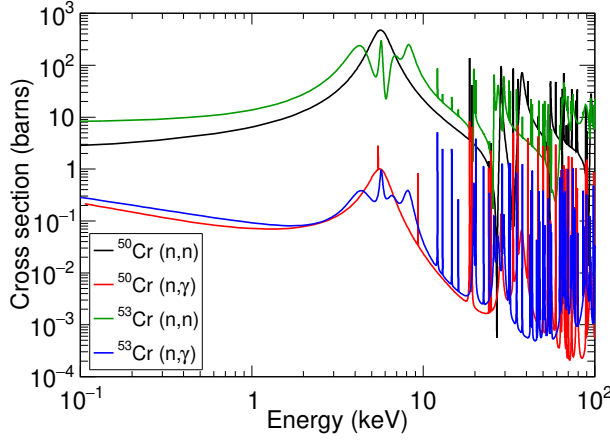


Figure 1.4: $^{50,53}\text{Cr}$ elastic and capture cross sections as featured in the JEFF-3.3 library. The probability that a capture reaction takes place in the main resonances is hundreds of times smaller than the neutron undergoing elastic scattering.

chromium oxide were performed.

It was in Brusegan et al. (1986) [23] in which C_6D_6 capture detectors, characterized by its very low neutron sensitivity (see Section 5.3.1), were employed for the first time. A more recent measurement was performed by Guber et al. (2011) [24] at the Oak Ridge Electron Linear Accelerator ORELA [25] (USA), in which neutron capture and transmission of natural and ^{53}Cr were performed. The most recent evaluations are based on this last measurement. However, it was discovered after the publication of the results that the density of the sample had been determined incorrectly [26]. After correcting the experimental data, the new cross section was in disagreement with the comprehensive results from Stieglitz et al. These discrepancies are still not well understood, and are the reason behind the recent evaluation and experimental efforts, including this work.

Table 1.2: Summary of the $^{50,53}\text{Cr}$ neutron capture cross section measurements performed using the time-of-flight technique. The new measurement proposed in this work aims to improve the previous ones.

Measurement	Stieglitz[19]	Beer[21]	Kenny[22]	Brusegan[23]	Guber[25]	This work
L (m)	27	0.7	40	60	40	185
E_n (keV)	1-200	1-300	1-200	1-200	0.01-600	1-100
Detector	Sc.tank	Sc.tank	C_6F_6	C_6F_6	C_6D_6	C_6D_6
^{50}Cr n(10^{-3}at/b)	8	18	5	7	-	0.6/1.9
^{53}Cr n(10^{-3}at/b)	14	14	8	12	14	1.2/5.9

In Fig. 1.5 some of the most recent evaluated capture cross sections of ^{50}Cr (left panel) and ^{53}Cr (right panel) in the s-wave resonance cluster (between 1 and 10 keV) are

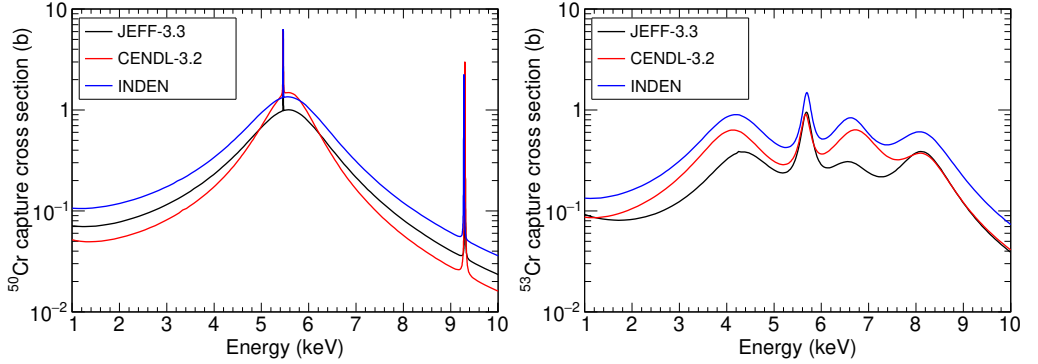


Figure 1.5: Evaluated capture cross sections of ^{50}Cr and ^{53}Cr in the main s-wave resonance energy region.

represented. For ^{50}Cr all major libraries (JEFF-3.3, ENDF/B-VIII.0, JENDL-5 and BROND-3.1) report the same resonance parameters, retrieved from Leal et al. [27]. In that work, a re-evaluation of the four stable chromium isotopes was performed based on Guber et al. and in another ^{50}Cr enriched transmission measurement by Harvey et al. also in ORELA from 90 to 800 keV, which has not been published. The only recent evaluation with different resonance parameters is CENDL-3.2, in which the several measurements from the 70s and 80s mentioned above were considered. The situation is very similar for the ^{53}Cr case: CENDL-3.2 uses the older experimental data and the rest adopt the parameters of Leal et al., based on the Guber et al. and Harvey et al. measurements.

As reported by Trkov [28], when performing k_∞ benchmarks with the JEFF-3.3 and ENDF/B-VIII.0 libraries, significant discrepancies are found between calculations and measurements which seem to be related to inaccuracies in the capture cross sections of chromium isotopes. Indeed, the benchmarks with the largest discrepancies are the ones which contain a relative large amount of Cr, like the KBR experiments for structural materials (HEU-COMP-INTER-005) focused on stainless steel KBR-09(SS) and chromium KBR-15(Cr). The rest of the KBR experiments are also affected because of the SS present in the fuel can and tubing. There was an attempt in BROND-3.1 to correct the ^{53}Cr capture cross section between 1 and 10 keV, with the aim of solving these discrepancies. They suggested to increase this cross section by re-evaluating the data from Guber et al., which are not very well understood as explained above and, in addition, it was misinterpreted as a cross section instead that as an experimental capture yield. Another benchmark very sensitive to chromium is PU-MET-INTER-002, related to SS-reflected assemblies, in which an almost 1% decrease in k_{eff} is observed when using in the calculations the chromium cross sections from BROND-3.1 instead of the

Table 1.3: MACS values at $kT=30$ keV (MACS_{30}) of ^{50}Cr and ^{53}Cr available in the cross section compilations and calculated from the evaluated nuclear data libraries.

MACS_{30} (mb)	^{50}Cr	^{53}Cr
<u>Compilations:</u>		
KADoNiS-1.0 (2014)	36.2(24)	42(10)
Mughabghab (exp) (2018)	40(4)	54(9)
Mughabghab (calc) (2018)	37(3)	36(5)
<u>Evaluations:</u>		
JEFF-3.3 (2017)	38.2	25.9
ENDF/B-VIII.0 (2018)	38.2	25.9
CENDL-3.2 (2020)	37.7	31.5
JENDL-5 (2021)	38.2	26.2
INDEN (2023)	45.0	51.7

previous ENDF/B-VII.1 [29]. The calculations indicate quite clearly that this sensitivity to chromium is dominated by the neutron capture in ^{50}Cr and ^{53}Cr , and that problems and limitations of previous data are propagated to the current chromium evaluations.

The discrepancies between cross sections are even more evident when comparing integral values. This is illustrated in Table 1.3, which summarizes the Maxwellian Averaged Cross Section at 30 keV or MACS_{30} (see Section 2.2.3) corresponding to different evaluations. The values from KADoNiS-v1.0 [30] and Mughabghab [31] data bases are also included. The observed differences range up to 24% for ^{50}Cr and more than a factor of two for ^{53}Cr .

The discrepancies between evaluations and their effects on criticality benchmarks specially sensitive to chromium led to the opening of an entry in the Nuclear Energy Agency (NEA) High Priority Request List (HPRL) [32] dedicated to chromium. It calls for a new measurement of the ^{50}Cr and ^{53}Cr neutron capture cross sections with an accuracy of 8 to 10% in the region between 1 and 100 keV, that would allow to solve the chromium puzzle.

1.4.2. The INDEN project

The latest efforts to improve the chromium evaluated data are mainly led by the INDEN project [33]. INDEN (International Nuclear Data Evaluation Network) is a network coordinated by the IAEA that aims to improve the evaluations and to add new ones regarding light isotopes, structural materials and resonant absorbers, following up the successful CIELO Pilot project [34, 35]. After the evaluation of iron within CIELO [36], new works to re-evaluate the chromium cross sections have been carried out, focusing on a careful analysis of the multiple-scattering correction in previous experiments.

In this new evaluation, instead of starting from the cross sections provided by Stieglitz et al. and Guber et al., since the thin sample approximation is not valid (see Section 2.2.2), the analysis was made on the related capture yield instead. A simultaneous fit of the capture yield and transmission coefficients was performed with the code SAMMY [37], including the finite-size of the samples and paying special attention to the mentioned multiple-scattering corrections. In this process, they concluded that the thickness reported for the ^{53}Cr -enriched sample of Guber et al. was not correct, and thus neither was the decreasing on the capture cross section reported in the latest evaluations. For this reason, a new resonance analysis was performed using the data of Stieglitz et al. and those of the natural samples of Guber et al. All the details can be found in [26].

In the last version available of the INDEN evaluation from August 2023, the capture cross section of ^{50}Cr below 10 keV and ^{53}Cr in the full resonance energy region are updated according to the analysis described above. For ^{50}Cr at energies higher than 10 keV, the cross section adopts the parameters from ENDF/B-VIII.0 (i.e. the ones from Leal et al. [27]). The resulting cross section is shown in Fig. 1.5 together with the other evaluations, illustrating the general increase of the capture cross sections proposed in INDEN. Regarding the MACS₃₀ (see Table 1.3), it is noticeable the major increase of 22% for ^{50}Cr and 88% for ^{53}Cr with respect to the previous libraries.

The new chromium evaluation was tested on several criticality benchmarks of chromium and SS, together with new INDEN evaluations of iron, silicon and oxygen, improving significantly the performance compared to previous evaluations. However, the proposed enhancement in the chromium cross section must be confirmed by new neutron capture data because, overall, the old ones suffered from high neutron-scattering effects and the most recent from Guber et al. is not considered as reliable.

1.5. Objectives and strategy

As exposed earlier, the main issues that faced previous chromium measurements were the large neutron-scattering background and multiple-scattering effects in the yield related to the large scattering-to-capture ratio in chromium and the use of thick samples. The consequence is, in principle, an apparent enhancement of the capture cross section. In this work, the following aspects have been considered to reduce those undesired effects:

1. **Sample thickness.** The use of very thick samples enhances the probability that a neutron undergoes scattering interactions inside the sample before being captured, affecting the yield and requiring corrections that are far from easy to estimate. The solution is as "simple" as to use thinner samples, with the disadvantage that the irradiation times need to be increased to achieve the same amount of statistics.

2. **Sample characterization.** Previous measurements, at least that of Guber et al., suffered from uncertainties in the thickness of the sample, which affects directly the normalization of the capture yield. This is solved by using highly enriched chromium and ensuring an accurate characterization of the sample.
3. **Neutron sensitivity.** The chromium samples scatter many more neutrons than they capture, hence neutron background was an issue in many of the previous experiments. The solution is to employ low neutron sensitivity detectors and to minimize the amount of material in the vicinity of the samples and the detectors.

In order to overcome these issues, the solutions mentioned above have been implemented and two experiments have been designed and performed: a $^{50,53}\text{Cr}$ time-of-flight measurement at the n_TOF facility of CERN (Geneva, Switzerland) and a ^{50}Cr neutron activation measurement at the HiSPANoS facility of CNA (Seville, Spain). A detailed description of the experiments, data analysis and results is presented in this work.

The time-of-flight measurement aims at providing new accurate capture data that will be compared with the previous measurements and evaluations. The measurement is characterized by the use of very thin samples for the region that contributes the most to the neutron scattering (1-10 keV) to minimize the neutron-scattering effects, and thick samples for the rest of the range of interest (10-100 keV), both made of highly-enriched material. As displayed in Table 1.2, the largest thickness used in this work is thinner than all previous samples. The measurement took place in the EAR1 experimental area of n_TOF, which fulfills the requirements exposed above regarding neutron sensitivity (more details can be found in Section 5.2).

When ^{50}Cr captures a neutron, the product nucleus ^{51}Cr is unstable and thus opens the possibility of performing a neutron activation measurement. In this work, for the first time the MACS of ^{50}Cr at $kT = 30$ keV has been directly measured by the activation technique. This serves as a good complement to the time-of-flight measurement, since the MACS at that temperature is strongly influenced by the resonance region of interest.

The results obtained through both measurements satisfy the request made by the NEA and hopefully will help to understand the current differences between evaluated libraries. Also it is the first time that the cross section increase proposed by INDEN is tested experimentally.

After an introduction to neutron capture cross sections in Chapter 2, the activation measurement is described in Chapters 3 and 4, and the time-of-flight measurement in Chapters 5 through 7. Last, the results are further discussed and the conclusions presented in Chapter 8.

2 | Neutron capture cross sections

In this chapter, the key concept of the Compound Nucleus in the context of nuclear reactions is presented in detail, as it is key for the understanding and the analysis of the resonant structures observed in cross sections. Then, an overview is made about the facilities and experimental techniques more widely used for measuring neutron capture, focusing on the relevant topics regarding the capture measurement of $^{50,53}\text{Cr}$ presented in this work.

2.1. Neutron-induced reactions theory

When any nucleus is bombarded by neutrons, a number of different reaction can take place: capture (n,γ), fission (n,f), elastic (n,n) and inelastic (n,n'), production of light charged particles (n,p), (n,α),... The probability that one reaction takes place as a function of the neutron energy is related to the *cross section*, expressed in units of barn ($1\text{ b} = 10^{-24}\text{ cm}^2$), which is approximately the cross-sectional area of an uranium nucleus. The total cross section would be the sum of all the partial cross sections:

$$\sigma_{tot} = \sigma_{\gamma} + \sigma_f + \sigma_n + \dots \quad (2.1)$$

The reaction cross sections are highly dependant on the nuclear structure of the target nucleus and the energy of the neutron. This is reflected in the significant change on the cross section when comparing different nuclei, or even different reactions channels in the same nucleus. In addition, cross sections usually present narrow structures called *resonances* for some energy values that can raise their value many orders of magnitude between the valley and the peak. For example, as is shown in Fig. 2.1, between the main capture resonances of ^{50}Cr and ^{197}Au there is a distance of 3 decades in energy and more than 4 orders of magnitude in the peak cross section value.

The cross sections show some general trends as a function of the neutron energy:

- **$1/v$ region.** It covers everything below the energy of the first resonance, which depends on the nucleus. In this region, the cross section is inversely proportional

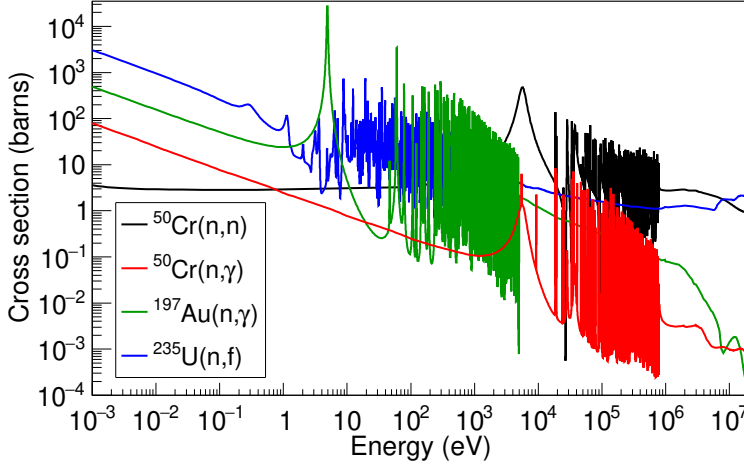


Figure 2.1: Some examples of neutron reaction cross sections for several nuclei.

to the square root of the energy, because it depends on the time spent by the neutron in the vicinity of the nucleus, so $\sigma \propto 1/\sqrt{E_n} \propto 1/v$.

- **Resolved Resonances Region (RRR).** It is characterized by the presence of resonances, reflecting the formation of a quasi-stable system with an excitation energy corresponding to an unbound level of what is known as the Compound Nucleus (CN). This is the relevant region for our work, so more details about the theoretical description of the CN are found in the next section.
- **Unresolved Resonances Region (URR).** In this region, the resonances are so close to each other that they begin to overlap, resulting in a smooth cross section.
- **High Energy region.** When the distance between resonances is much smaller than their width it can be considered a continuum. In this region, threshold channels like (n,xn) or (n,p) open up (see Section 2.2.1).

Depending on the time scale in which they take place, neutron interactions can be divided into two categories. If the interaction occurs in a very short time ($\sim 10^{-22}$ s), then is considered a *Direct reaction* in which the neutron only interacts with one nucleon. On the other hand, if the interaction is long enough, there is the possibility that a relatively long-lived system is created. As already mentioned, this is known as *Compound Nucleus reactions*, and is the reason behind the resonant structures of cross sections, as it is described in detail in the following.

2.1.1. The Compound Nucleus

The idea of Compound Nucleus (CN) was proposed by N. Bohr in 1936 [38]. It was observed that the interaction time between an incident fast neutron and the nucleus, of around 10^{-15} s, was much longer than the time that the neutron needed to transverse it, typically around 10^{-20} s. That means that some kind of compound system of relatively stability must form, which is the main principle of the CN model.

This model divides the neutron interaction in two steps:

1. First, the incident neutron and the target nucleus (AZ) form the CN ($^{A+1}Z^*$) with an excitation energy corresponding to that brought by the neutron to the system plus the neutron separation energy S_n of the ^{A+1}Z nucleus. This energy is distributed among all the nucleons, with the CN featuring energy levels, similar to a conventional nuclear system. The probability of forming the CN is therefore very small except if the energy of the system ($S_n + E_n$) matches the energy of one of the levels, being this the origin of the *resonances* observed in the cross section.
2. The second step is the decay of the CN by the emission of particles like γ -rays (capture process), a neutron leaving the nucleus in the ground (elastic scattering) or an excited (inelastic scattering) state, or even light charged particles like protons, tritons or alphas. With the CN model is also possible to describe the fission process.[39]. The de-excitation through one or another path is determined by probabilistic processes, and is independent of the CN formation mechanism.

A schematic view of the process regarding a neutron capture CN reaction is displayed in Fig. 2.2. The total excitation energy of the system E^* can be approximately expressed as

$$E^* \cong S_n + \frac{A}{A+1} E_n, \quad (2.2)$$

where S_n is the neutron separation energy, A is the atomic mass and E_n the neutron energy. The observed resonances in the cross sections correspond to the energy levels above S_n . Each resonant level decays with a half-life τ defined by the quantum properties of the system and related to the resonance width Γ by the uncertainty principle as $\Gamma = \hbar/\tau$. The total width of the state is the sum of the partial widths of all the possible de-excitation processes that the CN can undergo:

$$\Gamma = \Gamma_\gamma + \Gamma_n + \Gamma_f + \dots \quad (2.3)$$

In the case of capture, the de-excitation takes place through the emission of γ -rays according to the energy levels of the product nucleus, via the so-called *capture cascade*.

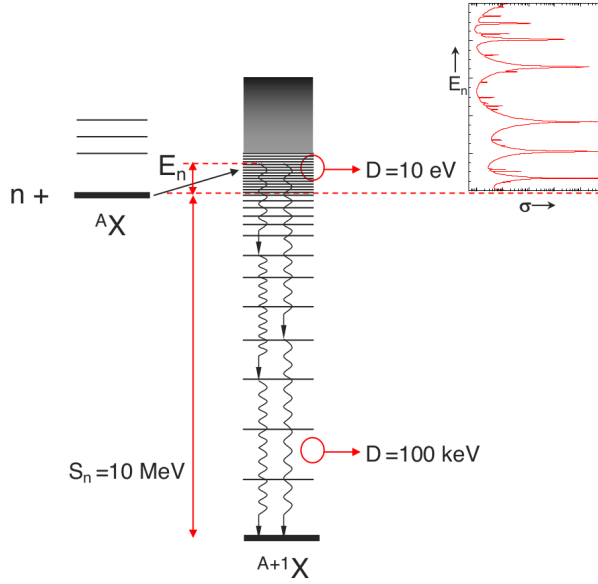


Figure 2.2: Schematic view of the formation of the Compound Nucleus after a neutron capture of energy E_n . The presence of resonances in the cross section is due to the energy levels above the neutron separation energy. From Ref. [40].

Conceptually, the Compound Nucleus model has had a great success through the years. The mathematical formalism to describe the model is known as R-matrix theory, and its main aspects are described in the following section.

2.1.2. R-matrix theory

The R-matrix formalism was proposed for the first time by Wigner and Eisenbud in 1946-1947 [41, 42]. In this Section we present an overview of the theory, applied to described capture resonances. A detailed mathematical development of the R-matrix formalism can be found in the extensive review of the theory by Lane and Thomas [43] and more recently by Fröhner [44].

The basic principle of the R-matrix is to split the space in which a reaction takes place into two regions, divided by the so-called channel radius a_c :

1. An external region where the nuclear forces are negligible, given by $r > a_c$. In this region, the system can be described by the wave functions of two free particles, either the incident (a neutron and the target nucleus in our case) or the reaction products (an excited nucleus and a γ -ray, two fission fragments, etc.).
2. An internal region $r < a_c$ where the nuclear forces come into play. Because the

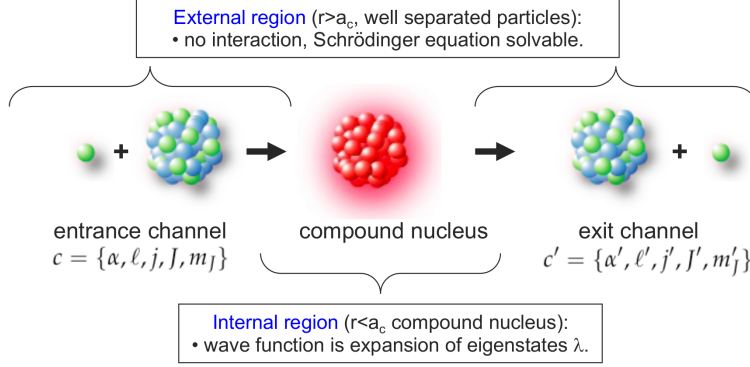


Figure 2.3: Schematic of the main principle of the R-matrix, in which the nuclear reaction space is divided into two regions (see text for details). From Ref. [45].

interactions are very complex, its exact form is unknown, but the internal wave function can be described as a lineal combination of eigenfunctions. These are defined by the non-relativistic Schrödinger equation of the in- and outgoing waves, matching them both at the boundary condition $r = a_c$.

In this formalism one talks about *reaction channels*, each one fully specified by

- α , which describes each particle that takes part of the reaction by its mass, charge, spin and any relevant quantum number,
- ℓ , the total orbital momentum,
- s , the total channel spin, defined as the vector sum of the spin of the incident particle and the nucleus $\vec{s} = \vec{i} + \vec{I}$, satisfying the relation $|i - I| < s < i + I$,
- J , the total angular momentum $\vec{J} = \vec{\ell} + \vec{s}$, satisfying $|\ell - s| < J < \ell + s$.

The non-relativistic Schrödinger equation, considering neutral particles and with a stationary ingoing plane wave plus a spherical outgoing wave as boundary conditions, was solved by Blatt and Biedenharn in 1952 [46], obtaining the following result:

$$\sigma_{cc'} = \pi \lambda_c^2 g_J |\delta_{cc'} - U_{cc'}|^2, \quad (2.4)$$

where $\sigma_{cc'}$ is the cross section of the incident channel c (defined as $c \equiv \{\alpha s \ell J\}$) and the exit channel c' integrated over all angles, the de Broglie wave length $\lambda_c = \hbar / (2\pi \mu_c v_{rel})$, with μ_c the reduced mass and v_{rel} the relative speed of the particles, g_J is the spin factor:

$$g_J = \frac{2J + 1}{(2i + 1)(2I + 1)}, \quad (2.5)$$

$\delta_{cc'}$ is the Kronecker delta and $U_{cc'}$ are the elements of the so-called collision matrix $\mathbf{U}$. This result indicates that, for $c \neq c'$, the cross section is proportional to the transition probability from channel c to channel c' , $|U_{cc'}|^2$, and to the probability that the total momentum is J . Since $\mathbf{U}$ is unitary (the sum of all probabilities must be the unity), the total cross section for the entrance channel c can be expressed as

$$\sigma_c \equiv \sum_{c'} \sigma_{cc'} = 2\pi\lambda_c^2 g_J (1 - \text{Re } U_{cc}). \quad (2.6)$$

The interactions taking place inside the nuclei are introduced in the theory by the channel matrix $\mathbf{R}$. Its elements correspond to the resonances, and take the following form:

$$R_{cc'} = \sum_{\lambda} \frac{\gamma_{\lambda c} \gamma_{\lambda c'}}{E_{\lambda} - E}, \quad (2.7)$$

with $\gamma_{\lambda c}$ the probability amplitude for the formation of compound levels λ through entrance channel c (or, equivalently, for the decay via channel c') and E_{λ} the energy of the resonance. The resonance parameters E_{λ} and $\gamma_{\lambda c}$ depend on the unknown nuclear interaction, so they are introduced in the theory as fit parameters adjustable from experimental data. Henceforth, Greek subscripts refers to compound levels and Roman subscripts to reaction channels.

Instead of $\mathbf{R}$, the collision matrix also can be expressed using the level matrix $\mathbf{A}$:

$$U_{cc'} = e^{-i(\varphi_c + \varphi_{c'})} \left(\delta_{cc'} + i \sum_{\lambda, \mu} \Gamma_{\lambda c}^{1/2} A_{\lambda \mu} \Gamma_{\mu c'}^{1/2} \right), \quad (2.8)$$

$$\Gamma_{\lambda c}^{1/2} \equiv \gamma_{\lambda c} \sqrt{2P_c}, \quad (2.9)$$

$$(\mathbf{A}^{-1})_{\lambda \mu} = (E_{\lambda} - E) \delta_{\lambda \mu} - \sum_c \gamma_{\lambda c} L_c^0 \gamma_{\mu c}. \quad (2.10)$$

The cross sections are expressed more often in terms of the partial widths $\Gamma_{\lambda c}$ defined as $\Gamma_{\lambda} \equiv \sum_c \Gamma_{\lambda c}$, instead of the probability amplitudes. The rest of the quantities depend only on the known in- and outgoing wave functions at the channel radius a_c : φ_c are the hard-sphere phases, L_c the logarithmic derivatives and $P_c \equiv \text{Im } L_c$ the centrifugal-barrier penetrabilities.

For neutral incident particles, the outgoing radial wave functions are proportional to the spherical Hankel functions of the first kind, which satisfy the following recursive

relations:

$$L_0 = ik_c a_c = iP_0, \quad L_\ell = -\ell - \frac{(k_c a_c)^2}{L_{\ell-1} - \ell}, \quad (2.11)$$

$$\varphi_0 = k_c a_c, \quad \varphi_\ell = \varphi_{\ell-1} + \arg(\ell - L_{\ell-1}), \quad (2.12)$$

where $k_c = 1/\lambda_c$ is the wave number. The channel radius is usually adopted as $a_c = (1.23A^{1/3} + 0.80)$ fm for neutron channels, with A the atomic mass.

2.1.3. Approximations of the R-matrix

In the previous section we have presented the main elements of the R-matrix theory. To apply this theory, one must invert the level matrix $\mathbf{A}$, but because of its complexity it is practically impossible to do so in any real scenario. For this reason, several approximations of the theory have been developed to simplify the problem.

One example is to consider only one level, which is the basic principle of the Single-level Breit-Wigner approximation (SLBW). In that case, Eq. 2.10 becomes

$$(\mathbf{A}^{-1})_{\lambda\mu} \rightarrow E_0 - E - \sum_c L_c^0 \gamma_c^2 \equiv E_0 + \Delta - E - \frac{i}{2} \Gamma, \quad (2.13)$$

where $\Delta = E_0 - E_r$ is the level shift or difference between the level and the resonance energies, and $\Gamma = \sum_c \Gamma_c$ the total width. Both quantities are real, energy dependent and well-known.

Another option is the so-called Multi-level Breit-Wigner approximation (MLBW), in which only non-correlated levels are considered by neglecting all the off-diagonal terms of $\mathbf{A}^{-1}$. In that case,

$$(\mathbf{A}^{-1})_{\lambda\mu} = \left(E_\lambda - E - \sum_c L_c^0 \gamma_{\lambda c}^2 \right) \delta_{\lambda\mu} \equiv \left(E_\lambda + \Delta_\lambda - E - \frac{i}{2} \Gamma_\lambda \right) \delta_{\lambda\mu}. \quad (2.14)$$

The most accurate approximation is the Reich-Moore approximation (RM) [47], in which one assumes that the photon channels contributions to the off-diagonal elements can be neglected. Eq. 2.10 becomes then

$$(\mathbf{A}^{-1})_{\lambda\mu} = \left(E_\lambda + \Delta_{\lambda\gamma} - E - \frac{i}{2} \Gamma_{\lambda\gamma} \right) \delta_{\lambda\mu} - \sum_{c \notin \gamma} \gamma_{\lambda c} L_c^0 \gamma_{\mu c}. \quad (2.15)$$

In this expression, $\Delta_{\lambda\gamma}$ from photon channels is usually absorbed in a constant E_λ , and $\Gamma_{\lambda\gamma}$ is often taken as a constant. This approximation is the preferred option when evaluating neutron resonances with analysis codes like SAMMY [37] (see Chapter 7).

2.1.4. Characteristics of the resonances

As mentioned before, the cross section resonances are defined by their energy E_0 , the partial widths Γ_c and the spin J , that have to be considered in the R-matrix formalism. These parameters are determined by performing reaction cross section measurements (see Section 2.2).

The quantity obtained from a typical measurement is the number of reactions per incident neutron, i.e. the reaction yield $Y_r(E_n)$ (see Eq. 2.22), which features the resonances. From the analysis of the yield one can extract information such as the resonance parameters mentioned above. The simplest value that can be extracted from an observed resonance is its area. Integrating the reaction cross section, and considering the thin sample approximation (see Section 2.2.2), the resonance area is

$$A_r = \int \sigma_r(E_n) dE_n = 2\pi^2 n \bar{\lambda}_0 g_J \frac{\Gamma_r \Gamma_n}{\Gamma}, \quad (2.16)$$

where $\bar{\lambda}_0$ is the reduced wave length of a neutron with energy E_0 , n the atomic thickness of the sample and Γ_r and Γ_n are the reaction channel and neutron scattering partial width respectively, with $\Gamma = \Gamma_r + \Gamma_n$. According to the previous expression, the resonance area is proportional to the so-called *reaction kernel* K_r , defined as:

$$K_r = g_J \frac{\Gamma_r \Gamma_n}{\Gamma}. \quad (2.17)$$

The definition of the reaction kernel implies that, if one of the two widths dominates, then the kernel is proportional to the other one. That is:

$$\begin{aligned} \Gamma_r \ll \Gamma_n &\rightarrow K_r \propto g_J \Gamma_r \\ \Gamma_r \gg \Gamma_n &\rightarrow K_r \propto g_J \Gamma_n \end{aligned} \quad (2.18)$$

If the measurement allows it, not only the area but the actual resonance shape can be studied through a fitting program like SAMMY (used in this work) or REFIT [48] that returns the resonance parameters.

In addition to the R-matrix expressions described above, these resonance analysis codes need to consider the experimental factors that affect the measured yield and its relation with the cross section. The main ones are the Doppler broadening effect due to the temperature of the nucleus, multiple-scattering and self-shielding because of the finite size of the samples and the Resolution Function (RF) of the facility. These effects are presented and discussed in detail along this work.

2.2. Capture cross section measurements

In this section we present an overview of the neutron beam production, the main neutron facilities worldwide and some experimental techniques for measuring neutron reaction cross sections.

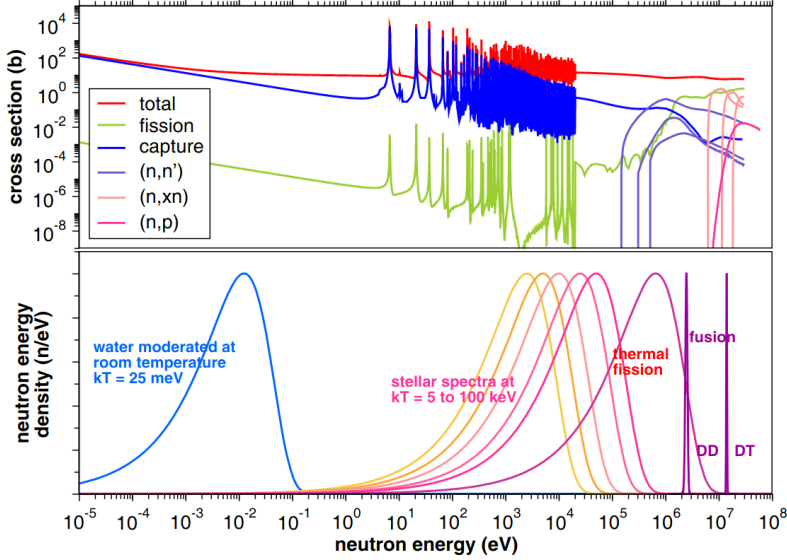


Figure 2.4: Schematic showing the typical cross sections of a heavy nucleus for several neutron reactions (top panel), along with some examples of the most common neutron beams produced for different applications. From Ref. [49].

2.2.1. Neutron facilities

Neutrons are of interest in many research fields and are used in a variety of applications. In each particular case, the energy of the neutrons involved is different, ranging from meV to tens or hundreds of MeV. Therefore, facilities are needed to produce beams from cold neutrons ($E_n < 25$ meV) to high energy neutrons ($E_n > 20$ MeV), including thermal ($E_n = 25$ meV - 1 eV), epithermal ($E_n = 1$ eV - 100s keV) and fast neutrons ($E_n = 100$ s keV - 20 MeV). The behaviour of the neutron cross sections in such a large energy span is illustrated in Fig. 2.4, where the $1/v$, RRR and URR regions, and threshold reactions are clearly shown.

The characteristics in terms of energy and intensity of the neutron beams produced to study nuclear reactions depend on the production method. These beams can be mono-energetic, a Maxwell-Boltzmann distribution centered on some temperature or a white spectrum if it covers several orders of magnitude. The energy scale of the most common



Figure 2.5: View of the Budapest Research Reactor (BRR). By courtesy of G. Torvund.

neutron distributions is illustrated in Fig. 2.4.

According to the neutron production technique, the facilities can be classified as follows:

- **Nuclear Research Reactors.** A very intense neutron flux is produced by fission reactions inside a reactor, where cold and thermal neutron beams are extracted to perform experiments like neutron imaging, neutron scattering or medical isotopes production, or the samples are introduced inside the reactor core for irradiation. Fast neutrons are also produced in some reactors [50]. A couple of examples are the ILL [51] (Grenoble, France), with the most intense neutron flux in the world ($1.5 \cdot 10^{15}$ n/cm²/s), and the Budapest Research Reactor (BRR) [52] in Hungary (shown in Fig. 2.5).
- **Low energy ion accelerators.** The neutrons are produced by means of nuclear reactions using light ion beams, mainly protons and deuterons, produced by low energy accelerators (<15 MV). These reactions can be divided in two groups. First, fusion reactions like $^2\text{H}(d,n)$, $^3\text{H}(p,n)$ and $^3\text{H}(d,n)$, resulting in mono-energetic neutron beams. Second, reactions like $^7\text{Li}(p/d,n)$ and $^9\text{Be}(p/d,n)$, which produce broad fast or epithermal beams when the ion energy is just above the reaction threshold. Some facilities of this kind are JRC MONNET [53] (Geel, Belgium), LiLiT [54] (Soreq NRC, Israel) or CNA HiSPANoS (Seville, Spain), described in detail in Chapter 3.
- **High energy electron accelerators.** In these facilities, electrons are accelerated using a linear accelerator (LINAC) up to hundreds of MeV, sending the beam towards a heavy target. When the electrons are slowed down inside the target, the Bremsstrahlung radiation produces neutrons by photo-fission or (γ,n) reactions.

If the neutrons are moderated, the outcome becomes a white spectrum. These reactions are not very efficient regarding the number of neutrons per pulse, but it is compensated by operating at a very high frequency (hundreds of kHz). Some examples are JRC GELINA [55] (Geel, Belgium) or nELBE at HZDR [56] (Dresden, Germany).

- **High energy ion accelerators.** Very energetic ion pulses (between 100s MeV and GeV) collide with heavy targets made of materials like Pb, W or Ta, producing neutrons via spallation [57]. After being moderated, white spectra in a wide energy range are obtained, as well as pretty intense fluxes. The disadvantage is that they are complex facilities that need large, resource-intensive accelerators. A few spallation facilities are LANSCE [58] (Los Alamos, USA), JSNS at J-PARC [59] (Japan), Back-n at CSNS [60] (China) or n_TOF at CERN [61–63], (Geneva, Switzerland), described in Chapter 5.
- **High power laser.** Nowadays, the possibility of producing neutrons using high power (100s TW and PW) ultra-fast (fs) lasers is under investigation [64, 65]. This technology would allow to build compact facilities with very high neutrons-per-pulse fluxes to perform time-of-flight experiments.

2.2.2. Cross section measurements: time-of-flight technique

With the *time-of-flight* technique it is possible to measure a neutron reaction cross section over a wide range as a function of the neutron energy. In these experiments, the measuring station is placed at a distance L from the neutron source, and the kinetic energy of the neutrons E_n is determined by the time span t between their production and the detection of the subsequent reaction by means of the non-relativistic relation:

$$E_n = \frac{1}{2} m_n v^2 = \frac{1}{2} m_n \frac{L^2}{t^2}, \quad (2.19)$$

where m_n is the neutron mass and v its speed. A time-of-flight facility must produce a pulsed neutron beam, so the neutron production time is well defined. Usually, a very intense γ -ray beam precedes the neutron one, so it can be used as the initial time reference. A schematic of a typical time-of-flight measurement is displayed in Fig. 2.6.

According to Eq. 2.19, in a facility of flight path L the relative energy resolution is defined as

$$\frac{\Delta E}{E} = 2 \sqrt{\left(\frac{\Delta L}{L}\right)^2 + \left(\frac{\Delta t}{t}\right)^2}. \quad (2.20)$$

This relation shows that the resolution improves for longer flight paths. Of course, the

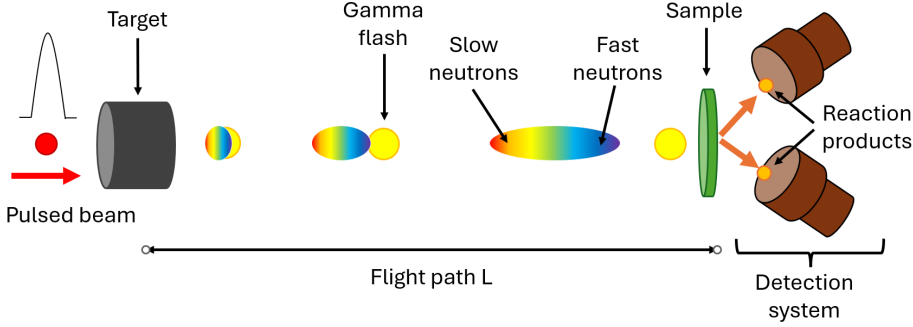


Figure 2.6: Schematic showing the main principles of the time-of-flight technique. If the flight path is long enough, the γ -flash arrives first to the detector system, and then the neutrons according to their speed, producing the desired reaction in the sample.

intensity of the neutron flux decreases with the distance, so depending on the kind of measurement a compromise in the flight path has to be found. A good example is the n_TOF facility, which offers an experimental area with a long flight path (185 m) with a very good energy resolution, and a second area with a short flight (19 m) path featuring a lower resolution but a very intense neutron flux (see Section 5.1).

The other factor that affects the resolution is the time determination. The detectors feature a finite time resolution, that marks the smallest time interval that can be distinguished by each detection system. In addition, if the neutrons are moderated along the flight path, then the relation between neutron energy and time-of-flight is not univocal. This is related to the Resolution Function (RF) of the facility, and how it affects to the measurements performed at n_TOF is explained in Section 5.2.3.

The quantity obtained in a time-of-flight measurement is the *reaction yield*, defined as the probability of a neutron undergoing a given reaction as a function of its energy. In general, the yield can be expressed as

$$Y_r(E_n) = \frac{C_r(E_n)}{\Phi(E_n)}, \quad (2.21)$$

where $C_r(E_n)$ is the number reactions occurred and $\Phi(E_n)$ the total number of neutrons traversing the sample. The reaction yield can be related to the cross section by considering the probability of the neutron to interact with the sample and the probability of that reaction being the one of interest:

$$Y_r(E_n) = (1 - e^{-n\sigma_t}) \frac{\sigma_r}{\sigma_t}, \quad (2.22)$$

where n is the atomic thickness of the sample in atoms/barn, σ_r the reaction cross section and σ_t the total cross section, both expressed in barns. Because of the finite size of the sample, a correction factor F has to be included in Eq. 2.22, that takes into account neutrons that scatter once or more times inside the sample before undergoing the specific reaction.

If the sample is so thin that the relation $n\sigma \ll 1$ is true, then the previous expression can be approximated as $Y_r(E_n) \approx n\sigma_r$, so the reaction yield becomes proportional to the atomic thickness and the cross section; this is known as the *thin sample* approximation. On the contrary, when the thickness is sizable, the multiple-scattering effects become more significant and their impact on the capture yield has to be considered.

In a neutron capture cross section measurement like the one described in this work, what is measured is the γ -rays produced after the neutron capture. The origin of these γ -rays is the de-excitation of the Compound Nucleus. This is a random process, which means that the cascades can follow many different paths through many different nuclear levels, specially in heavy nuclei. As a consequence, the energy and multiplicity of the emitted γ -rays is variable and thus a capture cascade is difficult to identify and be measured with accurately known detection efficiency. Several measuring techniques have been developed to overcome this issue:

- **Prompt Gamma Activation Analysis (PGAA)**. This technique is based on the detection of the characteristics prompt γ -rays emitted after a neutron capture [66]. If the energy and the intensity of each γ -ray is well known, since is a non-destructive technique is often used for material analysis. It requires of detectors with good energy resolution to distinguish the different γ -rays emitted in the complex cascade de-excitation scheme, like HPGe detectors. It can also be used for the determination of thermal capture cross sections [67].
- **Total Energy Detection (TED)** [68, 69]. This technique is based on the detection of only one γ -ray of each capture cascade by using detectors with very low efficiency. Another condition necessary to apply this technique is that the efficiency must be proportional to the energy of the γ -ray. It can be proved that, when these conditions are true, the efficiency of detecting a cascade is independent of the de-excitation path. Since the second condition is not fulfilled by the majority of detecting systems, the response of the detector must be weighted by a special function, using the so-called *Pulse Height Weighting Technique* (PHWT) [70]. The $^{50,53}\text{Cr}$ (n, γ) cross sections measured at n_TOF and presented in this work have been carried out using this technique, thus further details are given in Section 6.3.

- **Total Absorption technique.** This method is based on the detection of the full cascade, so a detection system with 100% γ -ray detection efficiency is needed. A typical configuration of a Total Absorption set-up is an array of detectors covering a 4π solid angle around the sample. A couple of examples are the Total Absorption Calorimeter (TAC) of n_TOF [71], made of 40 BaF₂ detectors, or the Detector for Advanced Neutron Capture Experiments (DANCE) at the Los Alamos National Laboratory (LANL), with 162 BaF₂ detectors [72].

2.2.3. Cross section measurements: activation technique

Integral cross sections measurements are based on the idea that, after irradiating the sample with a neutron flux, by measuring the number of produced nuclei on the reaction, the corresponding cross section can be extracted as:

$$\sigma_r = \frac{N_X}{n_{\text{at}}\phi}, \quad (2.23)$$

with N_X is the number of nuclei produced by the reaction, n_{at} the atomic thickness and ϕ the total number of neutrons. The result can be related with an averaged cross section, defined as

$$\langle\sigma_r\rangle_\Phi = \frac{\int \Phi(E_n)\sigma_r(E_n)dE_n}{\int \Phi(E_n)dE_n}, \quad (2.24)$$

where $\Phi(E_n)$ is the neutron distribution used in the irradiation and $\sigma_r(E_n)$ the point-wise cross section.

There are some atom counting techniques to determine N_X with a great precision, like mass spectroscopy (AMS) or radio-chemical techniques. On the other hand, if the product nucleus is unstable and its decay allows it, the number of reactions occurred can be determined by measuring its decay. This technique is known as *neutron activation*.

In the context of neutron cross sections, a very relevant quantity, specially in the s-process of stellar nucleosynthesis [73], is the Maxwellian Averaged Cross Section at a temperature of kT (MACS _{kT}), defined as

$$\text{MACS}_{kT} = \frac{\langle\sigma v\rangle}{v_T} = \frac{2}{\sqrt{\pi}} \frac{\int_0^\infty E_n e^{-E_n/kT} \sigma(E_n) dE_n}{\int_0^\infty E_n e^{-E_n/kT} dE_n}, \quad (2.25)$$

where $\langle\sigma v\rangle$ is the neutron capture reaction rate, v_T is the mean thermal velocity, E_n is the total kinetic energy in the center-of-mass system, k is the Boltzmann constant, T is the effective temperature and $\sigma(E_n)$ is the point-wise neutron capture cross section.

The standard value used as reference is the MACS at $kT = 30$ keV, corresponding to the

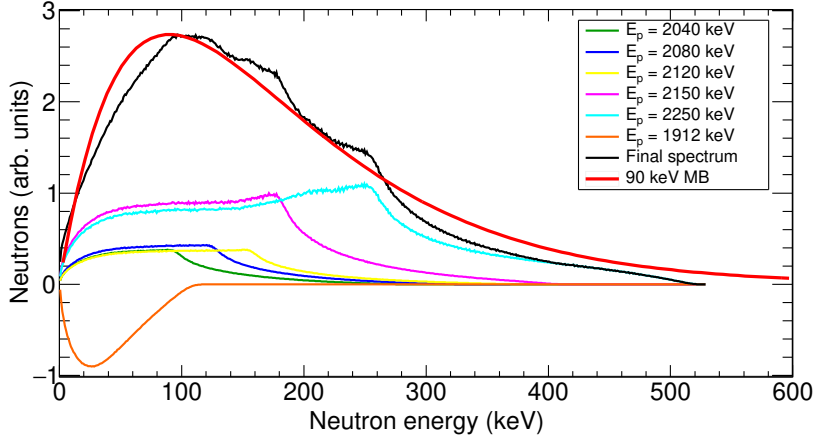


Figure 2.7: With a linear combination of several neutron spectra produced by the ${}^7\text{Li}(p,n)$ with proton beams of different energies, the resulting spectrum is a Maxwellian distribution at $kT = 90$ keV. The spectra represented here were simulated with PINO [82].

helium burning phase in red giant stars [74], although the temperatures of interest range from 5 keV up to 100 keV. This is very interesting because this maxwellian neutron energy spectrum can be produced in a facility using three reactions: ${}^7\text{Li}(p,n)$ with 1912 keV protons for a $kT \sim 25$ keV spectrum [75, 76], ${}^{18}\text{O}(p,n)$ with 2582 keV protons for $kT \sim 5$ keV [77] and ${}^3\text{H}(p,n)$ with 1099 keV protons for a $kT \sim 52$ keV one [78]. For the rest of temperatures, the measured MACS_{kT} has to be extrapolated from one of the mentioned temperatures (increasing the uncertainty when the temperatures are more different), or directly calculated using the point-wise cross section previously determined by time-of-flight using Eq. 2.25.

More recently, a new technique to produce spectra at different temperatures has been proposed by Reifarth et al. [79], in which several spectra are produced using the ${}^7\text{Li}(p,n)$ reaction with different proton energies. None of them is a Maxwellian distribution, but with the adequate linear combination of these spectra the equivalent total neutron distribution is a Maxwellian at a temperature of $kT = 90$ keV, as can be seen in Fig. 2.7. The feasibility of implementing this technique for activation measurements is being investigated at the moment [80, 81].

Even though it is not as precise as a time-of-flight measurement, integral quantities like the MACS_{kT} is a direct and relatively simple way to obtain valuable information about a reaction or to validate a certain evaluation, because it is just a weighted integral of the differential cross section, as seen in Eqs. 2.24 and 2.25. The MACS at $kT = 30$ keV of ${}^{50}\text{Cr}$ has been measured by means of the activation technique at the HiSPANoS neutron facility, thus many more details will be given in Chapters 3 and 4.

Part II

^{50}Cr activation measurement at HiSPANoS

3 | Activation measurement at HiSPANoS

As already introduced in the previous chapters, to solve the current discrepancies regarding the $^{50,53}\text{Cr}$ neutron capture cross section, we have designed and performed two experiments: a ^{50}Cr neutron activation measurement and a time-of-flight measurement of both isotopes. Although with an activation measurement the result is an averaged cross section (instead of the point-wise cross section obtained by time-of-flight), is a fundamental quantity to check the validity of previous experiments and evaluations.

As will be discussed throughout this work, the Maxwellian Averaged Cross Section or MACS of an isotope is a direct and relatively simple measurement, and can be really useful for the nuclear data evaluators and to compare different evaluations. Since ^{51}Cr is an unstable isotope with suitable decay characteristics, ^{50}Cr is a perfect candidate to perform a neutron activation measurement.

In this Chapter we will describe the HiSPANoS facility and the experimental set-up employed for the activation measurement performed at this facility.

3.1. Centro Nacional de Aceleradores (CNA) research center

The Centro Nacional de Aceleradores (CNA) [83] is one of the most important multipurpose-nuclear research facilities in Spain. Located in Seville, is a joint research center supported by Universidad de Sevilla (US), the regional government of Andalusia and the National Council of Scientific Research (CSIC). It covers a large number of research fields and it is open to external users. It has been recognized by the Spanish government as a Singular Scientific-Technical Facility (ICTS).

Six different facilities and laboratories are available at CNA, which are the following:

- **3 MV Tandem accelerator** [84]. The first ion beam accelerator built in Spain, it is able to accelerate many different stable ions with its 3 MV terminal. There are eight experimental beam-lines available, each one focused on a specific application. HiSPANoS is installed in this accelerator, so more technical details are described in the following Sections.
- **18/9 MeV cyclotron accelerator** [85]. It accelerates protons and deuterons up to 18 and 9 MeV, respectively. Its main use is the synthesis of radiopharmaceuticals, so it has seven targets for β^+ emitters production, and an additional one in an external line to perform research studies.
- **SARA (Spanish Accelerator for Radionuclide Analysis)** [86]. An Accelerator Mass Spectrometer (AMS) for ultrasensitive detection of long-lived isotopes. Because of its versatility, it is mainly used for the study of environmental radioactivity and nuclear residues.
- **MiCaDaS (Mini Carbon Dating System) accelerator** [87]. A compact radiocarbon (^{14}C) dating system. This dating technique has been used in the archaeology field and in forensic applications, among others.
- **PET/CT clinical scanners** [88]. Used by the Diagnostic Imaging Center (CDI) at CNA, it is available for clinical scanning of humans and animals. Studies of short-lived isotopes also have been carried out with these instruments [89].
- **^{60}Co irradiator** [88]. Dedicated to irradiations with photons, it is equipped with a 357 TBq (as of December 2013) ^{60}Co source. Its main uses are electronic irradiation and materials study.

3.2. The HiSPANoS facility at CNA

The HiSPANoS (HiSPAlis NeutrOn Source) facility is the first accelerator-based neutron source in Spain. As mentioned before, it is one of the experimental lines of the Tandem accelerator available at CNA.

The neutrons at HiSPANoS are produced via nuclear reactions when a proton or deuterium beam impinges on a D, Li or Be target. Thanks to the latest accelerator upgrade, now it is possible to operate in continuous and pulsed mode. The pulsing system was commissioned for the first time using epithermal neutrons produced by the $^7\text{Li}(p,n)$ reaction [90]. Several activation measurements have been successfully performed at HiSPANoS using an epithermal neutron flux, with similar techniques as the one described

in our work (see Refs. [91, 92]). More recently, the capabilities to produce continuous and pulsed fast neutron beams with deuterium reactions have been characterized. Monoenergetic neutron beams between 2 and 6 MeV, and broad white energy spectra up to 20 MeV can be delivered [93]. These neutron beams are available for different applications as nuclear technology, detector characterization and neutron imaging [94], among others. By moderating all these neutron fluxes, thermal neutrons can also be produced in the facility [95], although new efforts are being made to develop an optimized moderation system.

3.2.1. Ion acceleration system

HiSPANoS is installed at the 3 MV 9SDH-2 Tandem Pelletron accelerator from National Electrostatic Corp. (NEC), right after a 90 degrees magnet that deviates the beam in case that it has to be sent to one of the other seven beam-lines available in the accelerator. The layout of the accelerator is shown in Fig. 3.1. HiSPANoS is located at 0° with respect to the acceleration system, so the magnet must be switched off to allow the ions to reach the neutron line. There are three ion sources available at the accelerator:

- **Alphatross.** The ion production is achieved using radio frequency techniques on gases. This source is mainly used to produce alpha particles, but it can also produce H^- , NH^- and O^- ions.
- **Duoplasmatron.** A plasma is generated by ionizing a gas using electrons emitted from a cathode filament. This makes possible to produce very bright proton beams.
- **SNICS II.** It is based on the cesium *sputtering* of solid samples. A large variety of ions can be produced, and because a very high intensity can be achieved with H and D beams, it is the ion source usually used in neutrons experiments at CNA, including the one described in this work.

Once the ions are extracted from the source, they are then guided towards the Pelletron tank by an injection magnet. In the low energy region of the accelerator, just between the injection magnet and the entrance tank, the pulsing system is located. It is composed of two elements:

- The *chopper*, composed by a couple of deflecting plates perpendicular to the beam. A 650 kV voltage is applied to one of the plates, thus always deflecting the beam, being turned off to let the ions continue towards the beam-line. This system allows to form pulses with a temporal width between 40 and 250 ns, with a frequency from 31.2 kHz to 2 MHz.

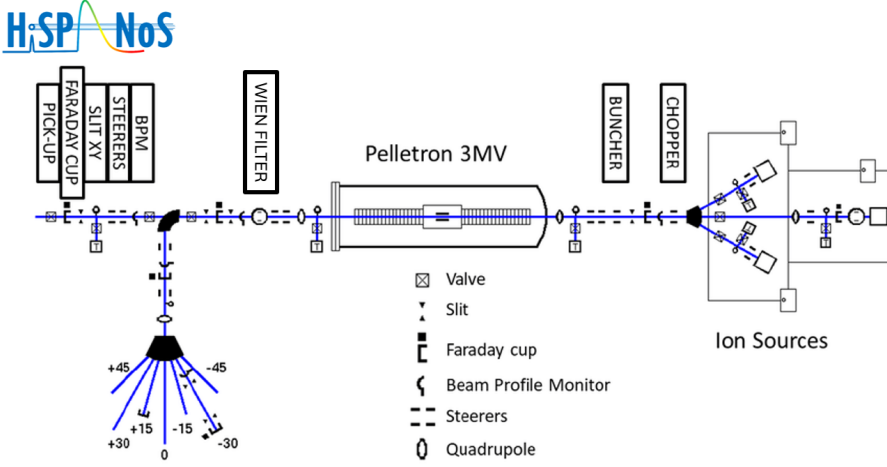


Figure 3.1: Schematic layout of the Tandem accelerator at CNA. The HiSPANoS beam-line is located after the 90 degrees magnet, at the left part of the picture. Figure adapted from Ref. [96].

- The *buncher*, to compress the "chopped" pulse. Using Radio-Frequency signals, it decreases the energy of the ions at the beginning of the pulse and increases it at the end. In this way, a temporal width of around 2 ns is achieved at the neutron source position, at the end of the beam-line.

When the ions enter the tank, they are accelerated in two steps. First, the positive voltage applied by the terminal in the middle of the tank accelerates the ions. Afterwards, the ions lose electrons in a *stripping* process, so they are accelerated towards the exit of the tank by a repulsive force. The ion beam then passes through a Wien filter that selects the required ion velocity and therefore the desired energy. The following element is the 90 degrees magnet, and afterwards the HiSPANoS line. The final energy of the ions is given by the following expression:

$$E = (q + 1)V + J, \quad (3.1)$$

where q is the charge of the ion, V the tank terminal voltage and J the injection voltage from the source. In addition, as explained in the following Section, the accelerator is calibrated regularly to determine with accuracy the actual energy of the ions.

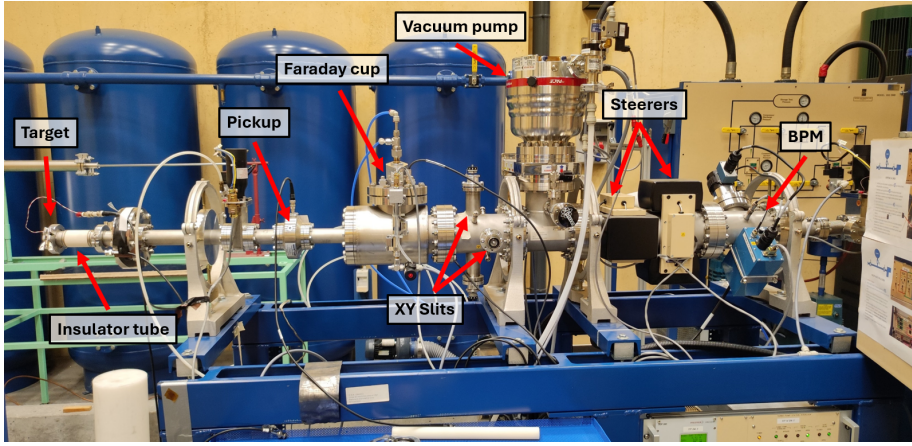


Figure 3.2: HiSPANoS beam-line, indicating the main elements.

3.2.2. HiSPANoS beam-line

There are several elements installed along the neutron beam-line to adjust and monitor the beam (see Fig. 3.2). Two magnetic Steerers are present to move the beam and center it in the desired position, and with Ta XY slits the maximum aperture of the beam can be defined. With a Beam Profile Monitor (BPM) the shape and position of the beam can be determined without interfering with it, and with a Faraday cup the current can be measured. In addition, there is a capacitive pickup to monitor the temporal width and frequency of the pulses.

The neutron target is placed at the end of the beam-line, coupled to a ceramic insulator tube. A bias of 300 V was applied to suppress the secondary electrons produced when the beam impinges on the target, that would otherwise affect the measurement of the current. The beam current at the target is monitored with an ORTEC 439 Digital Current Integrator module. This allows to register the current second by second during the whole irradiation.

The target is placed facing one of the corners of the tandem hall, so the closest distance to the walls is 4 meters. When performing time-of-flight measurements, the detectors can be placed in almost any angle relative to the neutron source, being the maximum distance in the forward direction 3 meters. This is due to the fact that neutrons can be scattered in the walls and enter the detector from behind, so if it is placed too close, the "rebounds" can affect the measurement (see Ref. [94] for more details). Longer flight paths are possible for wider angles.

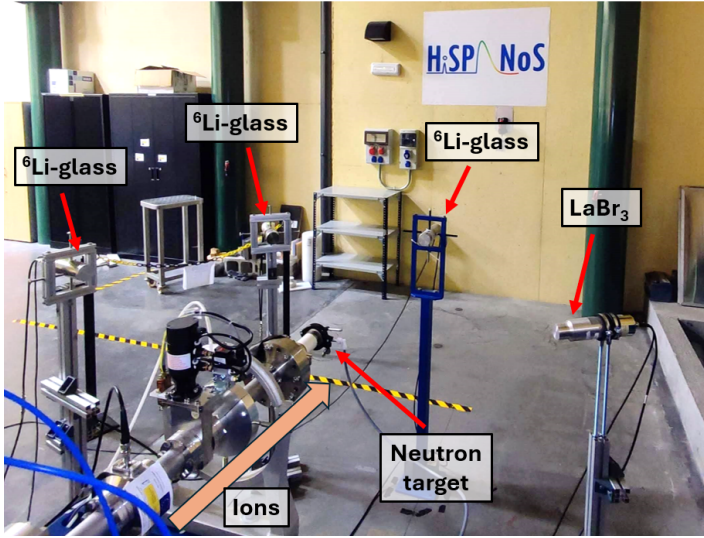


Figure 3.3: Example of experimental set-up for a time-of-flight measurement at HiSPANoS. The neutron detectors can be placed at different distances and angles, and the γ -flash can be characterized with a LaBr_3 detector.

3.2.3. Neutron detection set-up

One remarkable feature of HiSPANoS is the possibility of performing time-of-flight measurements. For that purpose, there are different kinds of detectors available depending of the energy distribution of the neutrons of interest in the measurement:

- Organic liquid scintillators for fast neutrons. Because of its large hydrogen content, the neutrons are detected by the recoil of H or C ions after a elastic scattering [97], or by nuclear reactions with C.
- ^6Li lithium-glass detectors for epithermal neutrons [98]. They are based on the $^6\text{Li}(n,\alpha)^3\text{He}$, detecting the energy released in the reaction.

The ^6Li -glass detector is the optimal choice for the characterization of a neutron distribution like the one used in this work, so more details about its performance can be found in Section 3.3.2. In addition, usually a LaBr_3 is placed along with the neutron detectors, to characterize and optimize the time resolution of the pulse by detecting the γ -flash that always appears with the neutron production reactions. A typical configuration for performing time-of-flight measurements at HiSPANoS is shown in Fig. 3.3.

In a time-of flight measurement, the time synchronization is fundamental. At HiSPANoS, the clock signal from the chopper is delayed and sent to the Data Acquisition System (DAQ) to open a coincidence window, so only signals detected in coincidence with the

clock signal in that window are registered. The DAQ is made of a CAEN V1751 digitizer with 8 channels of 10 bit resolution and 1Gs/s. Every time the chopper activates (so every time that the pulsed ion beam is generated), a logic signal is produced to open the acquisition window. This signal, all the signals from the detectors and the signal from the beam current impinging on the target are registered for their offline analysis. The most important quantities that are necessary in time-of-flight measurements are the total energy deposition and the energy deposition of the fast part of the signal, to discriminate neutrons from other elements as γ -rays, and the time stamp.

3.3. Experimental set-up for MACS determination

The standard methodology to perform a MACS measurement was established by Ratynski and Käppeler in [76], so our experimental methodology is based on the ideas presented in that reference. It describes how a quasi-maxwellian neutron spectrum (QMNS) at a temperature of $kT = 30$ keV can be produced via the ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction, with 1912 keV protons. As mentioned in Section 3.2, there have been several experiments at HiSPANoS to characterize the epithermal neutron spectrum produced by this reaction and to irradiate different isotopes in order to obtain a MACS at $kT = 30$ keV (MACS₃₀).

At HiSPANoS, there are two kinds of lithium targets for epithermal neutron production purposes: a 16 μm thick LiF target deposited on a 500 μm Cu backing and a metallic Li target. The LiF target is self-sustained and can be reused in a large number of irradiations, but the neutron production per incident proton is ~ 3.4 times lower than that with a metallic Li target. However, a fresh Li target has to be produced with every irradiation campaign, and because of its chemical properties it has to be manipulated in some special conditions. Because of the low neutron capture cross section of ${}^{50}\text{Cr}$, the metallic Li option was chosen for the irradiation, and the details of the characteristics of the target are described in the following section.

3.3.1. Irradiation set-up

A thick metallic Li target was inserted in an aluminium cooled target holder similar to the one reported in [99] (see Fig. 3.9). It was designed so that the target inside could be refrigerated with water or air, being the latter sufficient to avoid the melting of the lithium at 181 °C. Because of its high reactivity, the metallic Li oxidizes within minutes in contact with the air, so it has to be handled inside an inert atmosphere. A lithium disk of around 23 mm diameter and 200 μm thickness was extracted from a Li tube inside a chamber full of Ar, and was placed there inside the aluminium holder. At the end of the beam-line, before the Li target a 12 mm diameter and 175 μm thick

aluminium collimator was installed to limit the maximum aperture of the beam and to ensure that it impinges on the Li target completely.

A calibration in energy is performed annually in the Tandem accelerator by using very well-known resonances of the reactions $^{27}\text{Al}(p,\gamma)^{28}\text{Si}$ and $^{24}\text{Mg}(p,p'\gamma)^{24}\text{Mg}$, which emit photons of 991.86 and 2409 keV respectively, allowing for an energy resolution of around 1 keV. Nevertheless, the proton beam energy was checked before the irradiation. A Thermo Scientific BIREM FHT-751 neutron dosimeter was placed at 0° close to the Li target. The accelerator terminal was set under the $^7\text{Li}(p,n)$ reaction threshold of 1880.6 keV, increasing the energy while registering the neutron dose. The results were compared to the expected neutron production from PINO [82], a versatile online tool to simulate the neutron production based on the cross section of Liskien and Paulsen [100]. In this way, the beam energy was determined with an accuracy of 2 keV. In addition, simulations were performed in order to access the effect of a 2 keV variation in the correction factors and the final MACS (see Section 3.4). The simulations proved that the variations would be below 1%.

The injection voltage of the ion source was set to 59 kV, and the terminal of the accelerator to 920 kV, corresponding to a proton energy of (1912 ± 1) keV according to all the verifications described above. From previous time-of-flight measurements, the proton energy spread was determined to be 3.5 keV, mainly due to the pulsing process of the beam (see the following Section for more details).

3.3.2. Epithermal neutron field characterization by time-of-flight

As mentioned earlier, an exhaustive commissioning campaign was carried out to characterize the epithermal neutron field produced at HiSPANoS [90]. Before the ^{50}Cr irradiation campaign, an additional time-of-flight measurement was performed to check that the neutron spectrum produced was the expected one. This served as an additional validation of the irradiation set-up.

A ^6Li -glass detector from SCIONIX was placed at 515 mm and 0 degrees with respect to the neutron target. This detector is based on the $^6\text{Li}(n,\alpha)^3\text{H}$ reaction, so what is detected is the energy produced in the alpha-tritium break-up reaction, equivalent to 4.783 MeV (the Q-value of the reaction) plus the kinetic neutron energy. Since, with our beam configuration, the neutrons are produced in a continuum with a maximum energy of ~ 120 keV, it is impossible to determine the neutron energy distribution just with the deposited-energy spectrum. On top of that, when the γ -flash is very intense like in our case, the signals from γ -rays and neutrons cannot be distinguished (see left panel of Fig. 3.4). Because of this, the time-of-flight technique is necessary in these cases.

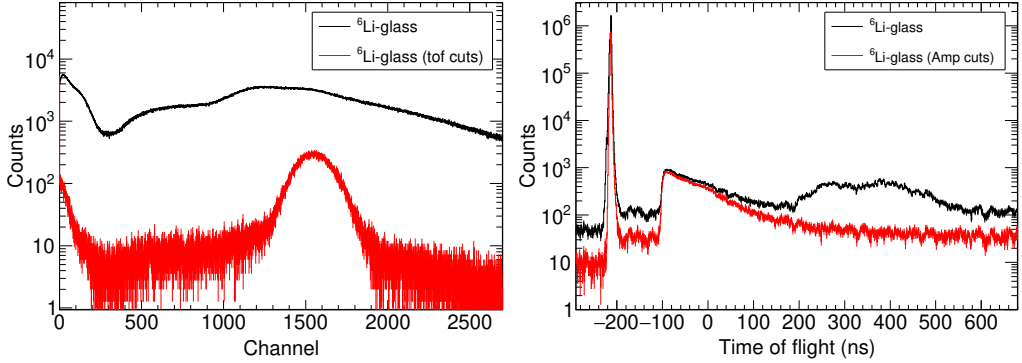


Figure 3.4: Left: Energy deposited spectrum before (black) and after (red) applying cuts in time-of-flight, to find the signals relative to neutrons. Right: Time-of-flight spectrum before (black) and after (red) applying the cuts in deposited-energy shown at the left panel, reducing the background.

The Li-glass detection efficiency is given by the ${}^6\text{Li}(n,\alpha)$ cross section and the thickness of the glass. The thicker the glass, more neutrons are going to react with the crystal and the measuring time needed to accumulate enough statistics will decrease. However, if the glass is too thick, the relation between the flight path and the time-of-flight is not univocal, since the neutron can interact at any position inside the crystal. In addition, neutrons emitted in wider angles travel a longer distance. All these effects are defined by the Resolution Function (RF) of the facility (see for example the n_TOF facility RF in Section 5.2.3), and has to be determined via Monte Carlo simulations. To minimize the influence of the RF, the measurement was performed with a Li-glass of 3 mm thickness, the thinnest available at HiSPANoS.

The chopper was set to form 45 ns proton pulses with a frequency of 500 kHz, and the buncher was tuned until a 2 ns pulse width was achieved. In this configuration, the target was irradiated during 3.5 hours with protons of 1912 keV, receiving 378.3 μC in total. The result of the measurement is displayed in Fig. 3.4. At the left panel, the energy deposited (in ADC channels) spectrum shows that there is no separation between the γ -rays and neutrons signals, because of the large amount of γ -rays produced. In the time-of-flight spectrum(right panel) the first peak observed is the γ -flash, and then the neutrons. The "bump" visible between 300 and 600 ns corresponds to delayed-background, mostly γ -ray signals of de-excitation and decay of different elements in the target holder, beam line and the experimental hall. Furthermore, a flat background is visible during the whole time window, corresponding to neutrons being scattered in the walls and the floor and going back to the detector.

To clean these histograms, a two-step process was performed:

1. The neutrons are located in the time-of-flight histogram at around 100 ns after the γ -flash. That time-of-flight cut was applied to the energy-deposited spectrum, so now the neutrons signals are visible between channels 1200 and 1900 (left panel of Fig. 3.4).
2. Only the signals between those channels are considered, resulting in the time-of-flight spectrum of the right panel of Fig. 3.4. As it is obvious, all of the γ -rays signals inside the cut are going to be included in the spectrum.

The final outcome is that the γ -flash is reduced, and the "bump" on top of the neutrons has disappeared (right panel of Fig. 3.4). But the neutron-scattering background is still present, so it also has to be removed. One common way of doing so is by measuring with a *shadow bar* [101]. A shadow bar is some element placed between the neutron source and the detector capable of capturing every neutron in the trajectory of the detector, so only the scattering background is detected (no neutron reaches the detector directly). In this work we have implemented another technique, that has already worked in other measurements at HiSPANoS. After measuring with a shadow bar, what one gets is basically the same flat background visible in the normal measurements [90]. What we propose is to fit that background directly in the neutron spectrum, and apply the time-of-flight equation to that value, to obtain the equivalent background in neutron energy units. The process is shown in Fig. 3.5, so the resulting background of the right panel now can be subtracted to the neutron energy spectrum.

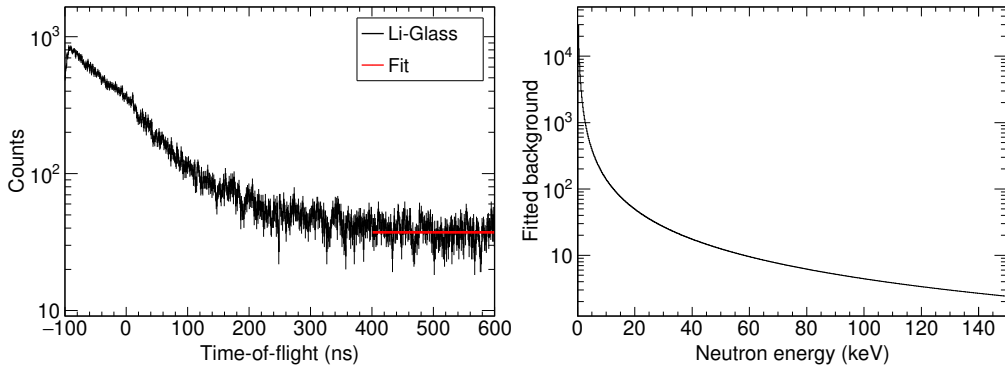


Figure 3.5: Left: Detail of the last part of the time-of-flight spectrum, in which the neutron-scattering background is fitted as a flat background. Right: Fitted background, which is flat in time-of-flight, presents a quadratic shape in the neutron-energy equivalent.

The clear time-of-flight spectrum is then transformed into neutron energy and the scattering background is subtracted. The data is weighted by the ${}^6\text{Li}(n,\alpha)$ cross section and the Li-glass thickness to correct for the efficiency, and normalized by the number of

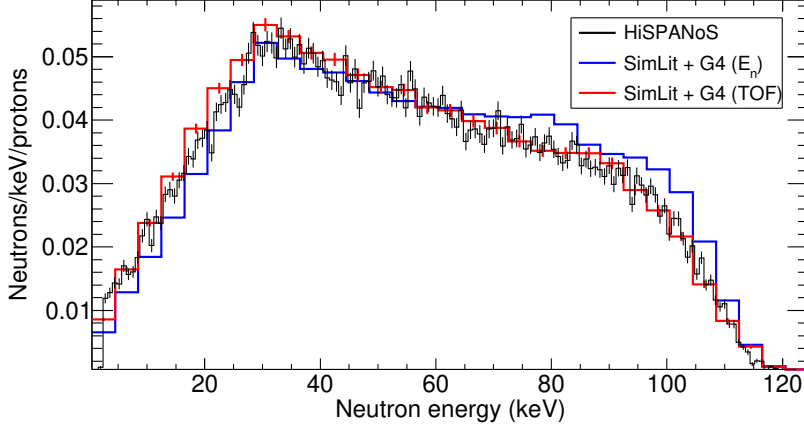


Figure 3.6: Neutron energy spectrum measured at HiSPANoS (black line) compared with the simulated distribution by scoring the energy of the neutrons (blue line) and the time-of-flight of a neutron capture inside the crystal (red line) to take into account the RF effects.

protons registered. The final result is shown in Fig. 3.6.

To compare this result, Monte Carlo simulations were performed with a realistic model of the neutron target and the Li-glass detector (see Section 3.4 for more details). The results of the simulations are shown in Fig. 3.6. To take into account the RF, not only the energy of the neutrons arriving at the detector was scored (blue line), but also the time between the emission of a neutron and a ${}^6\text{Li}(n,\alpha)$ reaction (red line). To the *simulated* time-of-flight spectrum, the same treatment was applied to the one for the experimental one, so the result is more realistic.

As can be seen in Fig. 3.6, the results are perfectly compatible. A slight decrease of the neutron energy is appreciable in the measured and simulated one when compared to the energy of the neutrons arriving to the detector. This is due to the RF, because of the neutrons that suffer some scattering inside the Li-glass or are emitted with wider angles, so their time-of-flight is larger than the corresponding energy. Also, according to the kinematics of the neutron production reaction, the maximum energy should correspond to 108 keV. The fact that the spectrum ends at 120 keV is due to the proton energy spread of ± 3.5 keV, which was again confirmed by the measurement and simulations. Thanks to this measurement, we confirmed that the neutron flux irradiating the spectra was the correct one.

3.3.3. ${}^{50}\text{Cr}$ sample

A high purity of enriched ${}^{50}\text{Cr}_2\text{O}_3$ was purchased for the measurement. A natural sample would not fulfill the necessities of the experiment for several reasons:

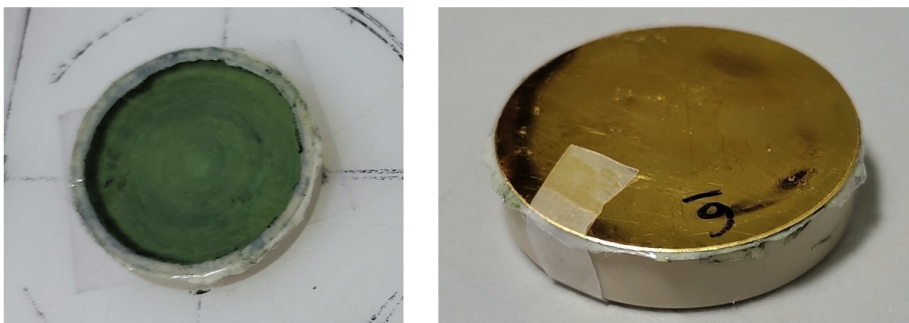


Figure 3.7: Left: Enriched $^{50}\text{Cr}_2\text{O}_3$ powder pressed inside a PEEK capsule. Right: Two gold foils glued at both sides of the capsule, to determine the Cr MACS₃₀ relative to the standard ^{197}Au MACS₃₀.

1. The abundance of ^{50}Cr in natural chromium is only 4.34%, so a very thick sample would be needed in order to produce enough ^{51}Cr nuclei.
2. As a consequence, in a natural sample more than the 95% of neutron interactions would be undesired captures and scattering, and the main goal was to reduce as much as possible the multiple-scattering correction.
3. The decay parameters of ^{51}Cr (see Section 4.1) would make the irradiation time necessary to accumulate statistics even longer.

For similar reasons, the n_TOF measurement also needed of enriched samples, so the same sample was used in both facilities.

The material of the sample was purchased from Trace Science International, consisting in a fine powder of 94.6(4)% ^{50}Cr enriched oxide (containing 4.92%, 0.4% and 0.08% of ^{52}Cr , ^{53}Cr and ^{54}Cr respectively). The powder was pressed into a 20 mm diameter, 0.5 mm thick and 4.5 mm tall PEEK capsule, adjusting the amount to the required atomic thickness (see Table 3.1). A very thin Mylar layer was glued at the top of the capsule to avoid losing material in case of detachment. The final outcome is shown at the left panel in Fig. 3.7.

A series of X-ray Computed Tomography (CT) images were performed and analyzed by the Mechanical and Materials Engineering (MME) Group at CERN, in which the thickness and the homogeneity of the pressed sample was assessed. Local variations of the order of 10% were detected in the surface of the powder, as well as a slightly concave meniscus in the walls of the capsule, as can be seen in Fig. 3.8. To estimate the effect of these inhomogeneities on the correction factors (see Section 4.2.1), GEANT4 [102, 103] simulations of the neutron capture and scattering in a sample perfectly homogeneous

Table 3.1: Physical characteristics of the ^{50}Cr and ^{197}Au samples.

Sample	Diameter (mm)	Mass (mg)	n_{at} (10^{-4}at/b)
^{50}Cr	20.0(1)	723(3)	18.73(7)
$^{197}\text{Au}_{\text{front}}$	22.0(1)	778.1(8)	6.258(7)
$^{197}\text{Au}_{\text{back}}$	22.0(1)	795.4(8)	6.397(6)

and with the mentioned imperfections were performed. The resulting correction factors and MACS₃₀ values varied less than 1%.

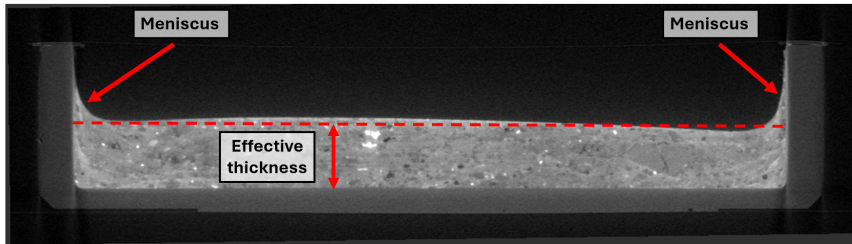


Figure 3.8: X-ray Computed Tomography (CT) of the ^{50}Cr sample. The meniscus at capsule walls and the variation in the thickness are pointed out. This image served to estimate the inhomogeneities and to investigate its effect on the MACS₃₀ determination.

As it is usual in neutron capture activation measurements, the MACS₃₀ of ^{50}Cr was measured relative to the ^{197}Au standard cross section. In order to estimate the accuracy of the corrections, two gold foils were glued at both faces of the sample. The final outcome of the process is shown in the right panel of Fig. 3.7. Usually only one gold sample is enough to determine the MACS₃₀ with very good accuracy, but because of the thickness and scattering problems of our sample we decided to use two foils. In this way, the effect of the neutron scattering upstream and downstream the sample, as well as the accuracy of the correction factors for each case, can be assessed. The physical characteristics of the samples are summarized in Table 3.1.

A schematic drawing of the whole target-sample assembly is displayed at the left panel of Fig. 3.9, where the Li inside the target holder and the Cr inside the PEEK can be seen. A picture of the assembly is shown at right panel of Fig. 3.9.

3.3.4. Activation detection set-up

As each activated sample emitted just one detectable γ -ray (see Section 4.1), it was not necessary to employ detectors with a very good energy resolution, but rather to use a fast detector with a relatively high efficiency. The decay of the samples was measured

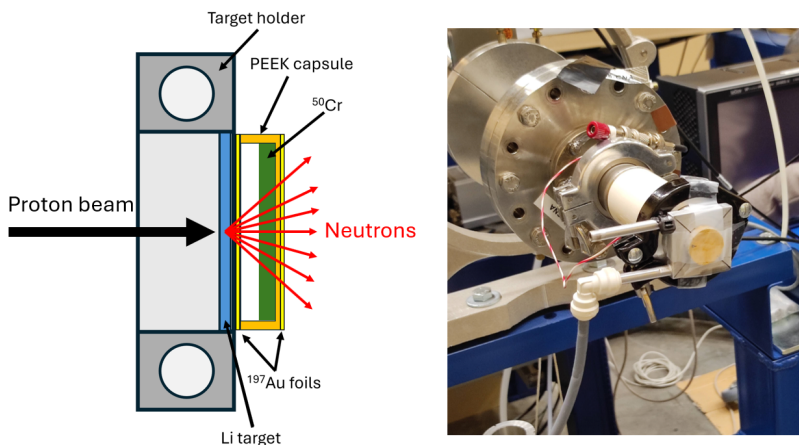


Figure 3.9: Schematic drawing (left) and photo (right) of the target holder with the Li inside and the sample formed by the Cr inside the PEEK and two Au foils.

with a cylindrical 1.5"×1.5" LaBr₃(Ce) Canberra detector. It was coupled to a PMT and connected to a 14 bits CAEN DT5730 digitizer.

The detector was fixed to a table with marks on it to determine precisely the distance at which the samples was placed. This was important since the samples were measured at different distances because of the very low activity in the chromium sample (see Section 4.1.1 for more details).

3.4. SimLiT-GEANT4 simulations

There are multiple correction factors that have to be taken into account for an accurate MACS determination. The Li target is placed inside an aluminium target holder, so the neutrons will undergo some scattering and the original spectrum is going to be slightly altered. There is also a geometrical factor due to the cylindrical shape of the samples, thus the *apparent* thickness of the samples is going to be larger for neutrons produced in wider angles. In addition, the scattering produced by the 1.8 mm Cr sample and the PEEK is going to modify the spectrum as well.

The most precise way of determine the actual neutron distribution traversing each sample is via simulations. A detailed model of the target and samples assembly was implemented in GEANT4, as seen in Fig. 3.10. To ensure that the neutron interactions were being accurately simulated, the High Precision (HP) physics list of GEANT4 were used, including evaluated neutron cross sections libraries [104, 105]. SimLiT [106] is a code developed to produce neutron energy and angular distributions regarding the ⁷Li(p,n)

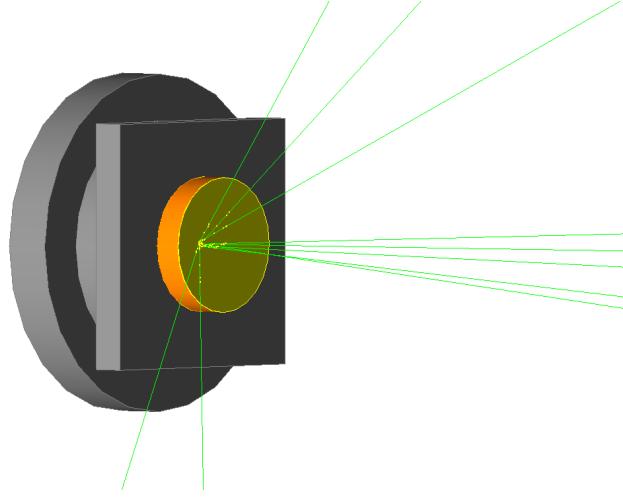


Figure 3.10: GEANT4 model of the activation set-up, including the Li target holder, the Cr sample inside the PEEK capsule and the two gold foils. The green line represents neutrons being emitted.

reaction (among others), so it was coupled to the GEANT4 model. The simulation consisted then in the production of a realistic neutron distribution and the transport of these neutrons along the described geometry. The energy of the neutrons when traversing each sample was scored, assigning a *weight* equal to the distance travelled inside the sample divided by the thickness. This is equivalent to weighting by the cosine of the angle when the "very thin sample" approximation is considered, which is not valid here because of the thickness of the ^{50}Cr sample.

The simulated neutron distributions normalized to its maximum is displayed in Fig. 3.11, along with ideal MB distributions of $kT = 25$ and 30 keV. The visible peak at the Cr and Au_{front} samples and the dip in the Au_{back} is due to the main ^{50}Cr resonance at 5.6 keV, where neutrons have the highest probability of undergoing an interaction. There is also a slightly decrease in energy regarding the Au_{back} , because of the neutrons losing energy after being scattered in the preceding material. As can be appreciated in Fig. 3.11, the energy distribution of the neutrons does not exactly resemble a maxwellian shape. For this reason, a correction factor that takes into account the difference between the ideal and the experimental neutron field that irradiates the samples must be introduced in the analysis. This will be explained in detail in Section 4.2.1.

In Fig. 3.12 the energy of the neutrons just before (left) and after suffering a scattering process (right) in the different samples is displayed. Is evident that, in the epithermal region of interest in our work, the neutron scattering is dominated by the ^{50}Cr elastic

cross section, even for the gold samples where the ^{50}Cr main resonance effect is also visible.

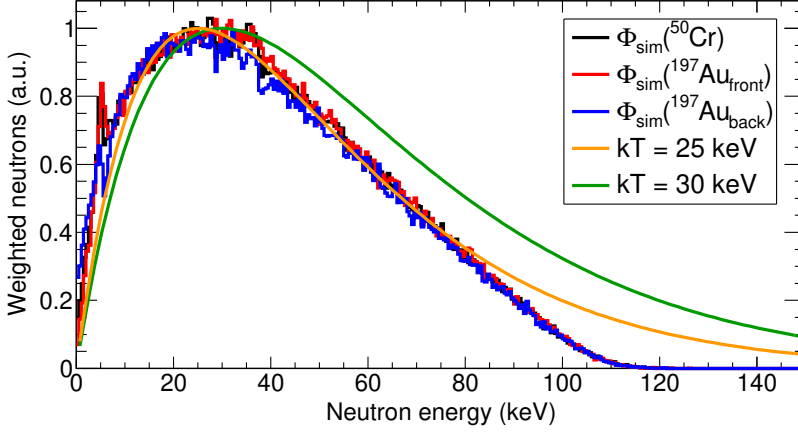


Figure 3.11: Simulated distribution of neutrons traversing each sample and weighted by the distance travelled inside them, compared with the ideal Maxwell-Boltzmann distributions at $kT = 25$ and 30 keV .

Last, the scattering and the *weighting* also affects the total number of neutrons impinging on the samples. The neutron flux in the chromium sample is 2.5% and 10% larger than the *front* and *back* gold samples respectively, probably due to the scattered neutrons in the PEEK capsule that go back to the chromium. This causes that a correction factor f_{Φ} has to be included in the analysis to take these differences into account.

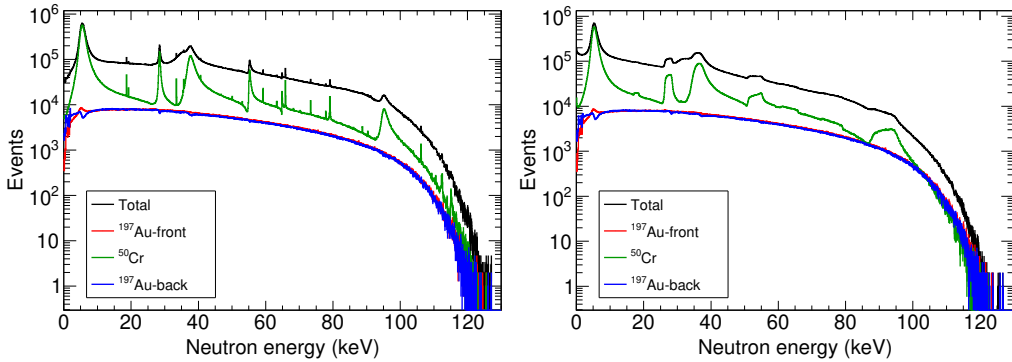


Figure 3.12: Neutron energy just before (left) and after undergoing a scattering (right), at the different elements of the sample, according to the simulations. The process is dominated by the ^{50}Cr elastic cross section.

4 | Analysis and determination of the ^{50}Cr MACS

In the previous Chapter we have described the experimental set-up and the work previous to the irradiation, specially to characterize the neutron flux impinging on each sample. In the following, we develop all the analysis regarding the activation to determine the number of produced ^{51}Cr and ^{198}Au nuclei and the relevance of the correction factors obtained with simulations. Lastly, the final MACS of ^{50}Cr reached in this work is compared with the previous values available on the libraries and evaluations.

4.1. Neutron activation analysis

As mentioned earlier, the activation technique can only be implemented if the product nuclei of the reaction are unstable, so the number of reactions can be determined through the analysis of the decay.

For the case of ^{50}Cr , after capturing a neutron the ^{51}Cr produced will decay to ^{51}V via electron capture with a half-life of $T_{1/2} = 27.704(3)$ days [107], emitting a single 320.1 keV γ -ray with an intensity of $I_\gamma = 9.91(1)\%$. Regarding ^{197}Au , ^{198}Au decays to ^{198}Hg via β^- with a half-life of $T_{1/2} = 2.6941(2)$ days, emitting mainly a 411.8 keV γ -ray with $I_\gamma = 95.62(6)\%$. The nice and suitable decay parameters allow for a relatively simple γ -ray detection set-up, even for the ^{51}Cr decay (the longer half-life and smaller intensity makes its measurement slightly more challenging).

4.1.1. LaBr_3 efficiency determination

The γ -ray efficiency of the $\text{LaBr}_3(\text{Ce})$ detector was obtained with a combination of measurements and simulations. The first step was to determine the efficiency curve measuring a series of calibrated radioactive sources: ^{22}Na , ^{60}Co , ^{133}Ba , ^{137}Cs and ^{152}Eu . The same sources were used to calibrate the detector in energy. According to the manufacturer sources, their activity was known withing 1.5%, so this number was included

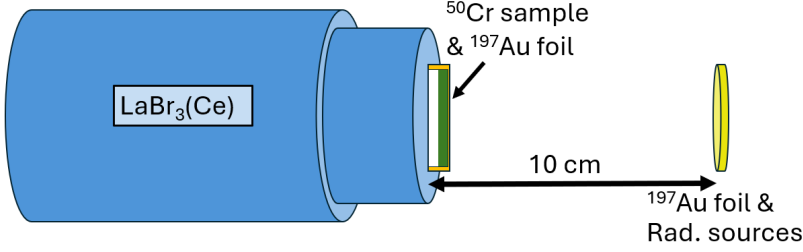


Figure 4.1: Schematic drawing of the $\text{LaBr}_3(\text{Ce})$ detector and the distances at which the samples were measured: the calibration sources at 10 cm, the gold foils at 10 cm and in contact with the detector and the Cr sample also in contact (not in scale).

on the efficiency uncertainty. The sources were placed at 100(1) mm of the detector, the same distance that the gold samples were measured. The efficiency curve shown in Fig. 4.2 was obtained by fitting the experimental points with the following analytical function:

$$\varepsilon_\gamma = \exp \left(\sum_{i=0}^3 a_i \ln^i(E_\gamma) \right). \quad (4.1)$$

In the case of ^{197}Au , some correction factor K needs to be added to the γ -ray efficiency ε_γ to take into account the geometrical extension of the gold foils and the self-absorption of the photons inside of them, so the total efficiency $\varepsilon = K\varepsilon_\gamma$ is the true efficiency for the corresponding E_γ and the measured sample. This factor was estimated with GEANT4 simulations of the sample and the detector assembly.

Regarding the ^{50}Cr sample, the induced activity was too low to measure the decay at the same distance that the gold samples, so it had to be measured almost *in contact* with the front face of the detector (see Fig. 4.1). To determine the efficiency in this case, several steps were followed:

1. The γ -ray efficiency for the ^{51}Cr sample inside the PEEK and in contact with the detector was simulated with GEANT4.
2. The ^{197}Au samples were also measured in contact with the detector, to obtain the increase factor of the efficiency.
3. The previous situation was simulated, to check if the relation between the efficiency for the ^{197}Au sample in contact and at 100 mm was reproducible through simulations.

Following this procedure the total efficiency regarding the ^{197}Au and ^{50}Cr samples are $3.39(6) \cdot 10^{-3}$ and $158(5) \cdot 10^{-3}$ respectively. The efficiency is almost 47 times larger for

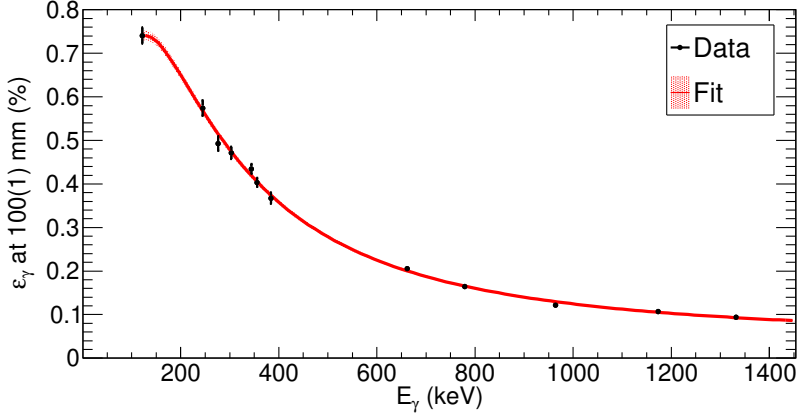


Figure 4.2: $\text{LaBr}_3(\text{Ce})$ γ -ray efficiency calculated from the radioactive calibration sources placed at 100(1) mm from the front face of the detector. The solid line corresponds to a fit to the analytical function of Eq. 4.1.

the chromium than for the gold samples, allowing for a better decay measurement, even when the main source of uncertainty affecting the experiment is this efficiency.

In the end, what is used to determine the ^{51}Cr production is the *relative* efficiency between the gold samples at 100 mm and the Cr sample in contact (see Eq. 4.5), to reduce systematic uncertainties. The results obtained in the points 2. and 3. were consistent within 1.7%, so this factor was included in the ^{51}Cr efficiency uncertainty. The ϵ values and uncertainties for each sample are summarized in Table 4.1 and 4.2 respectively.

4.1.2. Results of the activation

The irradiation lasted for 4.7 hours with an average proton beam current of 5.03(6) μA . The current registered by the Current Integrator is shown in Fig. 4.3. This information allowed us to estimate the decay during the irradiation.

The number of activated nuclei (N_{act}) in the samples was obtained from the activity at the end of irradiation (A_{EOI}) as

$$N_{act} = \frac{A_{EOI}}{\lambda} = \frac{C}{I_\gamma \epsilon} e^{\lambda t_w} (1 - e^{-\lambda t_m})^{-1} f_{irr}, \quad (4.2)$$

where λ is the decay constant, C is the total number of counts in the photopeak, I_γ the emission intensity, t_w the waiting time between the end of irradiation and the beginning of the measurement, t_m the duration of the measurement and f_{irr} a correction factor to

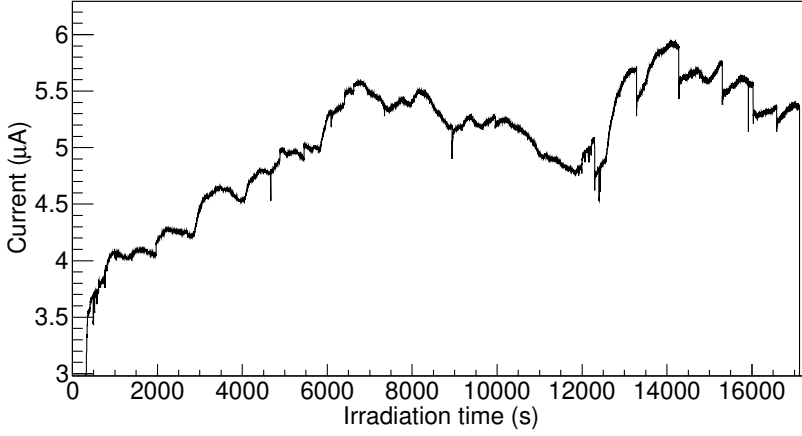


Figure 4.3: Proton current impinging on the Li target during the irradiation. With this information the correction factor f_{irr} to determine the number of activations can be estimated.

account for the decay during the irradiation.

An example of the γ -decay spectrum of ^{198}Au is shown in Fig. 4.4. One known issue with LaBr_3 detectors is their large intrinsic background, mainly due to the radioactive ^{138}La present in natural lanthanum and also alpha peaks from the U and Th natural decay chains [108]. For samples with enough activity the background is not a big problem, since it is very well characterized and can be subtracted for the spectrum. Another possibility is to fit the background in the vicinity of the region of interest, as it was done in this work (see Fig. 4.5).

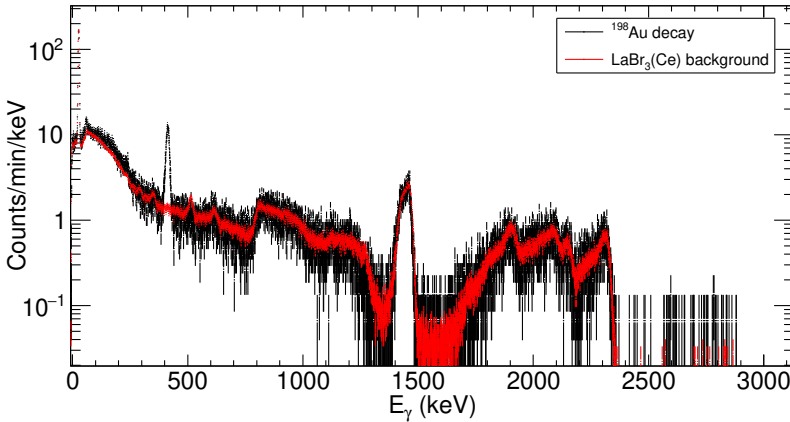


Figure 4.4: γ -decay spectrum of one of the gold samples along with a background measurement. Even the $\text{LaBr}_3(\text{Ce})$ detector has a considerable intrinsic background, it is perfectly characterized and can be subtracted with a background measurement of a couple of hours.

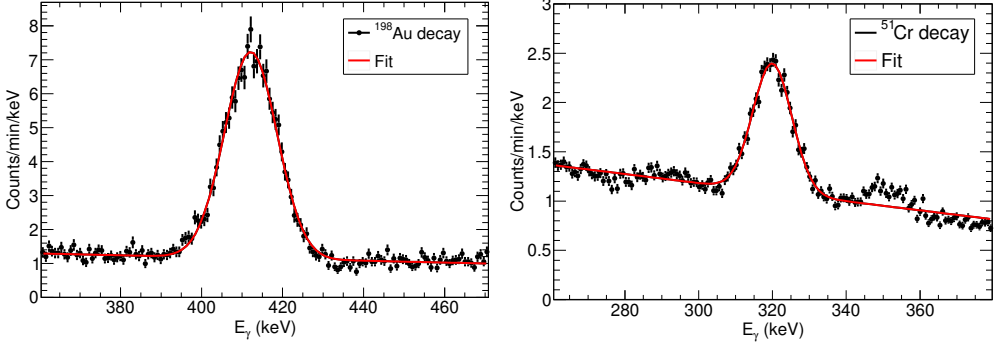


Figure 4.5: Photopeaks from the main γ -ray emissions of ^{198}Au (left) and ^{51}Cr (right) as detected with the $\text{LaBr}_3(\text{Ce})$ detector. The solid lines correspond to fits to a Gaussian peak on top of a linear background.

The number of counts C was determined by fitting the γ -spectrum to a Gaussian peak on top of a linear background, as shown in Fig. 4.5; f_{irr} was estimated using the current measurement during the irradiation. The value of the current was registered every second, so it was possible to estimate the number of captures and decays in every step of the irradiation. The desired value is not the *total* number of captures or decays, but the *fraction* between produced and decayed nuclei, so only the decay constant λ and the variation of the current is needed (not the number of captures per proton or similar quantities). The obtained values of f_{irr} for ^{51}Cr and ^{197}Au are 0.998(2) and 0.977(2) respectively, the first one being a smaller correction because of the longer half-life of ^{51}Cr (since obviously the current variation is the same for the three samples, but not λ).

The ^{51}Cr decay was measured for 4.5 hours, and the Au_{front} and Au_{back} for 15 and 20 minutes respectively. This measurement was part of a larger campaign, in which a large number of samples were irradiated and measured, thus the limited time available for measuring the decay and the waiting time before the gold samples could be measured. Despite this, the statistical uncertainty is less than 1.7% for all the samples .

The details of the measurement and the results of the activation are displayed in Table 4.1, and the uncertainties are shown in Table 4.2. The number of activations in the ^{50}Cr , $^{198}\text{Au}_{front}$ and $^{198}\text{Au}_{back}$ is $79(3) \cdot 10^6$, $434(11) \cdot 10^6$ and $425(10) \cdot 10^6$ respectively. The slight difference between both gold samples are due the different neutron spectra impinging on them, as a result of the Cr sample influence. The final uncertainty is 3.6% for the ^{50}Cr sample, and 2.6% and 2.4% for each gold sample. The statistical uncertainty is similar for all of them, and the values are dominated, specially for ^{50}Cr ,

Table 4.1: Summary of the ^{51}Cr and both ^{198}Au decay measurements, including all variables related to the calculation of the number of activated nuclei (N_{act}).

	E_γ (keV)	I_γ (%)	$C(10^3)$	$\varepsilon(10^{-3})$	t_w (min)	t_m (min)	f_{irr}	$N_{act}(10^6)$
^{51}Cr	320.0824(4)	9.91(1)	6.03(8)	158(5)	24.00(2)	270.08(2)	0.998(2)	79(3)
$^{198}\text{Au}_{front}$	411.8021(2)	95.62(6)	3.49(6)	3.39(6)	301.00(2)	15.03(2)	0.977(2)	434(11)
$^{198}\text{Au}_{back}$			4.52(7)		341.00(2)	20.00(2)		425(10)

Table 4.2: Sources of uncertainty (in %) contributing to the overall accuracy of N_{act} .

	^{50}Cr	$^{197}\text{Au}_{front}$	$^{197}\text{Au}_{back}$
I_γ	0.1		0.6
ε	3.3		1.8
C	1.3	1.7	1.5
$e^{\lambda t_w}$	0.01	0.08	0.07
$e^{-\lambda t_m}$	0.01	0.01	0.01
f_{irr}	0.2		0.2
Overall	3.6	2.6	2.4

by the efficiency determination (see Section 4.1.1).

4.2. ^{50}Cr MACS determination

4.2.1. Simulated correction factors

The neutron energy spectrum produced in the $^7\text{Li}(p,n)$ does not reproduce perfectly a maxwellian distribution, as it was shown in Fig. 3.11. There are a few methods proposed to obtain a neutron flux with a better maxwellian distribution, like shaping the proton beam using a carbon degrader [109]. When this is not the case, like in the measurement described here, what can be actually measured is the Spectrum Averaged Cross Section (SACS), defined as

$$\text{SACS} = \frac{\int_0^\infty \Phi(E_n) \sigma(E_n) dE_n}{\int_0^\infty \Phi(E_n) dE_n}, \quad (4.3)$$

with $\Phi(E_n)$ the neutron distribution used in the experiment. In our case, $\Phi(E_n)$ corresponds to the simulated neutron flux in each sample as shown in Fig. 3.11, so we will name it $\Phi_{\text{sim}}(E_n)$. To convert the measured SACS into the desired MACS_{kT} there is a factor $f_{S \rightarrow M}$ that can be obtained with the following relation:

$$f_{S \rightarrow M} = \frac{\int_0^\infty E_n e^{-E_n/kT} \sigma(E_n) dE_n / \int_0^\infty E_n e^{-E_n/kT} dE_n}{\int_0^\infty \Phi_{\text{sim}}(E_n) \sigma(E_n) dE_n / \int_0^\infty \Phi_{\text{sim}}(E_n) dE_n}. \quad (4.4)$$

Obviously this factor depends on the evaluated cross section chosen to perform the

calculations. In other measurements this dependence is small, but in our case it has a considerable effect in the final result. The reason is that, because chromium is a relatively light element, its main resonances are in the keV region, precisely the region of interest where the evaluations differ significantly.

The SimLiT-GEANT4 simulations were then performed including the JEFF-3.3 and the INDEN libraries (since ENDF/B-VIII.0, JENDL-5, BROND-3.1 and CENDL-3.2 are equivalent to JEFF-3.3), and the results are shown in Table 4.3. The results regarding the SACS and the correction factors of both gold samples when using each library are consistent within 0.4%, so the mean value was used in the rest of the analysis, being the SACS 669 and 701 mb and f_Φ 1.024 and 1.099 for Au_{front} and Au_{back} respectively. On the other hand, the $f_{S \rightarrow M}$ values for chromium are 0.793 with JEFF-3.3 and 0.753 with INDEN. There is a 5% difference due to the uncertainties present in the cross section evaluation in both libraries.

The uncertainty regarding the simulated factors is very difficult to estimate, since it would be incorrect to assign just the number of simulations or the uncertainty in the cross section libraries as the total uncertainty. Consequently, the uncertainty of the final MACS_{30} will be estimated through the spreading of the partial results, as described in the following section.

Given the sizeable difference on $f_{S \rightarrow M}$, the number of neutron captures in each sample was scored in the simulations, and the same analysis procedure explained in this chapter was applied, as if it was the real measurement. The MACS_{30} values of ^{50}Cr obtained this way was 37.8 mb with the JEFF-3.3 library and 44.3 mb with INDEN, matching the expecting values within 1.5% (see Table 1.3). The good level of agreement, better than the 5% difference in the $f_{S \rightarrow M}$ values, shows that the libraries are well implemented in the simulations and validates the analysis strategy.

4.2.2. Results and uncertainties

As mentioned along the text, the SACS of ^{50}Cr has been measured using the SACS of ^{197}Au as a reference, by means of the following expression [110]:

$$\frac{\text{SACS}_{\text{Cr}}}{\text{SACS}_{\text{Au}}} = \frac{N_{\text{act,Cr}}}{n_{\text{at,Cr}}} \frac{n_{\text{at,Au}}}{N_{\text{act,Au}}} f_\Phi, \quad (4.5)$$

where SACS_{Au} is obtained using Eq. 4.3, n_{at} is the atomic thickness and f_Φ is a factor that takes into account the different number of neutrons received by each sample (see Section 3.4), estimated with the SimLiT-GEANT4 simulations.

Combining the previous expression with Eq. 2.25 and 4.4, the aimed MACS_{30} is then

Table 4.3: Correction factors (f_Φ and $f_{S \rightarrow M}^{\text{Cr}}$) and cross sections (SACS and MACS₃₀) corresponding to the measurement with respect to either gold sample and using two different cross section evaluations.

	$^{197}\text{Au}_{\text{front}}$	$^{197}\text{Au}_{\text{back}}$
SACS(^{197}Au) (mb)	669	701
f_Φ	1.024	1.099
SACS(^{50}Cr) (mb)	44.2	51.8
<u>Simulations with JEFF-3.3:</u>		
$f_{S \rightarrow M}^{\text{Cr}}$		0.793
MACS ₃₀ (^{50}Cr) (mb)	37.9	38.5
<u>Simulations with INDEN:</u>		
$f_{S \rightarrow M}^{\text{Cr}}$		0.753
MACS ₃₀ (^{50}Cr) (mb)	35.7	36.4

See text for the discussion on the uncertainties.

determined as

$$\text{MACS}_{30}(^{50}\text{Cr}) = \frac{2}{\sqrt{\pi}} f_{S \rightarrow M} \text{SACS}(^{50}\text{Cr}). \quad (4.6)$$

The results obtained regarding Au_{front} and Au_{back} taken as the reference sample and for both JEFF-3.3 and INDEN libraries for the correction factors are displayed in Table 4.3. When using the JEFF-3.3 library, the MACS₃₀ of ^{50}Cr is 37.9 mb according to Au_{front} and 38.5 mb according to Au_{back} , and with INDEN, 35.7 mb and 36.4 mb (the uncertainties are discussed further in the text). The MACS₃₀ obtained using each gold sample indicate an agreement of 1.9%, even the sizeable differences in the respective f_Φ and $f_{S \rightarrow M}$ correction factors, providing confidence to the analysis strategy.

Because the experimental set-up allows it, another verification was made to consolidate the analysis by calculating the MACS₃₀ of the Au_{back} sample relative to Au_{front} . Following the same calculations described for Cr, taking into account that now $f_\Phi^{\text{Au}} = 1.073$ and $f_{S \rightarrow M}^{\text{Au}} = 0.771$ according to the simulations, we have obtained $\text{MACS}_{30}(^{197}\text{Au}) = 601(23)$ mb. This result is in agreement with the standard IAEA value of 620(11) mb [111] within 3%, therefore validating the analysis and results for ^{50}Cr .

The final MACS₃₀ of ^{50}Cr has been determined as the average of the four different results given in Table 4.3. This gives a value of 37.1 mb, with a 3% standard deviation of (1.1 mb). The spread in the partial results is not due to any of the uncertainty factors included in Table 4.2, so it serves as an estimation of the uncertainty introduced in the simulations of the correction factor. The total uncertainty is then the combination of the standard deviation and the values of Table 4.2, resulting in an overall accuracy in

the MACS_{30} of 5.3%. The final result of this work is then

$$\text{MACS}_{30}(^{50}\text{Cr}) = 37.1(20) \text{ mb.} \quad (4.7)$$

4.3. Comparison with previous data and evaluations

The most recent values available in the databases, the evaluated MACS_{30} from the libraries and the result of this work are summarized and compared in Fig. 4.6. This new value is the first direct experimental result obtained by ^{50}Cr neutron activation.

It is clear that our result is in agreement with the CENDL-3.2 and JEFF-3.3 evaluations, and with the rest of evaluations that adopted the same parameters that JEFF-3.3: ENDF/B-VIII.0, JENDL-5 and BROND-3.1. It is also in agreement with the values reported in the KADoNiS-1.0 and Mughabghab compilations. On the other hand, there is a 20% difference with the most recent evaluation INDEN.

It is very important to point out that we have estimated all the simulated correction factor to determine the MACS_{30} using JEFF-3.3 and INDEN as a neutron cross section libraries, and both results point out the same conclusion outlined above. It can be concluded that there is clearly a 20% overestimation in the evaluated cross section incorporated to INDEN.

This result is discussed further in Chapter 7 together with those from the differential cross section measurements presented in the next part of this work.

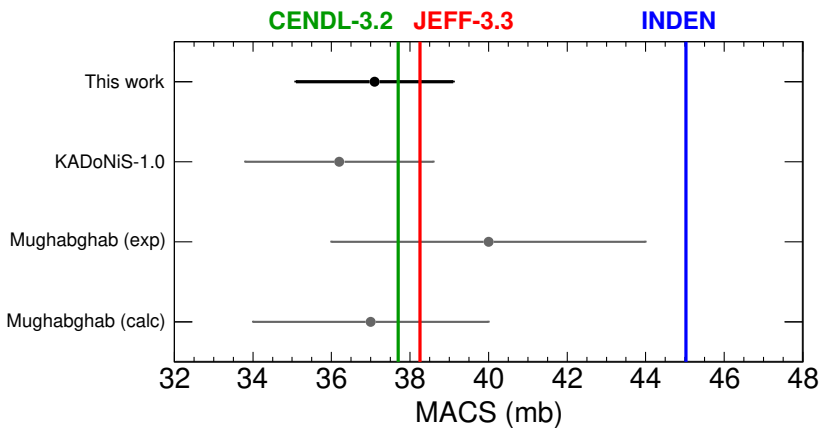


Figure 4.6: ^{50}Cr MACS at $kT=30$ keV from this work compared with previous values provided in databases and calculated from different cross section evaluations (JEFF-3.3 $\equiv$ ENDF/B-VIII.0 $\equiv$ JENDL-5 $\equiv$ BROND-3.1).

Part III

Time-of-flight measurement at n_TOF

5 | The experimental set-up at n_TOF-EAR1

The following Part of this work is dedicated to the time-of-flight measurement of the $^{50,53}\text{Cr}$ neutron capture cross section between 1 and 100 keV. In Chapter 5 we describe the n_TOF facility, focusing on the EAR1 experimental area, where our measurement took place, the capture detection set-up and the samples employed in the measurement. The steps followed to obtain a capture yield are detailed on Chapter 6, and the R-matrix analysis performed to obtain the final cross section is described in Chapter 7.

5.1. The n_TOF facility at CERN

The neutron Time-Of-Flight (n_TOF) facility at CERN has been in operation since 2001. Proposed originally by the Nobel Prize laureate C. Rubbia in 1998 [112], it was designed to measure neutron induced reactions with a high precision over a wide energy range (from meV to GeV). During its entire running time, a large number of measurements have been performed related to astrophysics (stellar nucleosynthesis), nuclear technology and nuclear medicine mainly, in addition to detector development, basic nuclear physics and others [113, 114]. There have been four upgrades during the CERN shutdowns to improve the performance and capabilities of n_TOF. The measurement described in this work was performed during n_TOF Phase 4, characterised by the installation of the third generation spallation target [115] during CERN Long Shutdown 2 (2019-2021).

The neutrons at n_TOF are produced by spallation [57]. The Proton-Synchrotron (PS) of CERN accelerator complex (see Fig. 5.1) sends 20 GeV/c² proton pulses to n_TOF with an average repetition rate of 0.8 Hz and a temporal root mean square (RMS) of 7 ns. Each proton pulse impinges on a cylindrical lead spallation target, producing around 300 neutrons per proton. Two kinds of proton pulses are sent to n_TOF: Dedicated and Parasitic, with an intensity of $\sim 8 \cdot 10^{12}$ and $\sim 3 \cdot 10^{12}$ protons per pulse, respectively.

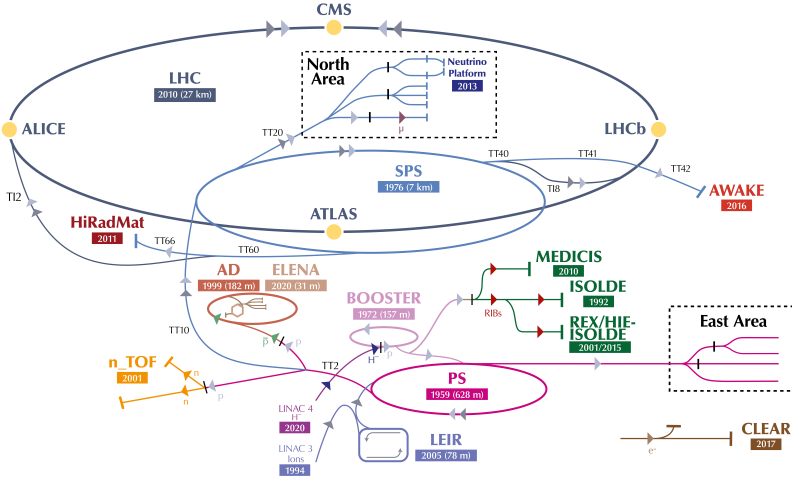


Figure 5.1: Schematic figure of CERN accelerator complex. n_TOF is one of the facilities that make use of the Proton-Synchrotron (PS) very intense proton pulse. From Ref. [116].

The produced neutrons are moderated in the vicinity of the target and then travel towards the different experimental areas available at n_TOF as indicated in Fig. 5.2:

- **EAR1** [61]. The first experimental area in operation, is located at 185 meters horizontally from the spallation target. Because of the long flight path, the area presents an excellent energy resolution.
- **EAR2** [62]. In operation since 2014, it was built vertically at 20 meters above the spallation target. The resolution is not as good as in EAR1, but the ~ 30 times more intense neutron flux allows for measurements of radioactive and/or low masses samples.
- **NEAR** [63, 117]. In commissioning since 2021, is located right next to the target, at only 3 meters. The extremely intense neutron flux provides the possibility of performing neutron activation, material irradiation or similar experiments.

The goal of the measurement described herein was to achieve a very good precision in the epithermal energy range between 1 and 100 keV, where the main resonances of chromium are found. Therefore, EAR1 was chosen because its better resolution allowed resolving the mentioned resonances.

The neutrons produced in the spallation target have an energy between MeV and GeV, so they have to be moderated in order to extend to lower energies the neutron beam arriving to the experimental areas. In this work, the neutrons travelling towards EAR1 were moderated with borated water. The fraction of neutrons that travels towards EAR1

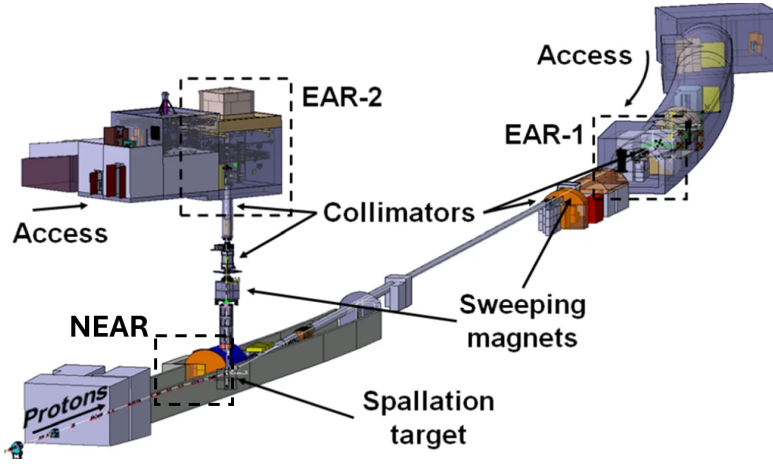


Figure 5.2: Layout of the n_TOF facility, showing the relative position of the spallation target, the main elements of the beam-lines and the three experimental areas (figure not in scale). Modified from Ref. [118].

enters the beam line and comes across a first collimator that removes its halo. Then, a sweeping magnet deflects the charged particles produced in the target, and a second collimator is placed at the end of the line to shape the neutron beam accordingly to the measurement to be performed. Usually a small opening collimator of 18 mm diameter is used for capture measurements and a larger one, of about 80 mm diameter, for fission measurements that requires larger samples.

5.2. Performance of the n_TOF-EAR1

5.2.1. Neutron flux

One of the main features of n_TOF is its very high instantaneous neutron flux, providing an excellent signal-to-background ratio that is specially relevant in the measurements of low mass and/or radioactive samples.

There are several systems at n_TOF to measure the value and energy dependence of the neutron flux, all based on the IAEA standard neutron reaction cross sections [111] depicted in Fig. 5.3 (see also Table 5.1):

- **Silicon Monitor (SiMon)** [119]: consisting on a enriched ${}^6\text{Li}$ neutron converter and four silicon detectors to register the alpha and triton particles created in the ${}^6\text{Li}(n,\alpha){}^3\text{H}$ reaction.

- **Micromegas (μ MGAS)** [120, 121]: gas detector loaded with ^{10}B and ^{235}U , to register the alphas and fission fragments from $^{10}\text{B}(\text{n},\alpha)^7\text{Li}$ and $^{235}\text{U}(\text{n},\text{f})$ reactions.
- **PTB** [122]: a calibrated H19 fission chamber from *Physikalisch Technische Bundesanstalt* (PTB) loaded also with ^{235}U samples.
- **Parallel Plate Avalanche Counters (PPAC)** [123, 124]: a gas fission chamber loaded with two ^{235}U samples, specially designed to be very transparent so that very high neutron energies (1 GeV) can be achieved.

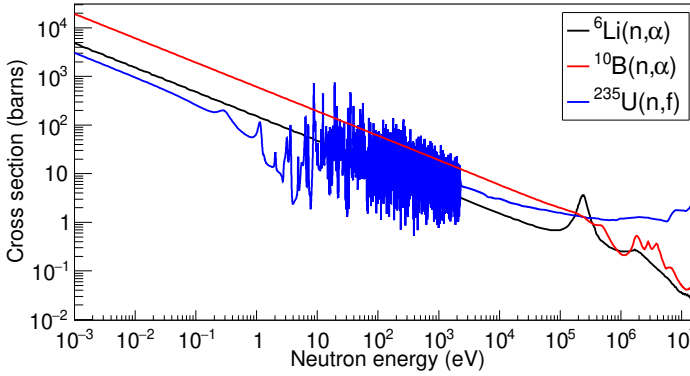


Figure 5.3: Standard neutron cross sections on which the different systems at n_TOF are based to determine the flux.

Table 5.1: Energy range in which the neutron reactions used to characterize the n_TOF flux are considered standard.

Reaction	Energy range
$^6\text{Li}(\text{n},\alpha)$	25 meV - 1 MeV
$^{10}\text{B}(\text{n},\alpha)$	25 meV - 1 MeV
$^{235}\text{U}(\text{n},\text{f})$	25 meV and 0.15 MeV - 200 MeV

These measurements are complemented with dedicated Monte Carlo simulations of neutron production and transport in the whole experimental area, with codes like MCNP [125] and FLUKA [126]. The outcome is the so-called *evaluated flux* shown in Fig. 5.4. The spectrum covers the range from thermal energies to 1 GeV, being dominated by the evaporation peak at 1 MeV, and the decrease in the number of thermal neutrons is due to the ^{10}B present in the moderator as already mentioned. The visible dips in the epithermal energy range are due to transmission resonances of the materials that the neutrons transverse before arriving to EAR1, mainly from ^{55}Mn and ^{27}Al . The evaluated flux at EAR2 is also shown for comparison: is much intense than at EAR1, and also the thermal peak is visible because there is no boron to capture the thermal

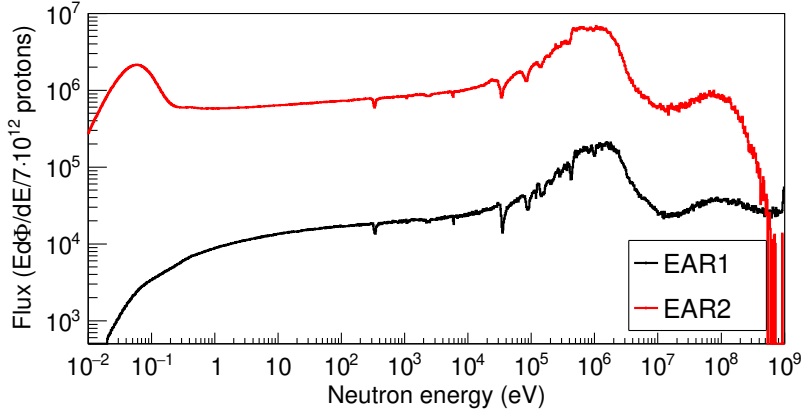


Figure 5.4: Evaluated neutron flux at EAR1 corresponding to the Phase 4 of n_TOF (in black). The flux in EAR2 (in red) is also displayed for comparison.

Table 5.2: Number of neutrons per nominal pulse ($7 \cdot 10^{12}$ protons) in EAR1 for the relevant energy ranges.

Energy range	Neutrons per pulse
1 eV - 10 eV	$2.6 \cdot 10^4$
1 keV - 10 keV	$5.0 \cdot 10^4$
10 keV - 100 keV	$7.0 \cdot 10^4$

neutrons in the moderator of EAR2. A summary of the number of neutrons per pulse in the different energy ranges of interest is displayed in Table 5.2.

5.2.2. Neutron beam profile

As mentioned previously, the size of the neutron beam at the position of the sample is defined by the second collimator. Although its diameter is only 18 mm, the beam opens as it travels further from the collimator opening, achieving 40 mm in diameter at the sample position. With samples smaller than this size, as is our case, a very precise knowledge of the beam profile is crucial to determine the fraction of neutrons impinging on the sample, which is known as Beam Interception Factor (BIF).

The determination of the beam profile was carried out with the PPAC detector in combination with MC simulations during the commissioning of Phase 4. It has been observed that the beam profile not only depends on the distance to the collimator: thermal neutrons are shifted down because of the gravity, and the fast neutrons are emitted more in the center of the spallation target compared to the moderated neutrons that are emitted from a larger area. For the intermediate energies of interest in this work, 1 eV to

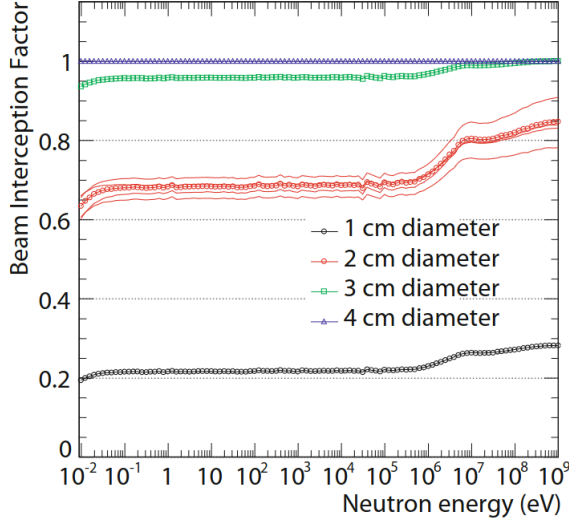


Figure 5.5: Beam Interception Factor (BIF) as a function of the neutron energy regarding the capture collimator of EAR1 and samples of several diameters. The red solid lines reflect the BIF value when moving the sample 1 mm vertically and horizontally. From Ref. [61].

100 keV, the BIF is constant within around 1%.

The red solid lines in Fig. 5.5, show that a misalignment of a 20 mm sample of only 1 mm alters the BIF value by up to 3%, making this factor one important source of systematic uncertainty in a measurement (these results correspond to Phase 2 [61], but being a purely geometrical effect it can be considered valid also for Phase 4). In our case, the chromium capture yield is determined relative to the ^{197}Au saturated resonance, thus what is important is that both samples are in the same position relative to the beam. Therefore, special care was taken to position the samples in the beam with sub-millimeter accuracy with the aid of a laser alignment system (see right panel of Fig. 5.12).

5.2.3. Resolution function

One important aspect regarding the time-of-flight measurements at n_TOF and similar facilities is the Resolution Function (RF) [127]. Because of the width of the proton pulse and the moderation of the neutrons, specially in the spallation target, the relation between time-of-flight and neutron energy is not univocal. In fact, it is a distribution: neutrons with the same kinetic energy arrive at different times. The RF determines this

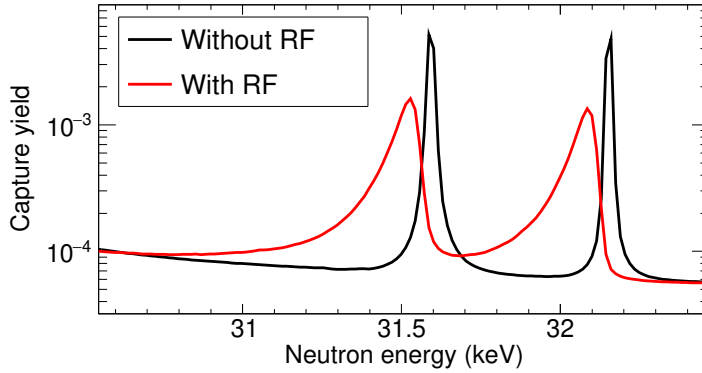


Figure 5.6: Effect of the Resolution Function on two capture resonances of ^{53}Cr . The yield has been obtained with the SAMMY code.

distribution, and it can be expressed in term of energy (E_n), time (t) or flight path (L):

$$R_E(E_n)dE_n = R_t(t)dt = R_L(L)dL \quad (5.1)$$

It is not possible to obtain the RF by any kind of measurements, so it has to be determined by Monte Carlo simulations [128]. A dedicated campaign was carried out during the commissioning of Phase 4 to this matter, validating it by measuring well-known resonances.

The RF affects the resonances by changing their shape and by shifting its energy when reconstructed from the measured time-of-flight, effects that are considered in the analysis of the observed resonances by including the RF in, for instance, the SAMMY code (see Chapter 7). This RF can be described by a analytical function or a numerical representation. In Fig. 5.6, the effect of including the numerical n_TOF RF on SAMMY when calculating a theoretical capture yield for ^{53}Cr can be appreciated.

5.2.4. The background at EAR1

Alongside neutrons, during the spallation process a large number of particles are produced, contributing to the background present in the experimental areas. Because most charged particles are deflected by the sweeping magnet, the remaining background is composed mainly by photons, and it has been exhaustively studied through simulations of the spallation process [128]. The background can be divided in two components: *prompt* and *delayed*.

The *prompt* component is the one arriving in less than 1 μs . It consists of very energetic

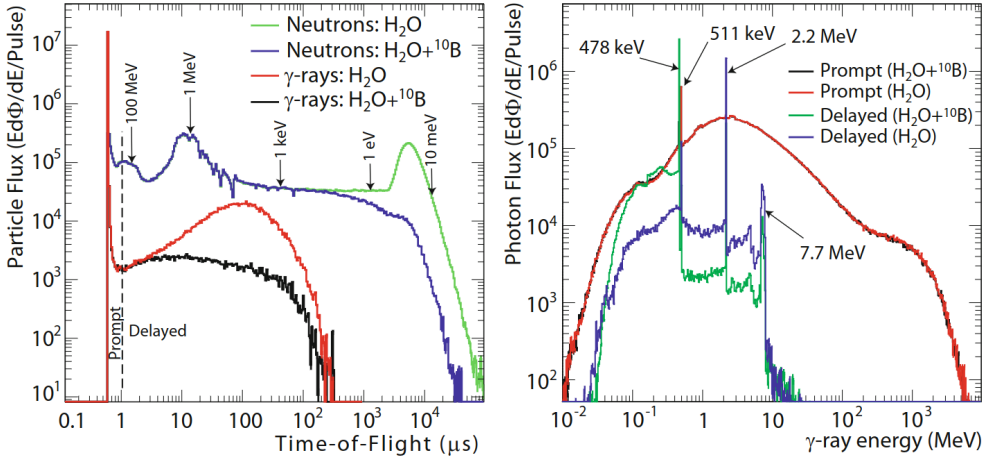


Figure 5.7: Left: Time-of-flight of neutrons and background for EAR1. There is a clear separation between the two background components (*prompt* and *delayed*). Right: Energy distribution of the background, where the 478 keV, 511 keV, 2.2 MeV and 7.7 MeV lines of the *delayed* component come from $^{10}\text{B}(n,\alpha)$, pair annihilation, $^1\text{H}(n,\gamma)$ and $^{26}\text{Al}(n,\gamma)$, respectively. From Ref. [61]

photons and relativistic charged particles from spallation reactions, in addition to a fraction of photons from the in-flight decay of short lived spallation products, mainly π^0 . All of this forms the so-called γ -flash, that cause a saturation on the response of the detectors during a certain time at the beginning of each acquisition. As it is made of particles travelling at a known speed (the speed of light c), the γ -flash serves as the reference time t_γ for the time-of-flight determination.

On the other hand, the *delayed* component arrives in times larger than 1 μs , and its origin are γ -rays from neutron captures in the target/moderator assembly. Therefore, the energy of these photons will be the characteristic one of the material where they have been produced, mainly from $^1\text{H}(n,\gamma)$, $^{10}\text{B}(n,\alpha)$ and $^{26}\text{Al}(n,\gamma)$ reactions (see right panel of Fig. 5.7). Thanks to the presence of boron in the moderator, the 2.2 MeV γ -ray component from capture in ^1H is shifted towards the 478 keV line from ^{10}B reactions. Because of its considerably lower energy, its effect is much less harmful in the measurements, specially in the case of the C_6D_6 detectors where the efficiency for low energy photons is very small (see Sect 5.3.1).

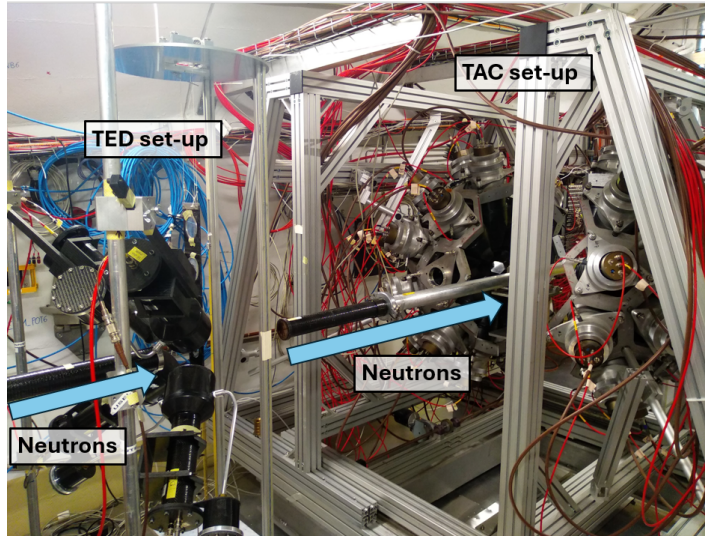


Figure 5.8: EAR1 at n_TOF. The four C_6D_6 detectors can be seen at the left, and the TAC assembly at the right, downstream the neutron beam.

5.3. Experimental set-up

There are two set-ups available at EAR1 to perform capture measurements, each with different features that makes them more suitable for one kind of measurement or another: the Total Absorption Calorimeter (TAC) [71] and an array of four optimized C_6D_6 detectors [69]. Both set-ups are shown in Fig. 5.8.

The TAC available at EAR1 in n_TOF consists of an array of 40 BaF_2 crystals covering 95% of 4π . Its working principle is having a 100% γ -ray efficiency to detect the full capture cascade, making it optimal for radioactive samples and/or with very low masses (for more details about the n_TOF TAC, see Ref. [71]). However, since it presents a very large neutron sensitivity and is not suitable for isotopes with high scattering-to-capture cross section ratio, it was discarded as the capture set-up of our measurement. The desired option was then the C_6D_6 detectors, which are based on the Total Energy Deposition (TED) technique, described in detail in the following section.

5.3.1. C_6D_6 Total Energy Detectors (TED)

The Total Energy Detection (TED) technique is based on the use of a detection system that fulfill two key features:

1. The efficiency for detecting a γ -ray ε_γ has to be very low, so that only one γ -ray of the capture cascade is detected. In this way, the efficiency for detecting the full

cascade ε_c is

$$\varepsilon_c = 1 - \prod_i (1 - \varepsilon_{\gamma i}) \approx \sum_i \varepsilon_{\gamma i}, \quad (5.2)$$

where the sum runs over all the emitted γ -rays in the cascade.

2. The efficiency for each γ -ray is proportional to its energy, so $\varepsilon_{\gamma} = kE_{\gamma}$. If one chooses $k = 1$, along with the previous condition, then the following is true:

$$\varepsilon_c = \sum_i E_{\gamma i} = E_c = \left(S_n + \frac{A}{A+1} E_n \right), \quad (5.3)$$

with E_c the energy of the cascade, S_n the neutron separation energy, A the atomic mass and E_n the captured neutron energy.

These two conditions make the efficiency for detecting a cascade proportional to its energy and independent of the de-excitation path.

The first time that this technique was implemented was in the 60s with the Moxon-Rae detector [68]. It consisted on a thin plastic scintillator coupled to a low Z converter material like graphite. The γ -rays from a capture event produced electrons in the converter, which are then detected in the plastic scintillator with a photo-multiplier tube. With this kind of system a very low and almost linear efficiency is achieved. But the handicap with these detectors is that the efficiency is in fact too low, and a good linearity can never be achieved in a real set-up, thus introducing large systematic uncertainties.

The next idea was to use large organic scintillators, removing the electron converter. The first time this model was used in an experiment was by Macklin and Gibbons [70] only a few years later with C_6F_6 detectors. Later on, this material was changed by deuterated benzene or C_6D_6 [129] to avoid the detection of scattered neutrons by the fluorine, which would lead to false events. This is nowadays the standard choice for the TED technique, although new approaches are recently under investigation [130, 131]. The C_6D_6 approach offers some advantages with respect to the previous ones:

- Better γ -ray efficiency, being still low enough to apply the TED technique,
- Very fast response and time resolution,
- Very low neutron sensitivity,
- Relatively cheap and simple to manufacture.

Several models of C_6D_6 detectors have been used at n_TOF for capture measurements because of its excellent features. The most recent, and therefore the one used in this work, is a model developed by the Legnaro National Laboratory (LNL). The goal com-

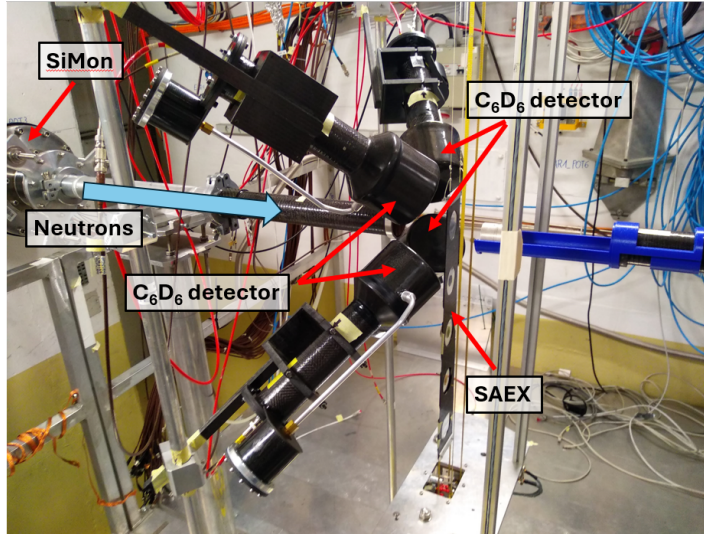


Figure 5.9: Capture set-up for the chromium experimental campaign at EAR1. The C_6D_6 detectors are placed in a 125° backward angle with respect to the sample. The sample exchanger (SAEX) where the samples are placed and moved vertically is also visible in the center of the picture.

pared to previous models [69] was to improve even more the neutron sensitivity, reducing the size of the PMT from 5 to 2 inches. Another important issue was the durability and safety inside the experimental environment, so the full detector is made by thicker carbon fiber walls of around 1.1 mm at the quartz window and 2 mm at the sides. To compensate this, a reduction in the amount of material in the PMT was included. The total active volume of the detectors is 1 L. More details about the design and manufacture of the detectors can be found in [132].

In C_6D_6 detectors, compared to the ideal Moxon-Rae ones, the proportionality condition is no longer fulfilled by construction. Instead, a methodology was developed in which the detected signals are weighted by an energy dependent Weighting Function (WF), so the weighted response of the detector makes the efficiency increase linearly with the energy. This is known as Pulse Height Weighting Technique (PHWT), and its implementation is discussed in detail in Section 6.3.

The four C_6D_6 detectors are placed at 8 cm from the sample at a backwards angle of 125 degrees with respect to the neutron beam. This position is better to minimize the energy of the detected in-beam photons undergoing Compton scattering in the sample and then reaching the detectors, and hence the efficiency and its contribution to the background. In addition, the dipole transitions after a capture could show some angular

dependence, so this effect also has to be minimized (at 125° , the second order Legendre polynomial is zero). The beam tubes and the supports for the sample and the detectors have been designed and optimized to minimize the neutron sensitivity of the whole set-up. A picture of the capture set-up is displayed in Fig. 5.9.

5.3.2. Chromium samples

A proper sample design is key to a successful capture measurement. Given the large scattering cross section of chromium compared to capture, the main goal of this experiment was to reduce as much as possible the scattering of neutrons inside the sample that affected the shape of the capture yield in previous experiments, thus thin samples are required. On the other hand, the samples must contain enough material to reach the necessary counting statistics while keeping the duration of the measurement within a reasonable time.

As already discussed in Section 1.5, the best way to sort out these problems is to use two samples: a very thin sample to measure the main resonances, i.e., between 1 and 10 keV, and a thick one for the rest of the energy range, up to 100 keV. The thin sample assures that the scattering caused by the main chromium resonances is minimized, while we achieve more statistics with the other sample for the rest of the resonances. In addition, the scattering effects can be observed and estimated by comparing the measurements of both samples. The physical properties of every sample are shown in Table 5.3.

Table 5.3: Physical properties of the enriched chromium samples. The thickness is the approximate value estimated by X-ray Computed Tomographies (CT).

Sample	Mass (mg)	Thickness (mm)	$n_{\text{at}}(10^{-3}\text{at/barn})$
^{50}Cr -thin	247(1)	0.62(3)	0.640(3)
^{50}Cr -thick	723(3)	1.81(9)	1.873(7)
^{53}Cr -thin	479(1)	1.61(5)	1.194(2)
^{53}Cr -thick	2362(5)	6.84(24)	5.885(12)

The same material used for the activation measurement of ^{50}Cr described in Chapters 3 and 4 was used for the n_TOF measurement, and the enriched $^{53}\text{Cr}_2\text{O}_3$ was purchased from the same company. The isotropic composition of each material is given in Table 5.4. The same methodology followed in that activation measurement was selected to manufacture the samples, pressing the oxide powder into PEEK capsules with 20 mm inner diameter (see Section 3.3.3). In fact, the HiSPANoS activation sample is the n_TOF ^{50}Cr -thick sample. For the case of the ^{53}Cr -thick, because of the large amount of powder employed, a 11.5 mm height PEEK capsule had to be crafted. The samples were held by Mylar at the center of an aluminium ring. Fig. 5.10 shows the ^{50}Cr -

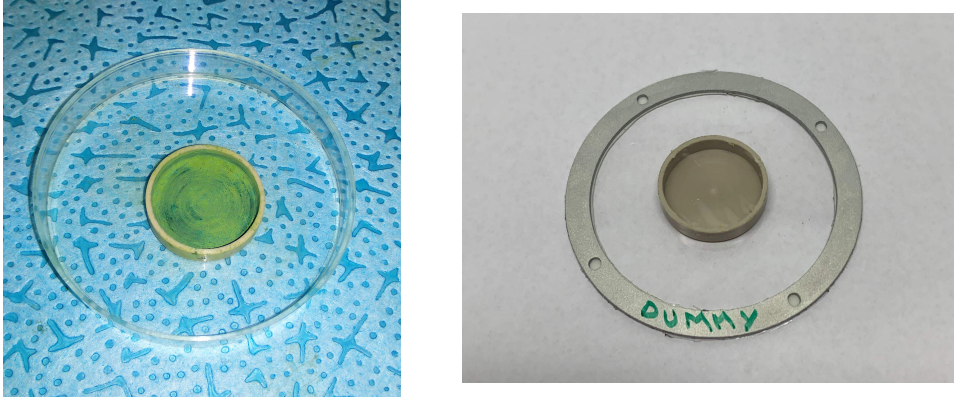


Figure 5.10: Left: ^{50}Cr -thin sample pressed inside its PEEK capsule. Right: Empty PEEK capsule sample.

Table 5.4: Isotropic composition of the enriched chromium oxide used for the measurement.

Sample	Enrichment (%)			
	^{50}Cr	^{52}Cr	^{53}Cr	^{54}Cr
$^{50}\text{Cr}_2\text{O}_3$	94.6(4)	4.92	0.4	0.08
$^{53}\text{Cr}_2\text{O}_3$	0.03	2.19	97.7(2)	0.08

thin sample (left panel) and one empty PEEK capsule (right panel). The uncertainty of the isotopic composition provided by the supplier has been considered as the mass and atomic thickness uncertainties. The availability worldwide of enriched chromium is extremely limited, so the amount used in this work was all the material that was possible to acquire. As a consequence, the statistics are not very good in the high energy part, specially for the ^{50}Cr case. Anyway, efforts were made to maximize the beam time for each sample (see Section 5.4).

As for the activation sample, several X-ray Computed Tomographies (CT) were performed by the MME Group at CERN to the rest of chromium samples to make sure that the inhomogeneities were small enough so they did not have a relevant effect on the neutron capture and scattering estimation. One example is shown in Fig. 5.11, where the meniscus are clearly visible on the sides of the sample, caused by its fabrication technique. As explained in Section 3.3.3, the simulations performed for the ^{50}Cr -thick sample showed that this meniscus had a negligible effect in the mentioned neutron processes. The average thickness of the samples estimated using the CT is also displayed in Table 5.3.

The samples were placed in the so-called *sample exchanger* (SAEX), a carbon fiber ladder in which a maximum of five samples can be placed at the same time (see left

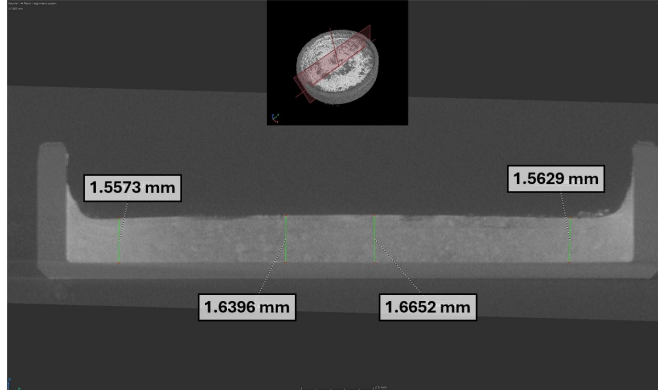


Figure 5.11: X-ray tomography of the ^{53}Cr thin sample. The TC of the samples allowed to manufacture the samples as homogeneous as possible.

panel of Fig. 5.12). The SAEX can only be moved remotely in the vertical position, so the sample in the beam can be changed without entering the experimental area and alignment errors are avoided. At the right panel of Fig. 5.12 the laser system available in EAR1 is visible, that allows to align the samples with each other and with the beam.

5.3.3. Background and auxiliary measurements

A series of ancillary measurements were performed during the experimental campaign, for background estimation or yield normalization. These measurements are the following:

- Dummy: the two versions of the empty PEEK capsule, to estimate the corresponding background in the measurement of the chromium samples.
- Empty: just the aluminium ring and the Mylar where the PEEK is placed, to estimate the beam-induced background that affects all the measurements.
- ^{197}Au : a 20 mm diameter (like the Cr samples) and 100 μm thick gold sample that serves to normalize the measured yields by means of the Saturated Resonance Method [133] (see Section 6.4.1). An 80 mm diameter sample with similar thickness was also measured to validate the calculation of the BIF.
- $^{\text{nat}}\text{C}$: a 20 mm diameter and 1 mm thick natural carbon sample (which can be considered a pure neutron scatterer), to estimate the background from neutrons scattered in the chromium samples and then captured in the detectors or surrounding materials, emitting a γ -ray.

A summary of the physical properties of the samples are displayed in Table 5.5. More details about the background determination can be found on Section 6.2.

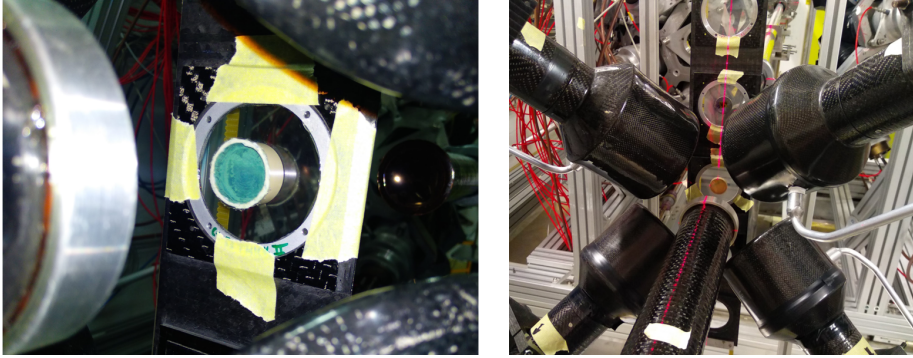


Figure 5.12: Left: ^{53}Cr -thick sample placed in the beam position. Right: General view of the C_6D_6 detectors, the SAEX and the laser system to align the samples in the correct position.

Table 5.5: Physical properties of the auxiliary samples used in the measurement. Two Dummies were irradiated, with the same diameter but different thickness.

Sample	Diameter (mm)	$n_{\text{at}}(10^{-3}\text{at/barn})$
Dummy (2)	22	-
^{197}Au	20	0.583
^{197}Au	80	0.644
$^{\text{nat}}\text{C}$	20	8.702

5.3.4. Beam monitors

The number of protons incident on the spallation target and neutrons arriving to the measuring station needs to be monitored in a bunch-per-bunch basis (a neutron pulse is known as a *bunch*). To this end, a series of ancillary system are available:

- **Beam Current Transformer (BCT).** It registers the number of protons impinging on the target pulse by pulse, serving also as the trigger for the data acquisition.
- **Pick-up.** A resistive Wall Current Monitor, that provides a signal proportional to the intensity of each proton pulse.
- **SiMon.** The same system used to assess the neutron flux (see Section 5.2.1) is used to monitor the number of neutrons arriving to the experimental area during the measurement.

The monitors are fundamental to normalize the different measurements to each other and to identify and then discard the "bad" bunches. For more details see Section 6.1.3.

5.3.5. Data Acquisition System (DAQ)

The measurements at n_TOF are very demanding in terms of signal storage and processing, thus the Data Acquisition System (DAQ) must be able to fulfill a series of requirements:

- The system must be able to handle a large number of detectors and beam monitors at the same time, meaning dozens of detectors when measuring with the TAC, at maximum counting rate.
- The counting rate can be very high when dealing with radioactive samples.
- The system must process fast signals (for instance from the C_6D_6) separated by very short times keeping the pile-up and the dead time at a minimum.
- The acquisition window has to be long enough to cover from the arrival of the γ -flash until the time corresponding to thermal energies, which is around 100 ms at EAR1.
- The data registered for each bunch has to be sent to the storage center before the following bunch arrives, about 1.2 seconds later. This means dealing with several GS/s of data transfer.

The original DAQ at n_TOF was designed to deal with all of these demands [134]. The upgraded current version [135] is based on SPDevices ADQ14DC flash analog-to-digital (ADC) units, a high performance ADC cards offering 4 channels with 14-bit resolution and 1 GHz sample rate. With their fast zero suppression algorithm they are able to exclude all the data below a certain threshold defined by the user for each channel, reducing the amount of data to be stored and analyzed.

The digitized output from each detector and monitor is stored temporally in local computers, and then sent to the CERN Tape Archive [136].

5.3.6. Pulse Shape Analysis (PSA)

Once the raw data has been stored, the signals are processed to extract the relevant information. The most relevant parameters for the capture yield analysis are the amplitude and area of the signals to extract the corresponding deposited energy, and the time stamp to determine the time-of-flight.

A generic Pulse Shape Analysis (PSA) routine has been developed in order to deal with the n_TOF data [137, 138]. This PSA aims to be versatile enough to include all the different kind of detectors available at n_TOF, with the minimum possible number of signal parameters.

The basic steps followed by the routine are the following:

1. Pulse identification based on the derivative properties of the signal.
2. Location of the γ -flash, by identifying the first signal that crosses a certain threshold or that saturates, which is the case for most of the detectors.
3. Baseline calculation, performed a certain time after the γ -flash signal.
4. Pulse shape fitting to a reference pulse shape after subtracting the baseline, so the amplitude, area and time-of-flight of the signal are extracted.
5. Reject the pulses lower than a certain threshold, to not include the signals generated by noise or rebounds. In this way, the size of the files is reduced substantially.

The extracted parameters of the signals, along with the information regarding the *event* (bunch number) in which it was generated, are stored in a ROOT file [139], in a process known as *event building*. The data at n_TOF is divided in *runs*, with a run corresponding to the irradiation of a given sample with about 4000 beam pulses, i.e. $\sim 2 \cdot 10^{16}$ protons on target. The ROOT files, once per run, are then ready to be analyzed to extract the capture yield, as described in the following Chapter.

5.4. Campaign beam distribution

The experimental campaign lasted for 43 days, with a total of $4.08 \cdot 10^{18}$ protons impinging on the spallation target. Because the number of protons was limited and many samples were included in the experimental campaign, a carefully planning of the beam time distribution was prepared, to try to optimize and maximize the statistics for each sample.

In addition, several calibration measurements were performed using radioactive sources (see Section 6.1.1). Some gain shift is usually observed in long C_6D_6 measurements, so to be able to correct this issue the detectors were calibrated once per week. Furthermore, to control the normalization and the reproducibility of the measurements, the gold sample was also measured in a weekly basis, to check that the BIF does not change during the campaign because of some displacement of the samples (see Section 6.4.1).

A summary of the samples and statistics is shown in Table 5.6.

Table 5.6: Summary of the number of protons registered by each sample during the whole n_TOF experimental campaign.

Sample	Protons (10^{17})
^{50}Cr -thin	5.49
^{50}Cr -thick	13.70
^{53}Cr -thin	6.52
^{53}Cr -thick	6.94
Dummy	3.05
Dummy (thick)	1.19
Empty	1.47
^{197}Au	1.07
$^{\text{nat}}\text{C}$	1.38
Total	40.81

6 | Capture yield at n_TOF-EAR1

After the data acquisition and Pulse Shape Analysis described in the previous Chapter, all the relevant information of the measurement is stored in ROOT files, one for each *run*. In this Chapter we will lay out the data sorting and analysis followed to obtain the capture yield.

6.1. Data reduction

The data reduction process starts with the calibration of the detectors and the flight path, then quality conditions are applied to the signals before the stability of all detection systems is verified throughout the full experimental campaign. The counting rates from the C₆D₆ detectors are corrected for dead-time effects before the Pulse Height Weighting Technique is applied. Then, the previously characterized backgrounds are subtracted and, last, the data are normalized using the Saturated Resonance Method, resulting in the aimed capture yields.

6.1.1. C₆D₆ energy and resolution calibration

A very accurate calibration of the C₆D₆ detectors, both in signal area to deposited energy and in energy resolution, is crucial for the implementation of the PHWT (see Section 6.3). During the experimental campaign, the detectors were calibrated once a week using the radioactive sources ¹³⁷Cs, ⁸⁸Y and ²⁴¹Am-⁹Be, which emit γ -rays of 0.662, 0.898 and 1.836, and 4.438 MeV, respectively. Due to the high chromium neutron separation energy combined with the hardness of the capture cascades, one high-energy point was needed in the calibrations, so the ¹⁹⁷Au cascade's end-point at 6.512 MeV was also included (see Section 6.3.1). Because of the low atomic number of the detector material, there are no photopeaks present in the measured spectra. Instead, the energy calibration is based on the Compton edges.

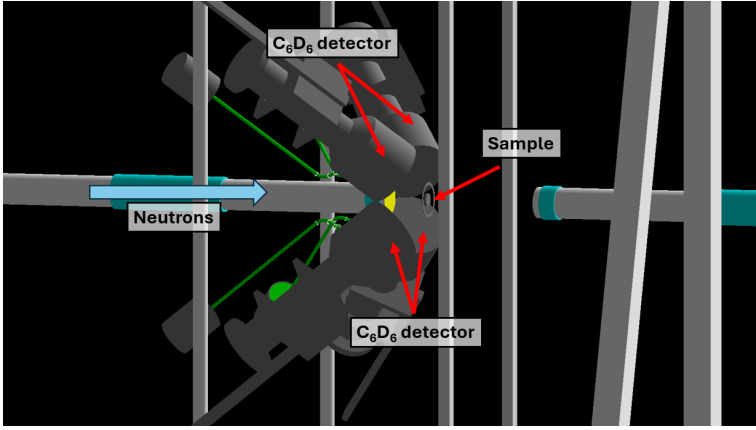


Figure 6.1: GEANT4 simulation of the EAR1 capture set-up. It includes a realistic geometrical model of the C_6D_6 detectors, as well as all the materials around them. As shown in the image, the distinct design of the sample used in each measurement is also included.

The experimental calibration was complemented by the simulated response of the detectors using a realistic GEANT4 model of the whole experimental set-up [140]. The model includes a very detailed version of the C_6D_6 detectors, as well as the elements that surround them as the beam line, the SAEX and the structure that holds the TAC assembly (see Fig. 6.1). In the simulations, the decay of the ^{137}Cs and ^{88}Y and the 4.438 MeV γ -ray from the ^{241}Am - 9Be source was used as the input, and the energy deposition was scored. Then, the energy spectra was broadened to reproduce the observed response of the detectors. The broadening was adjusted until it reproduced the measured spectra, and then fitted with an analytical expression. Some examples of the good agreement between measurement and broadened simulation are shown in Fig. 6.2.

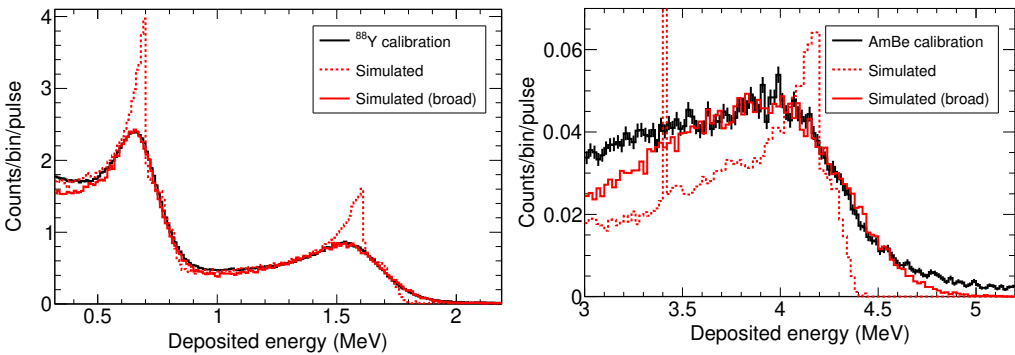


Figure 6.2: Comparison between calibration data and the simulated response of the detectors, including the resolution broadening, regarding ^{88}Y (left) and ^{241}Am - 9Be γ -ray (right) emission.

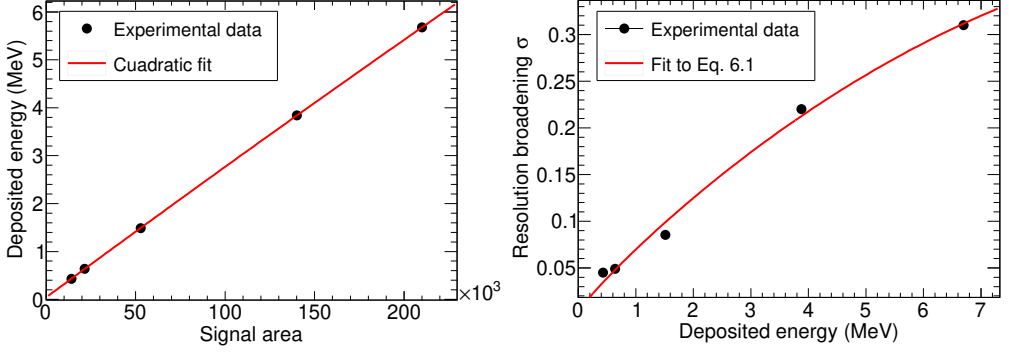


Figure 6.3: Left: Signal area to deposited energy calibration for detector #1 (first week). Right: Resolution broadening calibration for the same detector. The calibrations required a combination of radioactive sources measurements and simulations of the detection set-up.

The energy and resolution calibration of detector #1 are displayed in Fig. 6.3. A quadratic function reproduces the energy calibration better than a linear one (left panel). The resolution broadening as a function of the deposited energy $\sigma(E_\gamma)$ (right panel) was fitted using the expression

$$\sigma(E_\gamma) = a \cdot \sqrt{E_\gamma} + b \cdot E_\gamma + c \cdot E_\gamma^2. \quad (6.1)$$

The energy resolution is then obtained as

$$R(E_\gamma) = \frac{2.35 \sigma(E_\gamma)}{E_\gamma}. \quad (6.2)$$

In previous experiments, a significant gain shift has been observed affecting the C_6D_6 detectors, specially with high counting rates and/or radioactive samples. The stability of the detectors has thus been checked comparing all the calibrations performed along the 6 weeks long experimental campaign. A very good stability was observed, except for detector #4, where a very small gain shift can be appreciated (see Fig. 6.4). To minimize the effects of the gain shift, we divided the campaign into six periods, each one with its own energy calibration.

6.1.2. Detection threshold and signal rejection

Before proceeding to the yield determination, the data must be sorted to eliminate all possible signals that could correspond to noise. For this, a detection threshold of 150 keV in deposited energy is applied, since both the noise and most of the background

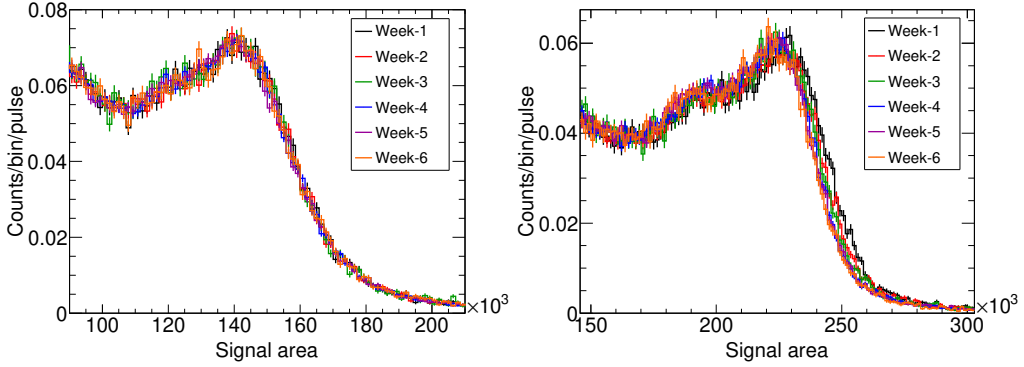


Figure 6.4: All calibrations performed with the ^{241}Am - ^9Be source during the experimental campaign, for detector #1 (left) and #4 (right). A small gain shift can be observed in the latter, so the data was divided into 6 sets, each one with its own calibration.

correspond to low amplitude signals (see left panel of Fig. 6.5). That value is a good balance between noise rejection and loss of efficiency. The threshold also eliminates signals rebounds that could have been identified as signals in the PSA process. This is an issue that is often observed in the C_6D_6 detectors on previous n_TOF measurements [141], and they usually appear around 600 ns after each signal, producing a low amplitude bump as can be also observed in the left panel of Fig. 6.5.

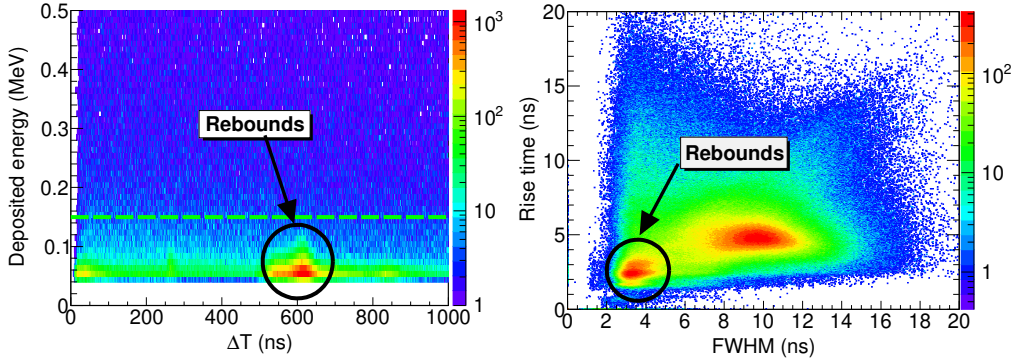


Figure 6.5: Left: Low amplitude signals coming from noise and rebounds, that get rejected under the 150 keV threshold (dashed green line). Right: When plotting the rise time versus the FWHM of the signals there is a separation between real signals and rebounds. The data displayed corresponds to the ^{197}Au measurement with detector #2.

An additional tool to discriminate these rebounds is the relation between the rise time and the FWHM, which is different for real signals and the rebounds (Fig. 6.5 right). As is shown in Fig. 6.6, after applying this cut and the threshold there are some visible

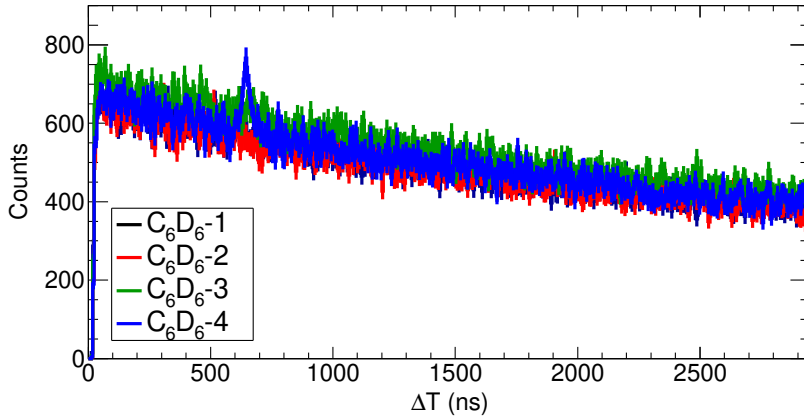


Figure 6.6: Time between consecutive signals for the ^{197}Au data. After applying the threshold of 150 keV, some rebounds are still visible in detector #4, but they will disappear in the PHWT procedure.

rebounds in detector #4, because their amplitude was higher and exceeded the threshold. However, when applying the Weighting Function (see Sect. 6.3) the low-energy γ -rays are strongly suppressed compared with capture signals, so their contribution to the final spectra becomes negligible.

6.1.3. Monitoring of the beam and detector performance

In order to obtain the capture yield, the counts registered by the C_6D_6 detectors for the different samples need to be normalized to the neutron flux, which is proportional to the proton beam current. Historically, the measurements at n_TOF are expressed relative to the nominal proton intensity per pulse, which is $7 \cdot 10^{12}$ protons/pulse. As mentioned in Section 5.3.4, the proton and neutron intensities per pulse are monitored by the BCT, the PKUP and SiMon (denoted as SILI from now on) in order to assess the stability of the measurement, and to identify and reject failed beam bunches. A dedicated analysis of the ratios of the three monitors against each other (BCT value, PKUP signal area and SILI counting rate) has been carried out for this matter, with the results displayed in Fig. 6.7.

The ratios between BCT and PKUP (top) clearly shows significant deviations from the $\sim 5 \cdot 10^6$ averaged value. The two blurred trends below this value at $\sim 4.4 \cdot 10^6$ and $\sim 3.7 \cdot 10^6$ correspond to Dedicated and Parasitic pulses for which the ratio value is anomalous. This can be better observed in Fig. 6.8. The anomalous behaviour is probably due to artifacts in the PKUP signals that made the PSA less reliable, thus hindering the normalization process. In addition, during the second week of experiment,

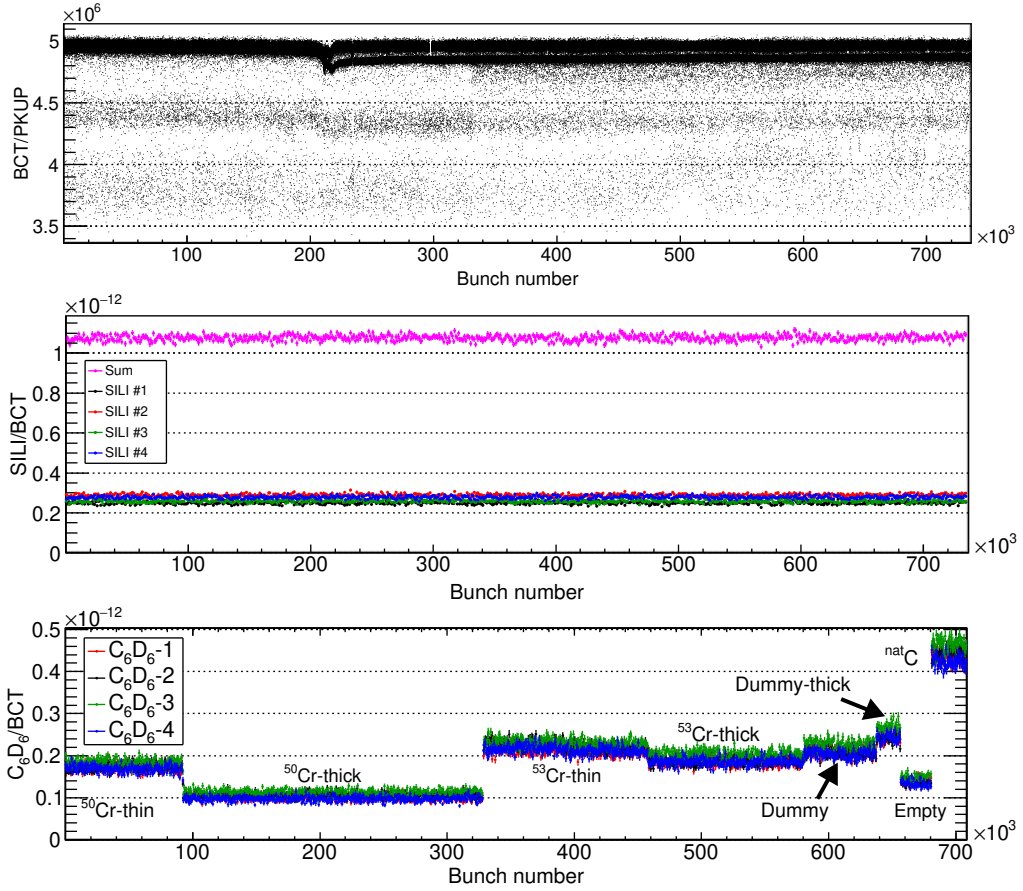


Figure 6.7: Top: Ratio between the BCT value and the PKUP signal area. Middle: Ratio between SiMon counting rate and the BCT value. Bottom: Counting rate of the C_6D_6 detectors normalized to the BCT nominal value. The counting rate for each sample corresponds to those in the neutron energy ranges of interest: 1-10 keV for the thin samples, 10-100 keV for the thick samples and 1 eV to 100 keV for the auxiliary measurements.

the PKUP signals changed slightly, resulting in different BCT to PKUP ratios, and therefore the PKUP was not considered as a reliable proton beam monitor.

The ratio between the SILI counting rate and the BCT value is shown in the middle panel of Fig. 6.7, where the data have been grouped every 1000 bunches to reduce statistical fluctuations. The counting rate corresponds to the integral of the triton peak (which, unlike the alphas, is well defined and separated from the noise), for neutron energies between 1 eV and 100 keV. The ratio values remain constant along the experimental campaign with a standard deviation of 2.5% (see Fig. 6.9). This serves to estimate the uncertainty in the normalization between different runs, samples, etc, thus contributing

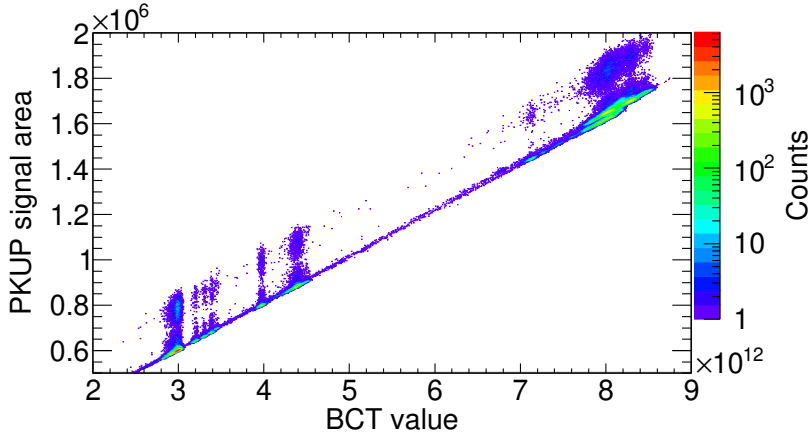


Figure 6.8: Relation between the BCT and the PKUP monitors. For an important fraction of pulses this relation changes, making the PKUP signal area not a reliable monitor.

with 2.5% to the overall uncertainty of the capture yields.

The last check to discard an anomalous performance during the experiment is the comparison of the counting rate of the C_6D_6 per bunch with the BCT nominal value, shown in the bottom panel of Fig. 6.7. In this, the C_6D_6 data correspond to the neutron energy range of interest for each sample: 1 to 10 keV for the thin samples, 10 to 100 keV for the thick ones, and 1 eV to 100 keV for all the auxiliary samples. After discarding bunches with very low proton intensity, a very nice behaviour of the detectors during the whole campaign is observed.

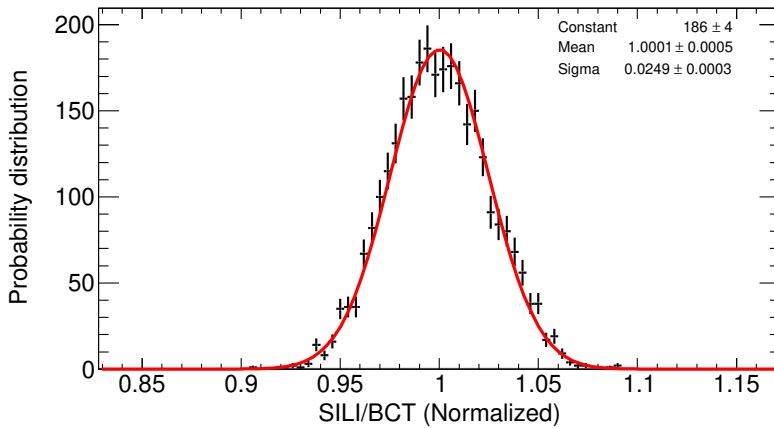


Figure 6.9: Probability distribution of the SILI/BCT ratio normalized to the average value. It serves to estimate the uncertainty associated to the normalization of the data to the number of neutrons.

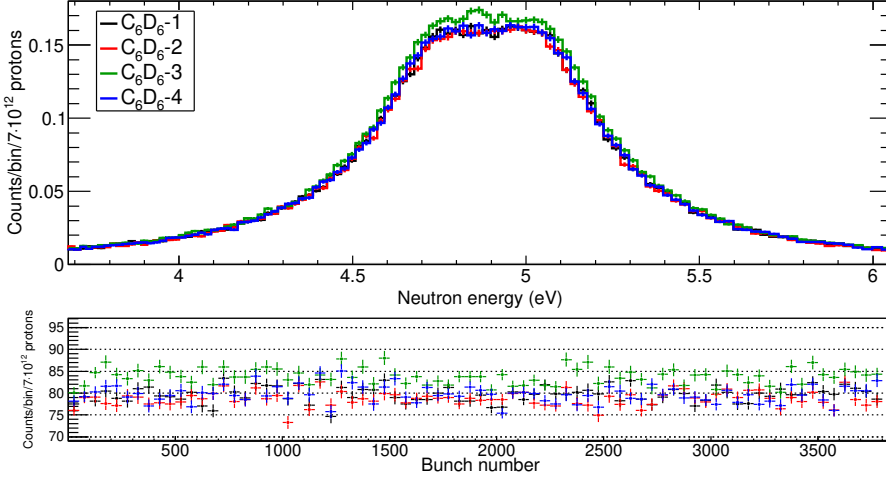


Figure 6.10: Counting rate as a function of the neutron energy (top) and per bunch (bottom) for the ^{197}Au saturated resonance, showing that C_6D_6 #3 was a little bit closer to the sample than the other detectors.

It should be noted that the counting rate of detector #3 was higher than the others. This is specially clear in the ^{197}Au saturated resonance data (see Fig. 6.10). The same behaviour was observed in the previous measurement performed at EAR1 during the 2022 summer campaign, in which the exact same experimental configuration was used. From simulations, it was found that this detector was 4 mm closer to the sample than all others. This is not an issue, as it is taken into account later on in the determination of the efficiency by means of the Weighting Functions to apply the PHWT (see Section 6.3).

6.1.4. Time-of-flight to neutron energy calibration

As mentioned in previous chapters, the time-of-flight technique is based in the determination of the neutron energy from the time it takes them to travel from the target to the sample, by means of a very simple non-relativistic relation:

$$E_n = \frac{1}{2}m_n v^2 = K^2 \frac{L^2}{t^2}, \quad (6.3)$$

where L is the flight path length, t the time-of-flight and $K = 72.2977 \cdot 10^3 \frac{\text{ns}\sqrt{\text{eV}}}{\text{m}}$. At n_TOF, the time-of-flight is determined as follows:

$$E_n = K^2 \frac{L^2}{(t - t_\gamma + L/c + t_{\text{off}})^2}, \quad (6.4)$$

where t is the time stamp of the signal as obtained by the PSA, t_γ the time of the γ -flash signal, t_{off} is related to the RF and c the speed of light, so L/c corresponds to the time required for the photons and relativistic particles that make the γ -flash to travel the flight path length. At n_TOF, given the size of the lead spallation target and the subsequent use of a moderator, the point of origin of the neutrons is not well defined, so neither is L . The usual way to determine the flight path is then to use well known resonances as reference.

However, as described in Section 5.2.3, because of the Resolution Function (RF) of the facility the relation between time-of-flight or flight path and energy is not univocal, but rather a distribution. This distribution makes the resonances broader and appearing shifted from their nominal energy. There are several methods to introduce this shift in the neutron energy determination [127]. A simple but effective strategy, is to introduce a time offset t_{off} to the time-of-flight in Eq. 6.4. This can be determined together with the effective flight path length L_0 from the comparison of the observed and nominal energy of well known resonances, in this case from ^{197}Au .

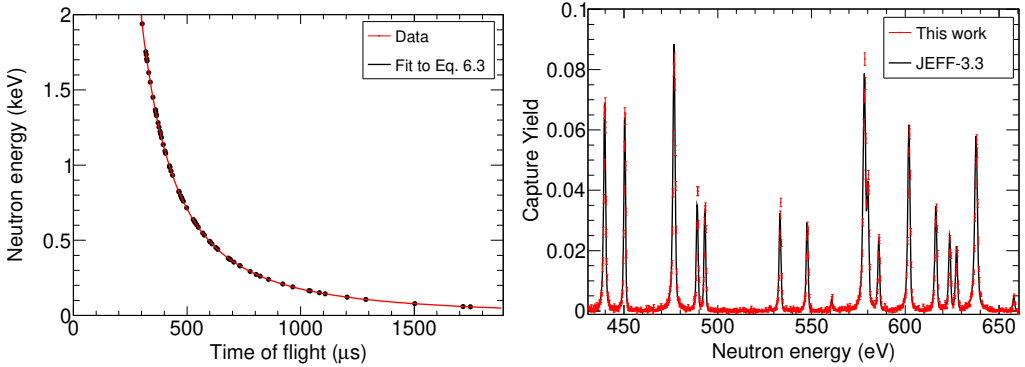


Figure 6.11: Left: Fit of the L and t_{off} parameters of the time-of-flight to neutron energy calibration. The points correspond to resonances of ^{197}Au . Right: Illustration of the good agreement (within 0.1%) of the resonance energies between this work and the reference JEFF-3.3 cross section.

First, a preliminary value of L_0 was estimated by adjusting the position of the 4.9 eV resonance. Then, the neutron energy spectrum obtained with this flight path was analyzed with SAMMY (see Chapter 7) including the numerical RF of n_TOF, leaving only the energy of the resonances as free parameters. Then, the nominal energies (from the JEFF-3.3 evaluation) are represented against the time-of-flight reconstructed from the energies given by SAMMY, (left panel in Fig. 6.11). A simultaneous fit of L_0 and t_{off} to this curve returns $L_0 = 183.94(4)$ m and $t_{off} = -17.57(4)$ ns. The right panel of Fig. 6.11 shows how nicely (within 0.1%) the energy of ^{197}Au resonances is reproduced

when using this calibration strategy.

6.1.5. Dead-time correction

Dead time is an issue that every detector has to deal with. It is a situation that happens when two signals arrive too close in time, producing pile-up. When this happens, the PSA has troubles distinguishing between the two signals. Although in our case the efficiency of the C_6D_6 is very low and there were never large counting rates (see top panels of Fig. 6.12), the dead time has to be corrected anyway to not underestimate the yield of the strongest resonances.

The detection set-up with this PSA suffers from the so-called paralyzable dead time, in which the second pulse is not identified but extends the dead time by overlapping the previous one. It is rather difficult to estimate the correction in this case, but it can be proven that, for small counting rates, the correction factor is very similar that for non-paralyzable systems. In the case of non-paralyzable dead time, the system cannot detect a signal during the dead time τ after the first one, and the true counting rate C is

$$C = C_d + \frac{C}{T}C_d\tau, \quad (6.5)$$

with C_d the measured counting rate during a time interval T . Therefore, using the dead time correction factor F_τ , the true counting rate C can be calculated from the detected one C_d as

$$C = \frac{C_d}{1 - \tau \frac{C_d}{T}} = F_\tau C_d. \quad (6.6)$$

According to the minimum ΔT observed between consecutive signals (see Fig. 6.6), the dead time is estimated to be $\tau=30$ ns. As shown at the bottom panels of Fig. 6.12, the correction is less than 1% in the range of interest, even for all the resonances of gold, being just 1.2% for the highest counting rate one at ~ 60 eV in ^{197}Au . The small value of the correction factor makes the uncertainty associated to dead-time negligible.

6.2. Background determination

All possible sources of background must be determined and subtracted from the chromium measurements to correctly determine the capture yield. As mentioned in Section 5.3.3, a series of auxiliary measurements were performed to this purpose. The background in a typical n_TOF measurement is the combination several contributions:

- The ambient background due to the radioactivity from the walls and the elements inside the experimental area. This background is assessed through measurements

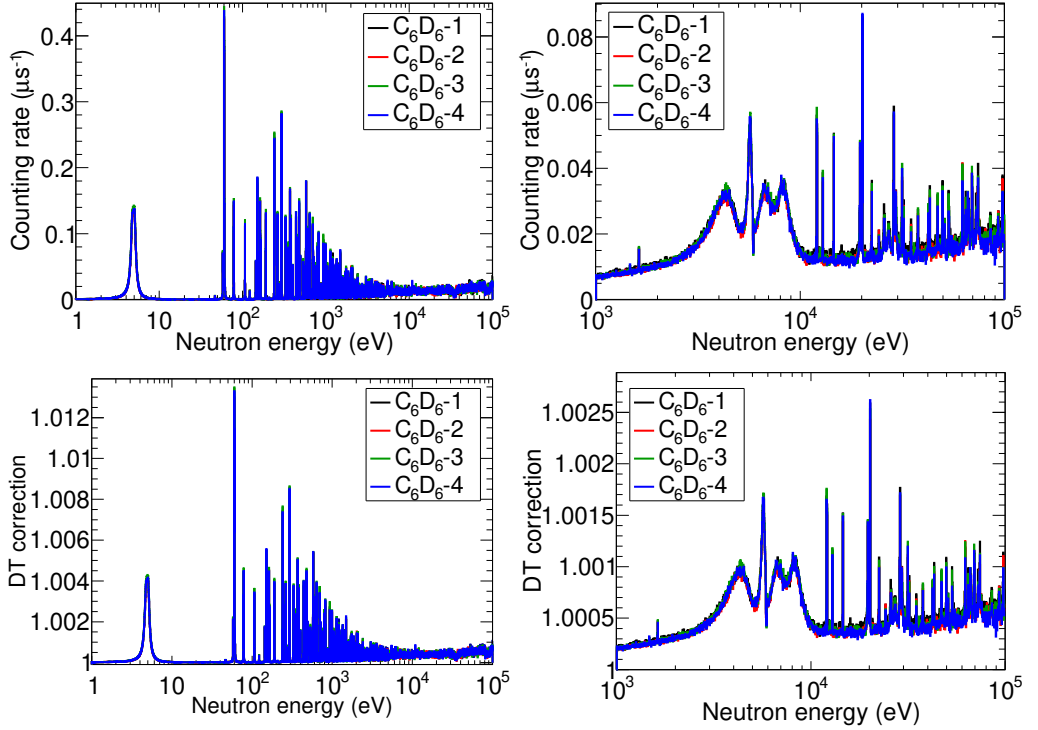


Figure 6.12: Top: Counting rates for dedicated pulses corresponding ^{197}Au (left) and ^{53}Cr -thick (right). Bottom: Dead time correction obtained from such counting rates.

without beam ("Beam-off") and is usually negligible compared to others.

- If the sample is radioactive, its intrinsic activity can produce a considerable γ -ray background, but this is not the case for ^{50}Cr and ^{53}Cr .
- In-beam γ -rays (see Section 5.2.4) can be Compton-scattered in the sample and then detected. To assess this background, a sample with a very high Z , like $^{\text{nat}}\text{Pb}$, is employed because it features a large γ -ray scattering-to-neutron scattering or capture ratio. However, given the low Z of chromium, this background is negligible in our case.
- Neutron scattering, defined as the background produced by neutrons scattered in the sample that are then captured somewhere else, thus producing a γ -ray background. There are several methods to estimate its contribution, like measuring the sample with neutron filters inserted on the beam line or the measurement of a low Z and low capture cross section sample, like $^{\text{nat}}\text{C}$, acting as a pure neutron scatterer.

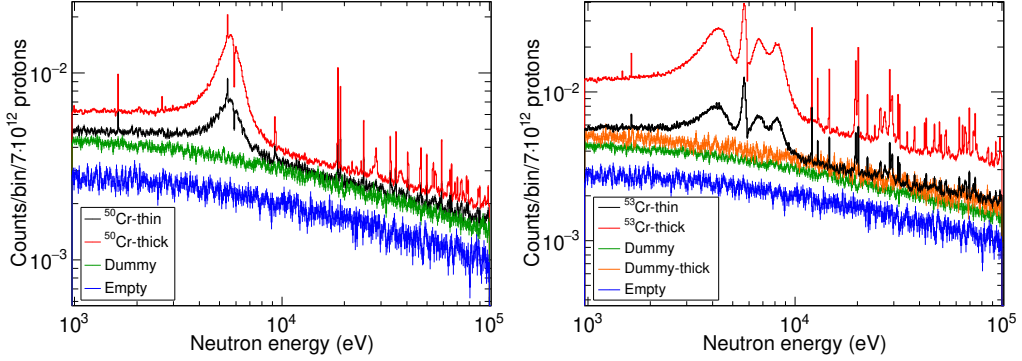


Figure 6.13: Counting rate per nominal proton pulse regarding ^{50}Cr (left) and ^{53}Cr (right), along with the Dummy and Empty measurements. They serve as an estimation of the background produced outside the samples.

- Sample holder related background, due to γ -scattering, neutron scattering or neutron capture in the PEEK container, in our case. Since it is a plastic ($\text{C}_{19}\text{H}_{12}\text{O}_3$), it can be assumed insensitive to γ -scattering. In the keV region where neutron scattering dominates for all its components, is also practically insensitive to capture.
- Any other neutron background contribution is determined by measuring without any sample, what is known as Empty sample. This contribution is inherent to any of the measurements mentioned above.

In Fig. 6.13 are shown the counting rates of the ^{50}Cr (left) and ^{53}Cr (right), the Dummies and the Empty. The Dummy counting rate for the ^{53}Cr -thick sample corresponds to a PEEK capsule slightly larger than the one used for the rest of the Cr samples. Obviously the background measurements have limited statistics and thus feature fluctuations that would be better to avoid. To minimize them, the counting rate of both Dummy measurements are parameterized (see Fig. 6.14) using the following form:

$$B(E_n) = a_0 + \sum_{i=1}^3 b_i (1 - e^{-c_i E_n}) e^{-d_i E_n}, \quad (6.7)$$

so, the parameterized function is subtracted to the measured chromium spectra instead of the measured Dummy background.

A perfect estimation of the background should be able to reproduce the counting rate in the valleys between resonances. However, as can be seen in Fig. 6.13, this is not the case when considering only the Dummy contribution, specially in the thick samples cases. This means that the background caused by neutron-scattering in chromium is not

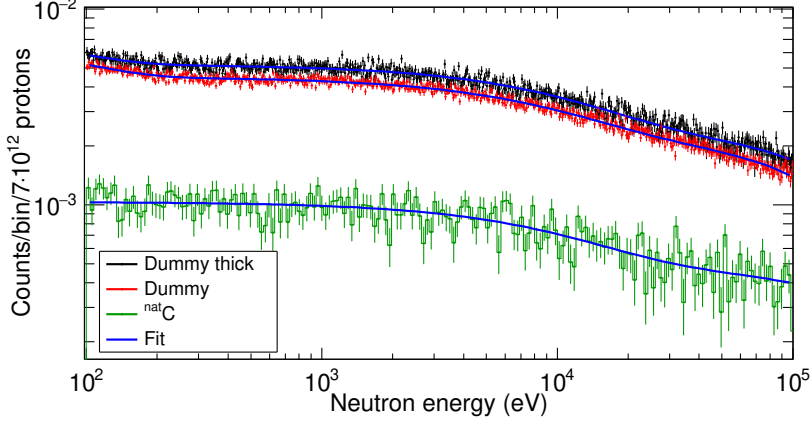


Figure 6.14: Fitting performed to the Dummy (red), Dummy-thick (black) and $^{\text{nat}}\text{C}$ (green) counting rate to minimize the fluctuations in the background subtraction procedure. The $^{\text{nat}}\text{C}$ fitting has to be scaled to the mass of each chromium sample.

negligible. As mentioned earlier, this background is estimated through a $^{\text{nat}}\text{C}$ sample. To take into account the different physical characteristics and cross sections of the chromium and carbon samples, a scaling factor F_n is applied to the measured carbon counting rate, defined as

$$F_n = \left\langle \frac{\sigma_{\text{el,Cr}}}{\sigma_{\text{el,C}}} \right\rangle \frac{n_{\text{at,Cr}}}{n_{\text{at,C}}}, \quad (6.8)$$

with $\langle \sigma_{\text{el,Cr}}/\sigma_{\text{el,C}} \rangle$ the average ratio between Cr and $^{\text{nat}}\text{C}$ elastic cross sections in the whole energy range, and n_{at} the atomic thicknesses. The F_n value for every Cr sample is given in Table 6.1. As we did with the Dummy, the $^{\text{nat}}\text{C}$ was also fitted using Eq. 6.7, but only up to $i = 2$ because it was enough to reproduce the data, as shown in Fig. 6.14. These F_n values are only an approximation because they do not include any dependence with neutron energy, but it is far from straightforward to calculate such behaviour (see Ref. [142]), and our approximation is later shown to be sufficiently accurate in this particular case.

Table 6.1: Scaling factors F_n to estimate the neutron-scattering background of the Cr using a $^{\text{nat}}\text{C}$ sample.

Sample	F_n
^{50}Cr -thin	0.25
^{50}Cr -thick	0.72
^{53}Cr -thin	0.59
^{53}Cr -thick	2.91

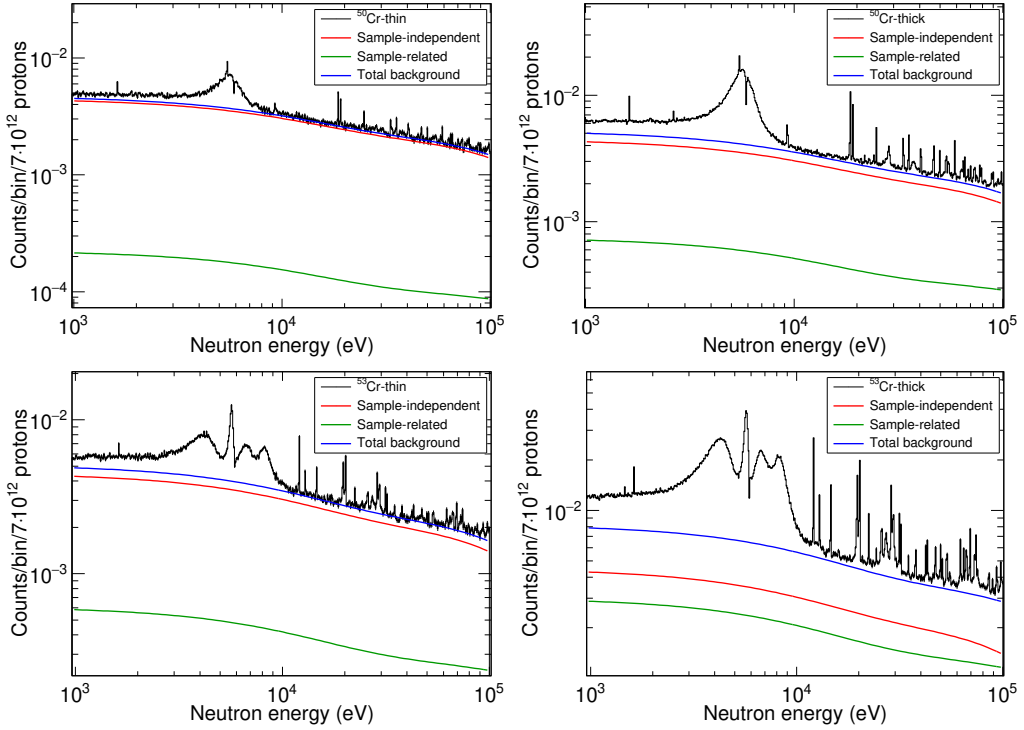


Figure 6.15: Sources of background considered for the ^{50}Cr (top) and ^{53}Cr (bottom), fitted to some analytical function as described in the text.

The final outcome of the background determination is displayed in Fig. 6.15. When adding the neutron-scattering contribution the total background is closer to the chromium measurements in the valleys between resonances, as it should. However, the chromium counting rate is significantly above the total background at low neutron energies, in the tail of the strong and broad s-wave resonances of the few keV region. The reason behind this is the mentioned missing neutron energy-dependence in F_n . In previous n_TOF measurements, this was determined by simulating the contribution of the scattered neutrons as a function of the time-of-flight in a realistic GEANT4 model of EAR1 [142]. In our case, the reproduced background is able to reproduce very well the counting rates between resonances, except for the low-energy part below the main resonances in the thicker samples. But since they are not used to analyze the low energy region, the approach followed herein is considered a good approximation. This is discussed further in Chapter 7 in the context of the capture yield analysis with the code SAMMY.

6.3. Implementation of the PHWT

The implementation of the Total Energy Detection (TED) technique requires, as discussed in Section 5.3.1, the efficiency of the detectors to be very low and proportional to the energy of the γ -ray detected. As the C_6D_6 detectors do not fulfill the second requirement, the strategy is to apply a mathematical treatment to the signals so the response features the desired proportionality to the γ -ray energy. This is the basis of the Pulse Height Weighting Technique (PHWT) proposed by Macklin in 1967 [70].

When a γ -ray with energy E_γ enters the detector, the efficiency for detecting it $\varepsilon(E_\gamma)$ is given by

$$\varepsilon(E_\gamma) = \int_0^\infty R(E_d, E_\gamma) dE_d, \quad (6.9)$$

with $R(E_d, E_\gamma)$ the response of the detector as that γ -ray deposits an energy E_d . When working with a discrete distribution of energies, Eq. 6.9 transforms to

$$\varepsilon(E_{\gamma i}) = \sum_j R_{ij} E_j. \quad (6.10)$$

The goal is to apply a weight to each signal by using the so-called Weighting Functions (WF), so that the resulting weighted efficiencies $\varepsilon^w(E_{\gamma i})$ are

$$\varepsilon^w(E_{\gamma i}) = \sum_j W_j R_{ij} = E_{\gamma i}. \quad (6.11)$$

Usually, the WF are defined as a function of the deposited energy E_d with the form

$$W_j = \sum_k a_k (E_j)^k. \quad (6.12)$$

In our case, a value of $k = 5$ is enough to obtain satisfactory WF.

The input to find the WF is the response of the detection system to mono-energetic γ -rays. Because it is not possible to produce a sufficient number of mono-energetic γ -rays in the laboratory, the response is obtained by Monte Carlo simulations. The same GEANT4 model of EAR1 used to calibrate the detectors (see Section 6.1.1) is used to obtain the γ -rays response, taking into account that each sample needs its own WF (gold and four Cr samples, in our case), because their geometry is different and thus the transmission of the γ -rays that need to escape the sample where they are produced.

A large number (10^6 events for each energy) of 56 mono-energetic γ -rays between 50 keV and 10 MeV have been simulated, broadening the deposited energy spectra according to Eq. 6.1. The simulated response of the detectors R_{ij} to these γ -rays is shown in Fig.

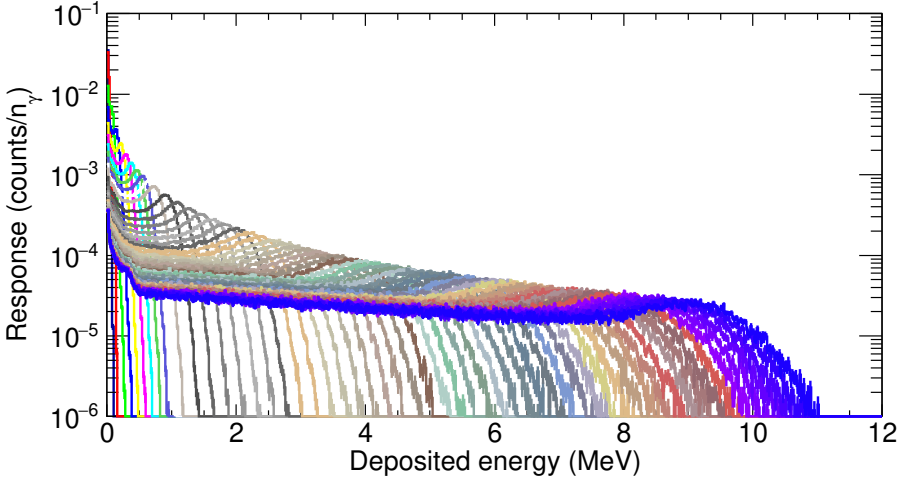


Figure 6.16: Simulated response of detector #1 to a series of mono-energetic γ -rays between 0.5 and 10 MeV, after the resolution broadening is applied.

6.16. To find the parameters a_k of the WF, the following χ^2 must be minimized:

$$\chi^2 = \sum_i \left(E_{\gamma i} - \sum_{j=E_{min}}^{E_{max}} W_j R_{ij} \right)^2, \quad (6.13)$$

with the minimization performed with the Minuit toolkit of ROOT.

One issue to be considered is that the actual measurements feature a detection threshold of 150 keV, which needs to be considered when applying the PHWT. There are two ways of handling this:

- Not introducing a minimum energy when calculating the WF ($E_{min} = E_{thr} = 0$), and then introducing a threshold correction factor to the yield calculation in order to compensate for the signals lost below the detection threshold.
- Applying the detection threshold to the simulation, so the correction is introduced implicitly in the WF. In this case, some correction is still needed for those γ -rays with energy lower than $E_{\gamma} < E_{thr}$.

We have performed a systematic study to investigate each methodology and chose the one that offers the best results. For that reason, we have obtained the WF for the gold sample introducing different thresholds. If the WF is perfectly calculated, then the following relation is true:

$$Q = \frac{\sum_j W_j R_{ij}}{E_{\gamma i}} = 1. \quad (6.14)$$

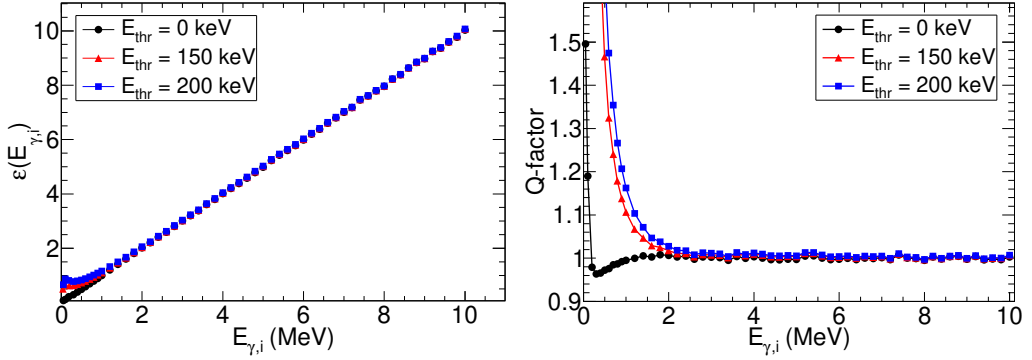


Figure 6.17: Left: Weighted efficiency obtained by choosing different values of E_{thr} . Right: The quality of the WF for each case given by the Q-factor defined in Eq. 6.14

In other words, how close this Q-factor is to unity offers information about the goodness of the WF. On the left panel of Fig. 6.17, the weighted efficiency obtained with different E_{thr} values are shown, with the corresponding Q-factor displayed on the right panel. When E_{thr} increases the WF are less capable of obtaining a good weighted efficiency. Not introducing a threshold seems to be more accurate option to obtain the WF, so is the option chosen in this work.

Another effect to consider in the simulations is the neutron self-shielding. If the sample is too thick or it features a very high capture cross section then the captures do not occur uniformly inside the sample, but more in an exponential distribution decreasing along the beam direction within the sample. Therefore, the capture cascades are emitted following the same distribution in the sample, which has to be included in the simulations. This effect is negligible when the product of the area density and the total cross section $n_{at}\sigma$ is very low, which is the case for all chromium samples in all the energy range and thus an homogeneous γ -ray emission inside the sample can be assumed. On the other hand, for the ^{197}Au sample at the 4.9 eV saturated resonance all neutrons are absorbed in the first ~ 30 μm , which has to be and has been considered in the corresponding simulations.

The WF of ^{197}Au and ^{50}Cr -thin samples are shown in Fig. 6.18 as an example. The differences between them, although small, show that indeed the WF are sample and detector dependent (detector #3 is around 4 millimeters closer to the sample than the others).

A last note on the application of the PHWT, is that to obtain a capture yield from a weighted spectrum, the background that will be subtracted also had to be weighted. The background spectrum for each chromium and gold sample is thus weighted by the same WF than the actual sample.

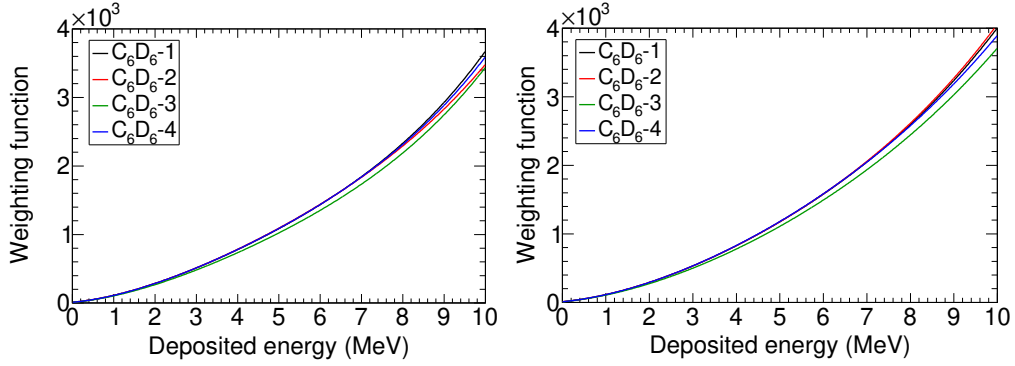


Figure 6.18: Weighting functions corresponding to the ^{197}Au (left) and ^{50}Cr -thin (right) samples.

6.3.1. Capture cascades simulation

In order to correctly apply the PHWT we need to simulate very accurately the capture cascades, because some additional correction factors need to be considered:

- f_{thr} , to take into account all the non-detected γ -rays because of the detection threshold.
- f_{mc} , as there is some possibility to detect more than one γ -ray per cascade (multiple-counting), although the detection efficiency is small.
- f_{ic} , because of the possible emission of electrons instead of γ -rays coming from internal conversion (IC).

Since all effects have been simulated at the same time, the final correction factor F_{WF} obtained is equivalent to the product of f_{thr} , f_{mc} and f_{ic} .

The correction factors F_{WF} have been calculated as well by means of Monte Carlo simulations, using as input the capture cascades. These cascades have been simulated with the cascade generator software NuDEX [143], serving as input to the GEANT4 simulation of our set-up. To produce a capture cascade, NuDEX takes the level scheme and the branching ratios from ENSDF [107] corresponding to the compound nucleus and simulates the de-excitation process starting at the neutron capture level. The missing levels at high excitation energy, because they are not present in the data bases or because they are in the continuous and cannot be accessed experimentally, are randomly generated according to statistical models of the nuclear level density. A complete set of levels and branching ratios is called a *realization*. Once the realization is fixed there is the possibility to generate the cascade beginning at different initial levels, recreating a capture produced in different resonances or neutron energies. This is called a *subrealization*.

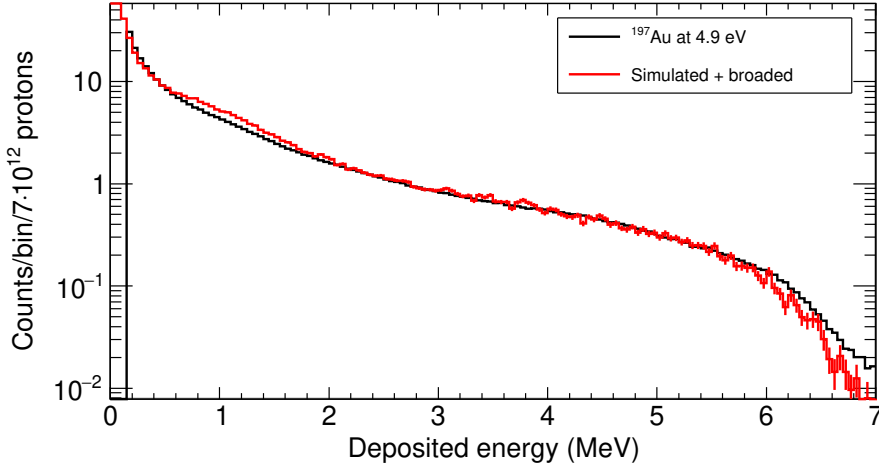


Figure 6.19: Measured ^{197}Au capture cascade in the saturated resonance at 4.9 eV (black) and simulated cascaded obtained with NuDEX and GEANT4 (red).

The simulated ^{198}Au cascade is displayed in Fig. 6.19 in comparison with the measured cascades from the saturated resonance at 4.9 eV. Since the simulation reproduces adequately the experimental cascades, the latter can be used in the estimation of the correction factor F_{WF} . However, as explained below, this is not true for the chromium case. For the matter of the discussion presented herein, the chromium cascades corresponding to a thermal neutron capture have been simulated and implemented in the following.

To find F_{WF} , the response to N_c simulated cascades considering a detection threshold of 150 keV is weighted to obtain the weighted efficiency $\varepsilon_{c,exp}^w$. The deviation of this value to the expected one, which is the energy of the cascades E_c (see Eq. 6.11), gives the correction factor:

$$F_{WF} = \frac{\varepsilon_{c,exp}^w}{\varepsilon_c^w} = \frac{\sum_i^{N_c} \sum_j W_j R_{ij}^c}{N_c E_c} \quad (6.15)$$

The values of F_{WF} obtained for each sample and detector are summarized in Table 6.2. The uncertainty of these factors has two contributions: statistical from the total number of cascades simulated (negligible in this case) and the systematic uncertainty of applying the WF. To estimate the latter, we have used the Q-factor of Eq. 6.14, but weighting the full simulated response of the cascades, without any detection threshold. The deviation of the Q-factor from the unity in this case reflects the intrinsic uncertainties of the weighting process. This has been performed for several ^{197}Au and chromium capture cascades, obtaining an average deviation of 1.7%, so we have consider this value as the systematic uncertainty of the WF. Observing the F_{WF} values from Table 6.2, the

Table 6.2: Correction factors necessary to apply to the weighted spectra to obtain the capture yield. The systematic uncertainty of the individual values is 1.7% (see text for details).

Sample	F_{WF}				Average
	C ₆ D ₆ #1	C ₆ D ₆ #2	C ₆ D ₆ #3	C ₆ D ₆ #4	
¹⁹⁷ Au	0.971	0.958	0.963	0.972	0.966(7)
⁵⁰ Cr-thin	0.981	0.978	0.980	0.979	0.979(1)
⁵⁰ Cr-thick	0.971	0.998	0.972	0.964	0.976(15)
⁵³ Cr-thin	0.973	0.969	0.985	0.980	0.977(7)
⁵³ Cr-thick	0.985	0.995	0.994	1.007	0.995(9)

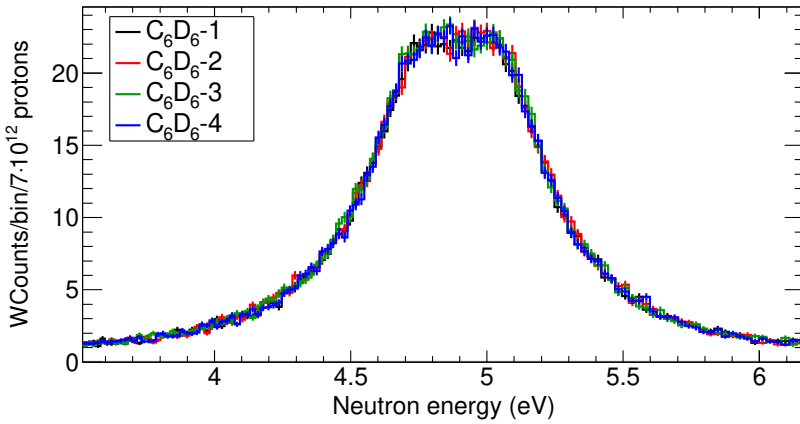


Figure 6.20: Weighted spectra of the ¹⁹⁷Au saturated resonance, illustrating the agreement between all detectors regardless of their distance to the sample.

correction is slightly larger for ¹⁹⁷Au, which makes sense because the detection threshold affects more its softer cascade. Then, the values for all chromium samples are similar, as they should, except for ⁵³Cr-thick, which is 4 to 10 times thicker than the others.

The weighted spectra for the ¹⁹⁷Au saturated resonance are displayed in Fig. 6.20. Contrary to what was shown in Fig. 6.10, the data from detector #3 data is now in agreement with all others because the different sample-to-detector distances are considered in the weighting procedure. This result reinforces the validity of the calculated WF.

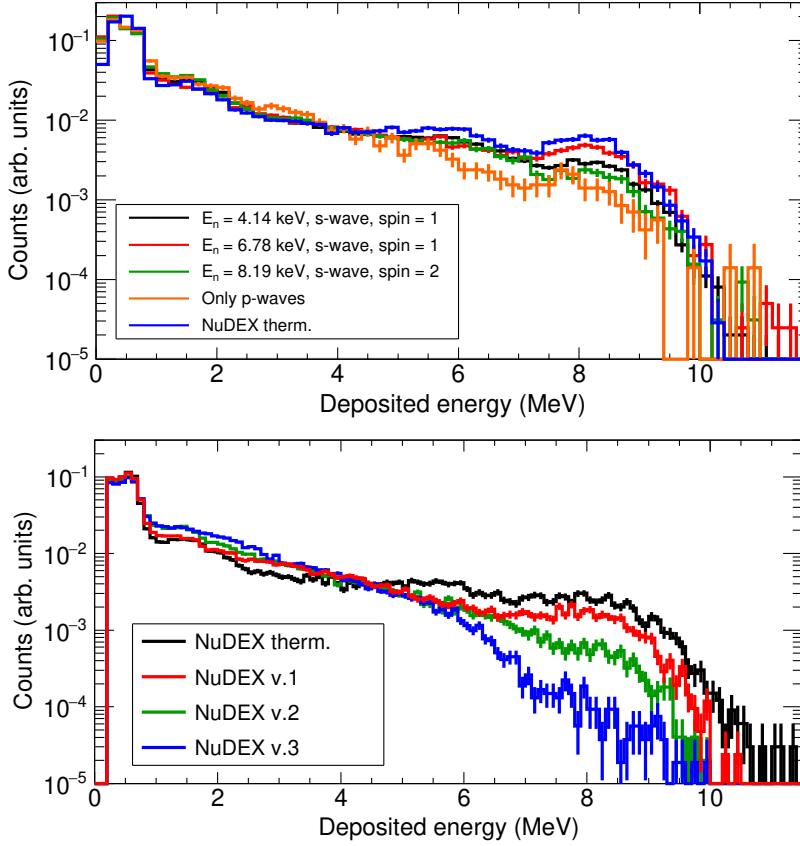


Figure 6.21: Response of the C_6D_6 detectors to ^{53}Cr capture cascades from different resonances. Top: Measured response to the three main s-waves resonances and a sum of the cascades of multiple p-waves resonances, along with a simulated thermal cascade generated with NuDEX. Bottom: simulation of the the same thermal cascade and 3 additional NuDEX subrealizations, i.e. resonances.

6.3.2. Chromium cascades

For heavy nuclei, which are the majority in n_TOF measurements, the capture cascades are simulated generally as capture of thermal neutrons. This is because the level density is very large in these and the cascade can thus follow a great number of de-excitation patterns, being therefore no difference between the cascades from different resonances. This is the case for the ^{198}Au cascades, but it is not necessary true for a light isotope like chromium. When the number of levels is relative low, the cascade pattern may change significantly depending on the energy and spin of the resonance it comes from, because is strongly determined by the *primary transitions*. For this reason, it is not straightforward

the implementation of the PHWT in the chromium cascades, and the assumption that the cascade is going to be more or less the same no matter the resonance is not true.

To confirm what was expected according to the theory, the measured response of the detectors to capture cascades of ^{53}Cr (thick sample) from different resonances have been extracted. We have chosen this sample because is the one with better statistics; in addition, ^{53}Cr has spin $I^\Pi = 3/2^-$, so it features s-waves with two different spins $J = 1, 2$ (see Chapter 2) that should results in slightly different cascade patterns.

The top panel of Fig. 6.21 shows the background (Dummy) subtracted detector response to the cascades from the three main s-waves resonances and also to the sum of several p-waves (the statistics from individual ones are insufficient). We have also included the simulated cascades for thermal neutrons from NuDEX for comparison. It is obvious that there are sizeable differences between them all, specially in the high energy part of the spectrum, which is dominated by the primary de-excitations.

In principle, the change in the de-excitation pattern depending on the resonance of origin is caused by changes in the primary transition probabilities [144], affecting more those nuclei with a smaller number of levels. To prove that the differences observed arise from this fact, we have produced and simulated several NuDEX subrealizations of the same realization for ^{53}Cr . This is shown in the bottom panel of Fig. 6.21, where the differences observed are indeed of the order of those measured.

The confirmed fact that the decay pattern changes from one resonance to another makes it necessary to estimate how much it affects the correction factor F_{WF} . In Mendoza et al. [145] a dedicated study was performed in relation to this. The calculations showed that indeed this effect results in a systematic uncertainty in the PHWT technique that is larger for light nuclei. In the case of chromium, and considering a detection threshold of only 150 keV, a value of 0.7% for ^{50}Cr and 0.9% ^{53}Cr is estimated in the Ref. [145]. For this work, a slightly more conservative 1% uncertainty is considered.

6.4. Capture yield determination

After applying the PHWT, we have obtained a weighted capture spectra which efficiency is the excitation energy of the Compound Nucleus formed after a neutron capture, so the capture yield is calculated as:

$$Y(E_n) = F_{WF} \frac{C_w(E_n) - B_w(E_n)}{F_{BIF} \left(S_n + \frac{A}{A+1} E_n \right) \Phi(E_n)}, \quad (6.16)$$

where C_w and B_w are the total and background weighted counts, $\Phi(E_n)$ the evaluated neutron flux, F_{WF} is given in Table 6.2 for each case and F_{BIF} is the correction factor due to the Beam Interception Factor (BIF). As introduced in Section 5.2.2, the sample is smaller than the neutron beam, so it only sees a fraction F_{BIF} of the neutrons, which is calculated by means of the Saturated Resonance Method (SRM).

6.4.1. Saturated Resonance Method (SRM)

The Saturated Resonance Method (SRM) [133] is based on the measurement of a sample "thick" enough (see below) to make the yield saturate for a particular resonance because all neutrons in the beam have interacted with the sample. This happens if the condition $n_{at}\sigma \gg 1$ is fulfilled, so the yield is (see Eq. 2.22)

$$Y(E) = (1 - e^{-n\sigma_t}) \frac{\sigma_\gamma}{\sigma_t} \cong \frac{\sigma_\gamma}{\sigma_t}. \quad (6.17)$$

Furthermore, if the resonance is dominated by capture, then $\sigma_t \approx \sigma_\gamma$ and thus the yield becomes not only a constant but also close to unity, resulting in the desired saturated resonance. The yield in the saturation is almost independent of the exact value of the capture or total cross section, so the unavoidable uncertainty in the cross section of the employed sample does not propagate to the saturation value.

^{197}Au is the perfect option for this matter, since the capture cross section at the 4.9 eV resonance is $\sigma_\gamma = 2.7 \cdot 10^4$ b and thus saturation is achieved with a sample of just tens of microns. Furthermore, this resonance features $\Gamma_\gamma = 127$ meV and $\Gamma_n = 14.96$ meV according to the JEFF-3.3 library, which means that capture dominates by nearly a factor 10 over neutron scattering. In addition, gold is a mono-isotopic and stable element, and it is very easy to manufacture a high purity sample of the desired shape.

If we measure a gold sample with enough thickness and of the same diameter that the sample of interest, the yield will be constant at the top of the 4.9 eV resonance. Ideally it will be close to 1, but as mentioned before, only a fraction of the neutron flux is seen by the samples, and thus the ratio of the observed saturation value with the expected value provides the value of F_{BIF} . The expected saturation value can be accurately calculated with SAMMY.

In order to implement this, following the steps described in this Chapter, the capture yield of gold has been calculated from the weighted spectrum to which the "Empty" background has been subtracted. This yield is shown in the left panel of Fig. 6.22, along with the expected value provided by SAMMY and the fitting of the yield, illustrating that even if the saturated region is not flat, SAMMY is able to reproduce it perfectly.

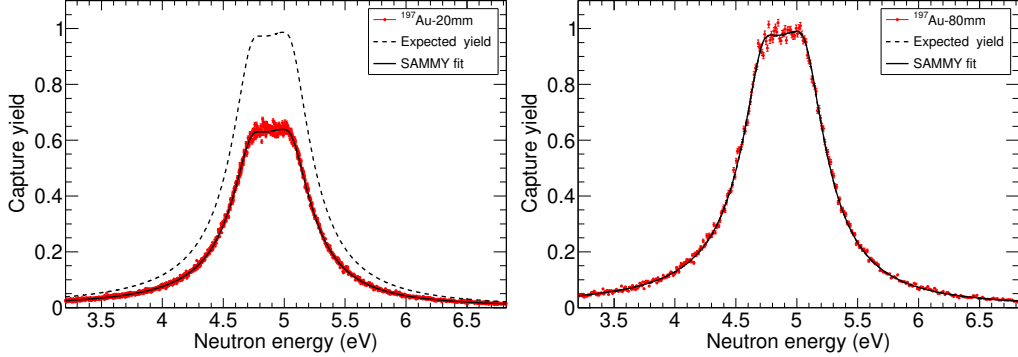


Figure 6.22: Capture yield corresponding to the 20 mm diameter (left) and 80 mm diameter (right) ^{197}Au sample fitted with SAMMY (solid line), and the expected yield (dashed line). These results allow for extracting the Beam Interception Factor (F_{BIF}).

As the gold sample was measured six times during the 6-week experimental campaign, the comparison of the F_{BIF} values obtained in these measurements and for each of the 4 detectors serves to test the reproducibility of the experiment. The average value features a reproducibility better than 0.5% when the values from each week are considered. Regarding the individual detectors, the F_{BIF} value for each one agree within 1.4%. The value considered in the analysis is thus $F_{BIF}=0.645(9)$.

This value is in agreement within uncertainties with the nominal value of 0.66(1) corresponding to the commissioning campaign of n_TOF Phase 4. Furthermore, the methodology for calculating the F_{BIF} has been validated through the measurement and analysis of a gold sample with 80 mm in diameter and similar thickness. Since this sample is larger than the ~ 40 mm of the beam, the full beam traverses the sample and indeed (see Fig. 6.22) a value of $F_{BIF}(80\text{ mm})=1.003(3)$ is obtained, in perfect agreement with the expected value of 1.

For the reasons explained above, what is important is the relative position of the ^{197}Au and chromium samples, so the possible misalignment between them has to be taken into account. We considered that the alignment is better than 0.5 mm because of the laser system of EAR1, so reflecting the analysis performed in [61], a conservative uncertainty of 2% has been considered for F_{BIF} .

6.4.2. Chromium yield

Once all correction and normalization factors have been determined, Eq. 6.16 is applied to obtain the capture yield of all the chromium samples. The data from each detector has been treated separately and the final reaction yield is the average of the four detectors.

The PHWT has been crucial to determine the efficiency and extract a yield; however, some remarks are due:

- Implementing the WF makes it possible that the C_6D_6 behave like TED detectors.
- The WF are geometry-dependent and hence every sample needs its own WF.
- Usually the background presents a very soft energy spectrum, so when weighting the background signals as well, the capture signal-to-background ratio improves. This may be more advantageous for a light nucleus like Cr, which features hard capture cascades with a high end-point energy.
- On the other hand, in the weighted neutron energy spectra the natural statistical fluctuations are enhanced because of the lack of statistics in the deposited energy spectra for each neutron energy bin.
- For isotopes with significantly different capture cascade patterns, the efficiency may be different for each one. The reason is that the main differences come from the primary de-excitations, corresponding to more energetic γ -rays, which are more affected by the weighting process because of the increasing shape of the WF (see Fig. 6.18). The consequence is that, for isotopes for which these differences are notable like our case, the PHWT implementation could change the relative intensity of the resonances after weighting the data.

For our specific case, the last point is going to be specially relevant. After determining the capture yield, the final step of the analysis is to use the code SAMMY to fit the resonance parameters. However, we observed that it was not possible to make any satisfactory fitting of the resonances from 20 keV and beyond. This is caused by the limited statistics in an involved way. The signals detected in each neutron energy bin follow the distribution illustrated in Fig. 6.21, however when there is limited statistics such distribution is sampled poorly and, because the weighting factor changes very rapidly with the signals energy (see Fig. 6.18), the overall weighting to the signals in a given neutron energy bin can be very different. The result is that the usual way of weighting all signals individually add fluctuations that deform considerably the resonances, modifying completely their shape and impeding the analysis.

There is an alternative methodology, developed for the first time by our group (see Lerendegui et al. [67]), in which an *averaged weighting factor* (AWF) is applied at once to the neutron energy counting rate spectrum, instead of the usual weighting of each individual signal contributing to the spectrum. This AWF is calculated as follows: the usual method is applied, and then the integral of each resonance is extracted for the weighted and unweighted neutron energy spectra. Afterwards, the ratio between both,

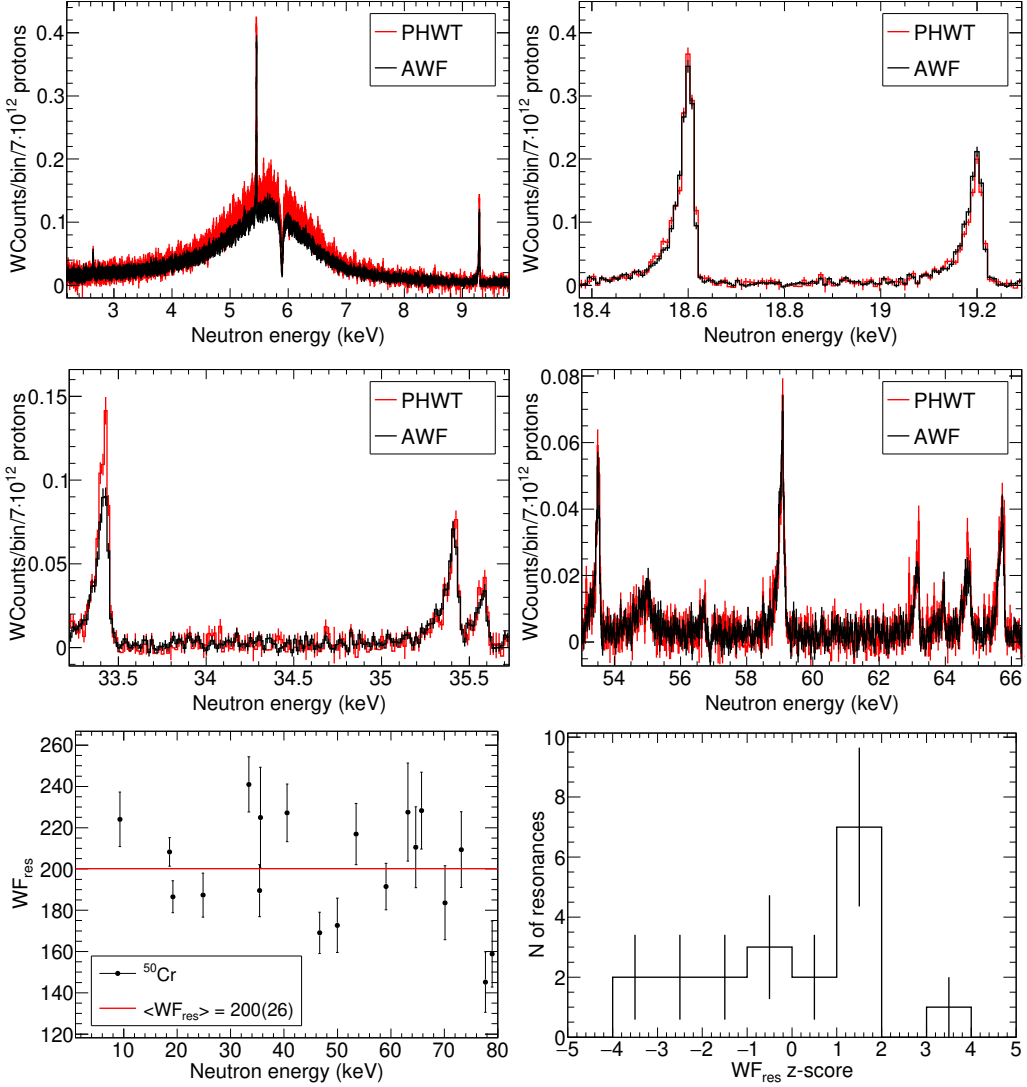


Figure 6.23: Top and middle: Comparison between the weighted spectra of ⁵⁰Cr obtained with the conventional PHWT (red) and innovative AWF (black) proposed by Lerendegui et al. Bottom: Summary of WF_{res} values (left) and their corresponding statistical distribution or z-score around the mean (right).

corresponding to the weighting factor of each resonance WF_{res} , is calculated for all resonances and an average value is obtained. In this way, the statistics are defined by the statistics accumulated over all resonances and not by that of each neutron energy bin, thus solving the issue related to the limited statistics.

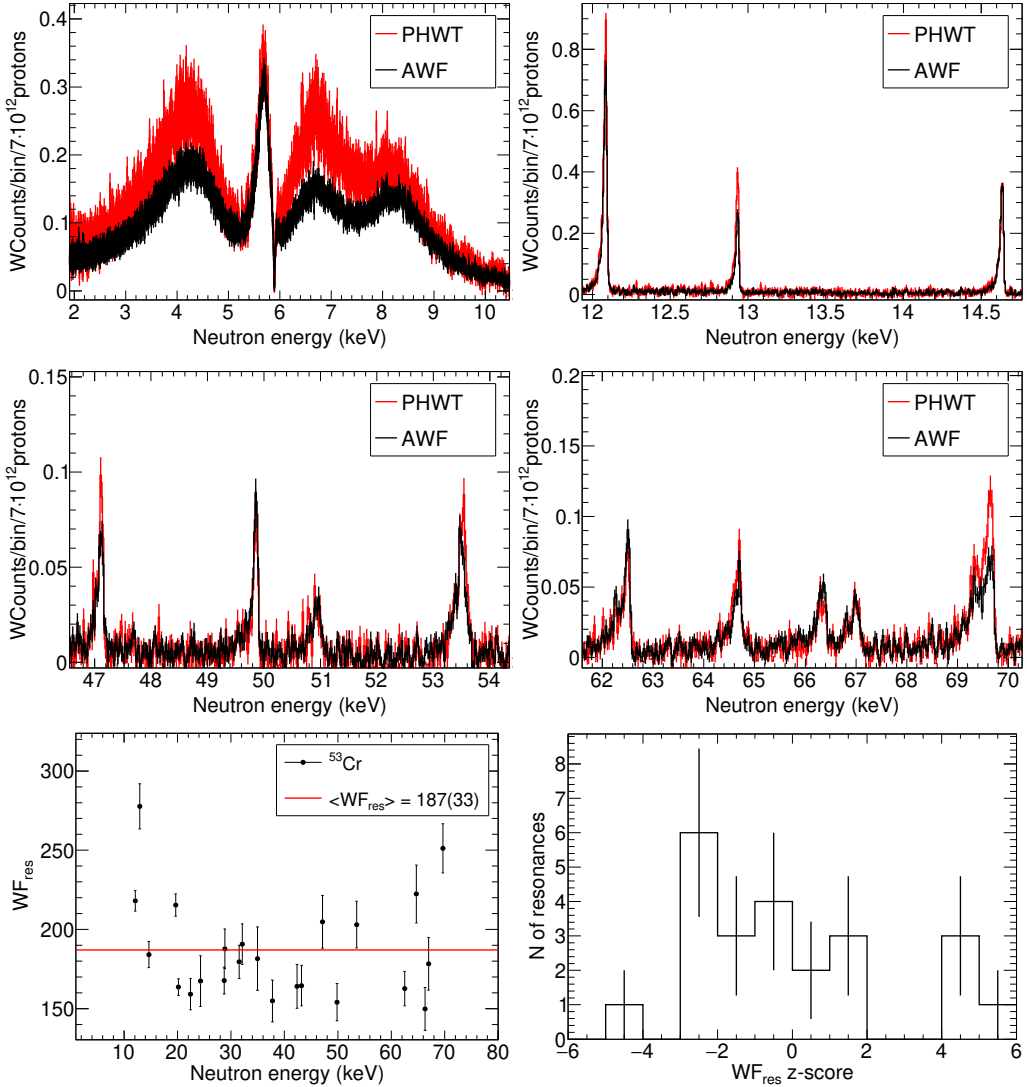


Figure 6.24: Top and middle: Comparison between the weighted spectra of ⁵³Cr obtained with the conventional PHWT (red) and innovative AWF (black) proposed by Lerendegui et al. Bottom: Summary of WF_{res} values (left) and their corresponding statistical distribution or z-score around the mean (right).

This methodology assumes that the de-excitation patterns is the same for all resonances, and such was the case for the ²⁴²Pu case for which it was developed; but not for chromium, as it was illustrated in Fig. 6.21. Even so, we tested the possibility of its implementation, calculating the weighted spectra of ⁵⁰Cr using both the conventional PHWT and the AWF proposed by Lerendegui et al.

The results are compared in Fig. 6.23, illustrating that they are compatible in some but not all cases, for instance for the s-wave resonance at 5.6 keV and the p-wave at 33.4 keV. The impossibility of using this method for the chromium data can also be illustrated by the statistical study displayed in the bottom panel. Even with the limited statistics (only 19 resonances), their WF_{res} values (left panel) are not distributed around the mean following the expected behaviour of a Gaussian distribution (right panel). For instance, only 1 instead of 5 resonances should be found beyond 2σ .

A very similar behaviour is observed for the case of ^{53}Cr -thick, as seen in Fig. 6.24. The disagreement in the main resonances between 1 and 10 keV is even more clear than in the previous case. On the rest of the range, there are clearly resonances that are incompatible between the two methods, for instance at 13, 47 or 69 keV. The same conclusions are extracted from the statistical study displayed in the bottom panel, where more than 40% of the resonances fall beyond 2σ .

Having confirmed that neither the conventional weighting of the individual signals (PHWT) neither the weighting of all the spectra at once (AWF) provide satisfactory results, we developed and implemented for the first time a new method based on the previous ideas of the AWF, but in which each resonance is scaled by its own factor WF_{res} : the *resonance weighting factor* technique (RWF).

The proposed strategy is as follows:

1. For the main chromium resonances between 1 and 10 keV and for ^{197}Au , because they are very strong and thus not affected by limited statistics, the conventional PHWT is applied.
2. For chromium from 10 keV onwards, each resonance is weighted by its own WF_{res} , which is not affected by the statistics of each individual bin but by the integral of the resonance. The ratio between the area of the unweighted and weighted resonances is calculated, chosen all the possible p-wave resonances with enough statistics (see Fig. 6.26). Those resonances are scaled by its individual weighting factor.
3. Some resonances feature so low statistics that an accurate WF_{res} cannot be calculated. In these cases, nothing can be made except scaling them to the average value of all other p-waves with enough statistics.

The result is shown in Fig. 6.25 for ^{50}Cr -thick (left) and ^{53}Cr -thick (right). The agreement is now very good in all cases and the fluctuations are kept minimized with respect to the conventional PHWT case. For some resonances (33.4 keV resonance of ^{50}Cr or the 47 keV one of ^{53}Cr) it might look like the data is not compatible, but it is an optical

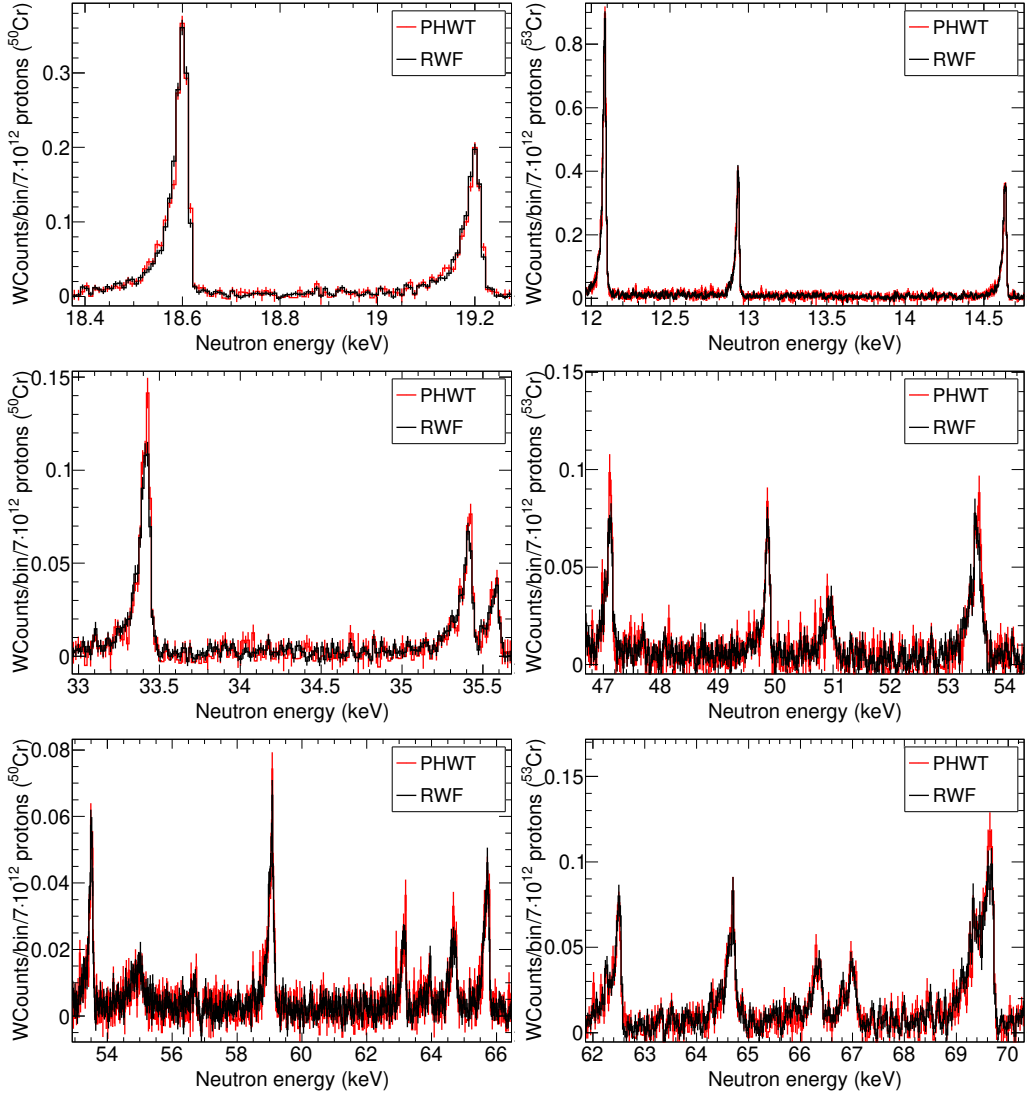


Figure 6.25: Comparison between the PHWT spectrum (in red) with the spectrum obtained by weighting each resonance by its own weighting factor (in black), showing the compatibility between both data (^{50}Cr -thick on the left column and ^{53}Cr -thick on the right one).

effect due to the large fluctuations on the weighted data. The method has thus proven to provide satisfactory results with only two limitations: *i*) having to apply an average WF_{res} value to resonances with too low statistics, and *ii*) its application to overlapping resonances would be problematic.

The implementation of the RWF technique introduce an additional uncertainty determined by the statistics of each resonance. In Fig. 6.26 the statistical uncertainty regard-

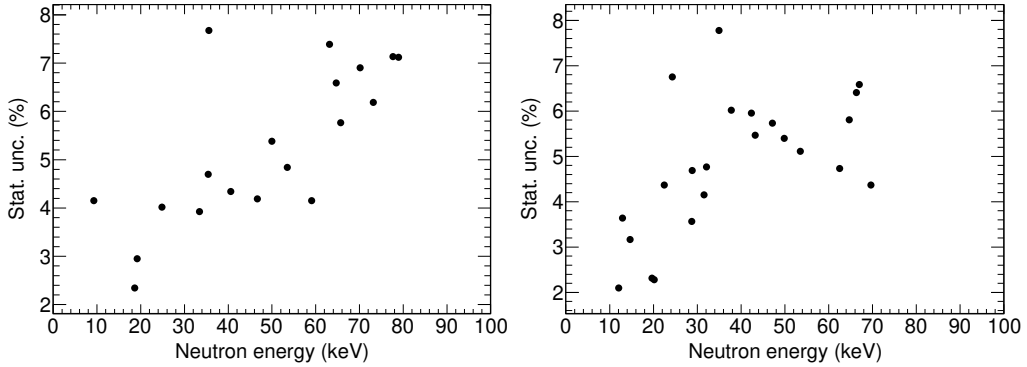


Figure 6.26: Statistical uncertainties extracted from the resonance area of ^{50}Cr (left) and ^{53}Cr (right). These have to be considered in the overall uncertainty of the final capture yield.

ing the total counts of each resonance is displayed. An increasing tendency is observed, because of the decrease in the resonance strength as a function of the neutron energy. Since the capture yield of the main resonances (below 10 keV) has been extracted using the classical weighting technique it is not affected by this source of uncertainty.

6.5. Summary of uncertainties of the capture yield

We have summarized in Table 6.3 all the sources of systematic uncertainty contributing to the calculation of the capture yield, which have been discussed in detail in the different Sections along the text:

- Sample thicknesses: the sample is weighted with high accuracy, thus only the uncertainty in the isotopic composition needs to be considered (Section 5.3.2).
- Normalization: uncertainty due to the normalization between different measurements using as reference the BCT and SILI monitors (Section 6.1.3).
- Neutron flux shape: the uncertainty in the shape of the evaluated flux in the energy ranges of interest is included (Section 5.2.1).
- PHWT: the study of the Q-factor and the simulated cascades allows to estimate the intrinsic uncertainty of the WF implementation (Section 6.3.1).
- Chromium cascades: the "convoluted" effect (expressed with $\otimes$ in Table 6.3) of the differences between the capture cascades and the correction associated to the detection threshold (Section 6.3.2).
- SRM normalization: the uncertainty of F_{BIF} is estimated from its variation in time and in between detectors (Section 6.4.1).

- Sample alignment: the possible misalignment between the gold and chromium samples has been taken into account (Section 6.4.1).
- RWF: the accuracy in the implementation of this new technique depends on the statistical uncertainty of each resonance (Section 6.4.2).
- Background subtraction: Since the background is flat in the energy range of interest, particularly in the valleys between resonances, and it is included in the SAMMY analysis, we consider that its contribution to the resonances' area uncertainty is negligible. (Section 6.2).

The final systematic uncertainty of the capture yield remains below 5% for the s-wave resonance cluster between 1 and 10 keV. For the rest of the resonances, the final uncertainty varies between 5% and 9% depending of the statistics of each one. In overall, the uncertainty is equal or smaller than 9%, fulfilling the requirements specified by the NEA needed to re-evaluate the $^{50,53}\text{Cr}$ neutron capture cross section.

Table 6.3: Contributions to the systematic uncertainty of the capture yields.

Contribution	Syst. unc. (%)	
	^{50}Cr	^{53}Cr
Sample thickness	0.4	0.2
Monitor normalization	2.5	2.5
Neutron flux shape	2	2
Saturated Resonance Method (SRM)	1.5	1.5
Sample alignment	2	2
<u>PHWT:</u>		
PHWT implementation	1.7	1.7
Cr cascades $\otimes f_{thr}$	1	1
RWF $\otimes$ statistics	2.3 - 7.7	2.1 - 7.8
Overall (1 - 10 keV)	4.5	4.5
Overall (10 - 100 keV)	5.1 - 9.0	5.0 - 9.0

7 | $^{50,53}\text{Cr}$ capture cross section resonance analysis

7.1. The code SAMMY

SAMMY [37] is a multilevel multichannel R-matrix code developed at the Oak Ridge National Laboratory (USA). It allows to perform the analysis of neutron (and charged particle) reaction cross section data, in the Resolved (RRR) and Unresolved Resonance Region (URR), by fitting the experimental yield through Bayesian methods. By default, the Reich-Moore (RM) approximation of the R-matrix is the formalism used by SAMMY (see Section 2.1.3).

There are several experimental conditions that SAMMY takes into account when fitting the shape of the resonances:

- **Doppler broadening.** The thermal motion of the target nuclei affects the shape of the resonances, causing a broadening that is higher for lower neutron energies [146]. There are several models to describe the Doppler broadening, but the most widely used is the so-called Free Gas Model (FGM) [147], being the desired option in our work. In this model, the broadened cross section σ_D takes the following form:

$$\sigma_D(E) = \frac{1}{\Delta_D \sqrt{\pi}} \int_0^\infty \left[e^{-4(E - \sqrt{EE'})^2 / \Delta_D^2} - e^{-4(E + \sqrt{EE'})^2 / \Delta_D^2} \right] \sigma(E') \sqrt{E'/E} dE, \quad (7.1)$$

where $\sigma(E')$ is the unbroadened cross section and Δ_D is the Doppler width given by

$$\Delta_D = \sqrt{\frac{4mEkT}{M}}, \quad (7.2)$$

where m and M are the neutron and target mass, k is the Boltzmann constant and T the effective temperature, $T = 295$ K in our case.

- **Resolution broadening.** As discussed in Section 5.2.3, the Resolution Function (RF) of the facility, which is related to the production and the moderation of the neutrons, also affects the resonances shape. In SAMMY, the RF can be introduced as a parameterized function, including specific parameters for the ORELA [25], GELINA [55] and n_TOF facilities, or as a numerical function obtained from simulations, which has been the adopted option in this work [128].
- **Self-shielding and multiple-scattering.** The finite size of the sample makes it possible that the reaction of interest occurs after some previous neutron scattering events. This is specially important in cases like chromium where neutron-scattering is the dominant process. In SAMMY, the capture yield is given by the sum of several components:

$$Y(E) = \sum_i Y_i(E), \quad (7.3)$$

where the index i indicates the number of scattering reactions occurred before the neutron is captured. The term $Y_0(E)$ is exactly calculated through Eq. 2.22, while $Y_1(E)$ already requires rather complex analytical expressions [37]. For the double-scattering and following terms $Y_{ms}(E)$, severe approximations have to be made to reduce the numerical calculation time, thus limiting the accuracy of the yield analysis for thick samples and/or isotopes with a high scattering-to-capture cross section ratio.

- **Multiple isotopes.** SAMMY allows to perform the analysis on samples with several isotopes, useful when the sample has an important amount of impurities or is a chemical compound, for instance an oxide, like in our case. The latter is important for a correct calculation of the multiple-scattering components of the yield.

7.2. Analysis strategy

The strategy adopted in the resonance analysis is the following:

1. The residual background present in the capture yield (see Section 6.2) is fitted to reproduce the valleys between resonances. In SAMMY, the background is described as the following:

$$B(E) = B_1 + B_2/\sqrt{E} + B_3\sqrt{E} + B_4e^{-B_5/\sqrt{E}}. \quad (7.4)$$

2. The region of the main s-wave resonances, from 1 to 10 keV, is analyzed using the thin samples, to minimize the multiple-scattering effects, which cannot be accu-

rately calculated with SAMMY and, furthermore, are affected by the uncertainty in the scattering cross section (which indeed is one of the main reasons for the inaccuracy of previous measurements).

3. The resonance parameters (E_n , Γ_γ and Γ_n) obtained below 10 keV are fixed for the analysis of the thick samples in the region between 10 and 100 keV, dominated by p-waves resonances.
4. Resonances are analyzed individually, except when some are overlapping or sitting on the tails of each other, in which case they are analyzed as a group.
5. To reduce uncertainties in the resonance parameters due to $\Gamma_\gamma - \Gamma_n$ correlations, only one of them is fitted if possible. For this, it is considered that the resonance area is proportional to $\Gamma_\gamma \Gamma_n / \Gamma$ (see Eq. 2.17) and thus to just Γ_γ when $\Gamma_\gamma \ll \Gamma_n$, which is true for most chromium resonances. If both resonance widths are comparable, then the fit is performed for all combinations of free parameters (Γ_γ , Γ_n or both). The results are then compared and, if the fit when leaving only one of the parameters free is as good or better than leaving both free, then the former is considered. The resonance energy, of course, is always fitted.
6. The spin of the resonances is chosen as the one given by the evaluations, unless the fit is not satisfactory, in which case another spin is proposed. There are a few cases in which the evaluations differ on the spin of a resonance, hence those are treated carefully and discussed later in a case by case basis.

7.3. ⁵⁰Cr resonance analysis

The measured capture yield and the one resulting from the SAMMY analysis of the ⁵⁰Cr resonances is compared to the evaluations from JEFF-3.3, CENDL-3.2 and the INDEN version of August 2023 (from now on denoted just as "INDEN"), since the rest of main evaluations like ENDF/B-VIII.0 or JENDL-5 contain the same resonance parameters as JEFF-3.3. A total of 30 resonances plus 3 not included in the current version of the evaluations are discussed in the following.

7.3.1. Main s-wave resonance at 5.6 keV

The cross section of ⁵⁰Cr between 1 and 10 keV is dominated by a broad s-wave resonance centered at 5.6 keV. For its analysis, the parameters from the three evaluations mentioned above were used for input of the a priori values required by the Bayesian method, resulting in a better reproduction of the measured yield when starting with the CENDL-3.2 parameters for this resonance and the negative resonances that describe the 1/v region.

The result is shown in Fig. 7.1, where a few features should be noted:

1. The effect of the aluminum dip present in the flux (see Fig. 5.4) has been removed,
2. The data points standing out at 1.5 keV correspond to the main resonance of ^{52}Cr , present in the enriched material (see Table 5.4) but not included in the analysis,
3. The narrow 5.46 keV resonance is a p-wave that is discussed in the next section.

First, an iterative fitting using both the thin and thick samples was performed, but the fitting of the thick sample was not as good (see right panel of Fig. 7.1). This is related to the mentioned multiple-scattering effects, which produce a deformation of the yield, specially in the ^{53}Cr -thick sample as discussed in the next section. Hence, the analysis is based on the fit of the thin sample only, for which both widths had to be left free. The result is displayed in the left panel of Fig. 7.1, where it is clear that our measurement is in agreement with JEFF-3.3 (and thus with ENDF/B-VIII.0 and JENDL-5) and in sizeable disagreement with CENDL-3.2 and INDEN. The resonance parameters obtained ($\Gamma_\gamma=3.226(15)$ eV and $\Gamma_n=1636(11)$ eV, with a -20% correlation) are compared with the ones from the different evaluations in Table 7.1. It is worth mentioning that although the measured resonance shape is quite different to that of CENDL-3.2, the corresponding resonance areas (i.e. the radiative kernels) are in good agreement; a casuistic that is discussed in more detail in Section 7.5.1.

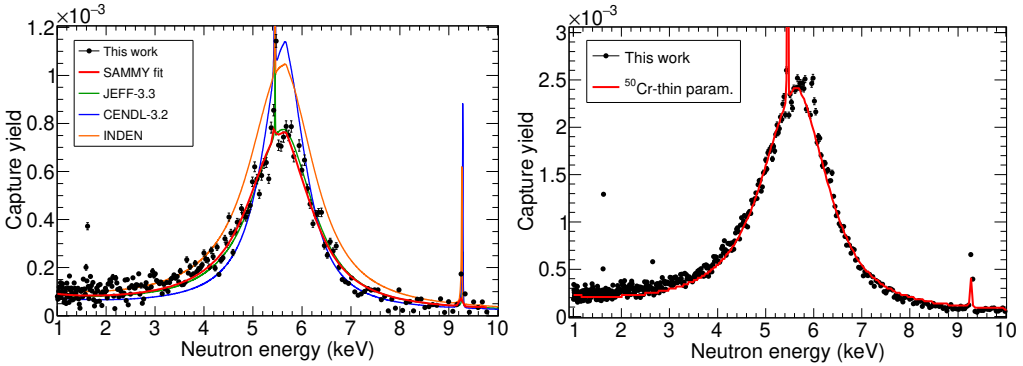


Figure 7.1: Left: Capture yield of ^{50}Cr -thin sample fitted with SAMMY, compared with the predicted one using different evaluations. Right: Capture yield predicted by SAMMY regarding the ^{50}Cr -thick sample, using the parameters obtained with the thin sample fitting.

Regarding the resonance energy, it had to be shifted with respect to the evaluations, resulting in $E_n = 5.581(6)$ keV instead of $E_n = 5.621$ keV according to the evaluations. The uncertainty in the energy has been estimated from the precision of the time-to-energy calibration (see Section 6.1.4). The bias on the value in the evaluations is most

Table 7.1: Resonance parameters of the ⁵⁰Cr main s-wave resonance obtained in this work compared with the values of the current evaluations. The uncertainty has been derived from the uncertainty of the widths provided by SAMMY.

	E (keV)	Γ_γ (eV)	Γ_n (eV)	Corr. (%)	K_γ (eV)
JEFF-3.3	5.621	3.250	1560	-	3.243
CENDL-3.2	5.621	3.250	1053	-	3.240
INDEN	5.621	4.561	1631	-	4.548
This work	5.581(6)	3.226(15)	1636(11)	-20	3.220(16)

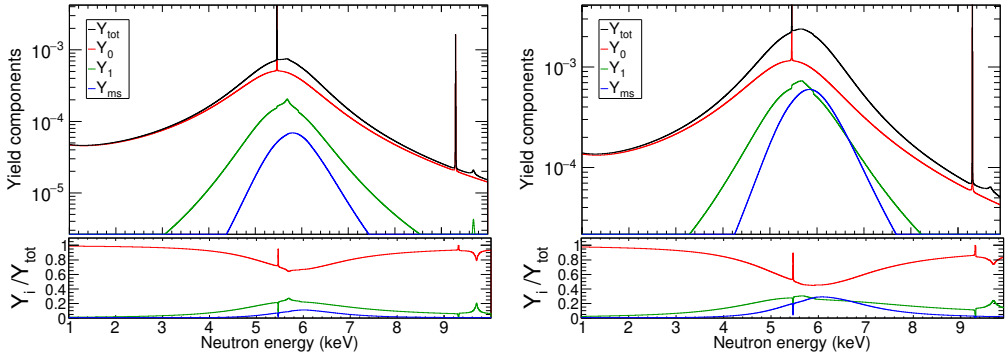


Figure 7.2: Individual yield components calculated with SAMMY for the ⁵⁰Cr-thin (left) and thick samples (right). The multiple-scattering components are more prominent in the thick sample, deforming the capture yield.

probably due, again, to the multiple-scattering effects in previous measurements. This makes the apparent energy of the resonances higher because of the high order multiple-scattering components of the capture yield. This is illustrated for the ⁵⁰Cr-thick in the right panel of Fig. 7.1, where the resonance is observed at a slightly higher energy than that reconstructed from the thin sample resonance parameters.

With SAMMY it is possible to calculate and visualize the individual components of the yield (see Eq. 7.3) and their relative contribution, as shown in the top and bottom panels of Fig. 7.2, respectively. In the the thick sample (right panel), Y_0 contributes less to the total yield than in the thin sample (left panel). In fact, Y_{ms} has the same magnitude than Y_1 , thus the effective energy shifting described above, and constitutes more than half of the total capture yield at the resonance maximum.

7.3.2. Resonances from 10 to 100 keV

The rest of the resonances were fitted with SAMMY following the strategy described in Section 7.2 using only the data from the thick sample because the thin sample did

not provide enough statistics for narrow and/or high energy resonances. From 10 keV, INDEN adopts the same parameters than JEFF-3.3, thus in the plots only JEFF-3.3 and CENDL-3.2 are displayed for comparison.

The complete set of resonance parameters can be found in Table A.1 of Appendix A. Here, we highlight the resonances that show some particularities (the cases depicted in the list are displayed in Fig. 7.3):

- **5.46 keV.** This very narrow p-wave resonance sits on top of the main s-wave one, being thus largely affected by it. Both resonance widths had to be fitted simultaneously.
- **20-24 keV.** At 21.8 keV there is a resonant-like structure that does not appear in the most recent evaluations, but it seems that it was included in JEFF-3.1 and then removed in JEFF-3.2. We propose to consider it again as a ⁵⁰Cr resonance, but a revision of previous data would be beneficial to confirm it. In the same region, at 22.9 keV there is another similar structure, but it corresponds to the second most intense resonance of ⁵²Cr.
- **24-25 keV.** It was not possible to obtain better parameters of the resonance at 24 keV than the ones from the evaluations, probably because it is too small. Since both JEFF-3.3 and CENDL-3.2 evaluations include the same parameters, which reproduce well the measurement, they were kept unchanged. In addition, the following resonance appears 500 eV lower in CENDL-3.2 than in our data.
- **35.5 keV doublet.** The second resonance of the doublet corresponds to a p-wave ($\ell = 1$), but in JEFF-3.3 its total momentum is assigned to $J = 3/2$ while in CENDL-3.2 is $J = 1/2$. Leaving all parameters free for both resonances, we found that the best fit corresponds to $J = 1/2$.
- **53 keV.** In our data, this resonance appears ~ 600 eV above, at 53.6 keV, than in the evaluations.
- **64 and 75 keV.** These two resonances were also removed from the evaluations in JEFF-3.2, but they were included in the previous version and they are clearly visible in our data (marked with arrows in Fig. 7.3). A satisfactory resonance analysis was possible for the one at 75 keV, but the statistics were limited for the 64 keV one and hence the parameters of JEFF-3.1 are adopted in this work.
- **80-100 keV.** In the high energy region the lack of statistics is more considerable, hence the analysis of several resonances is less reliable. For instance, there is a very small resonance at 89.6 keV included in JEFF-3.3 but not in CENDL-3.2, which our data cannot confirm or deny.

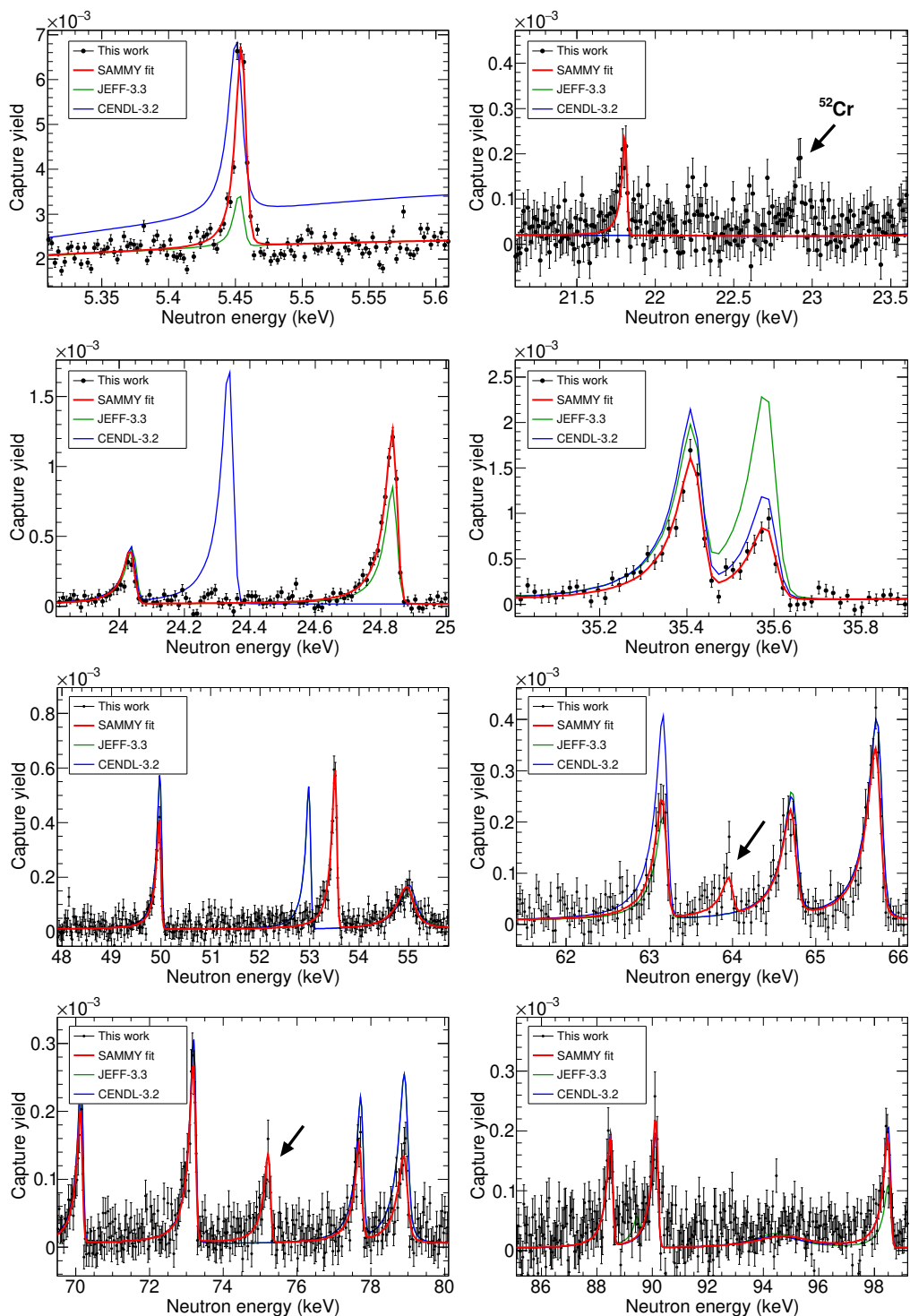


Figure 7.3: ^{50}Cr capture yield fitted with SAMMY, along with the one predicted using JEFF-3.3 and CENDL-3.2 for comparison. See text for more details.

The quantitative analysis and the implications of the cross section derived from the new resonance parameters is discussed in Section 7.5, by means of the individual resonances (radiative kernels) and integral (MACS) cross section values (see Section 2.2.3).

7.4. ^{53}Cr resonance analysis

For ^{53}Cr , JEFF-3.3 is again equivalent to ENDF/B-VIII.0 and JENDL-5, but different than INDEN, so both are included along with CENDL-3.2 in the following figures for comparison. A total of 51 resonances have been analyzed.

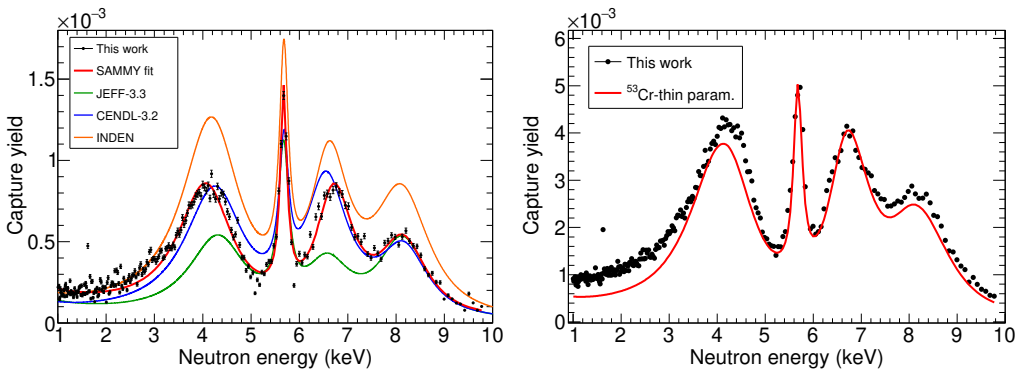


Figure 7.4: Left: Capture yield of ^{53}Cr -thin sample fitted with SAMMY, compared with the predicted one using different evaluations. Right: Capture yield predicted by SAMMY regarding the ^{53}Cr -thick sample, using the parameters obtained with the thin sample fitting.

7.4.1. Main resonances between 1 and 10 keV

The strategy followed for the analysis of ^{53}Cr is very similar to the ^{50}Cr case in this same region, with the particularity that the capture cross section between 1 and 10 keV is characterized by a cluster of four s-wave resonances. As a consequence, the fitting had to be performed to the whole structure at the same time. The result for the thin sample is shown in the left panel of Fig. 7.4, along with the expected yields corresponding to the JEFF-3.3, CENDL-3.2 and INDEN evaluations. In this case, the same trial of iterative fitting performed for the ^{50}Cr using the thin and thick samples produced even worst results, because the ^{53}Cr -thick sample is the most massive of all the chromium samples measured (see Table 5.3) and thus multiple-scattering effects are more prominent. Therefore, this region is analyzed using only the thin sample data. Then, when calculating the yield for the thick sample using those parameters, as expected the result is quite far from the measured yield (see right panel of Fig. 7.4). The first resonance at 4.1 keV is specially affected by multiple-scattering and neutron-scattering background,

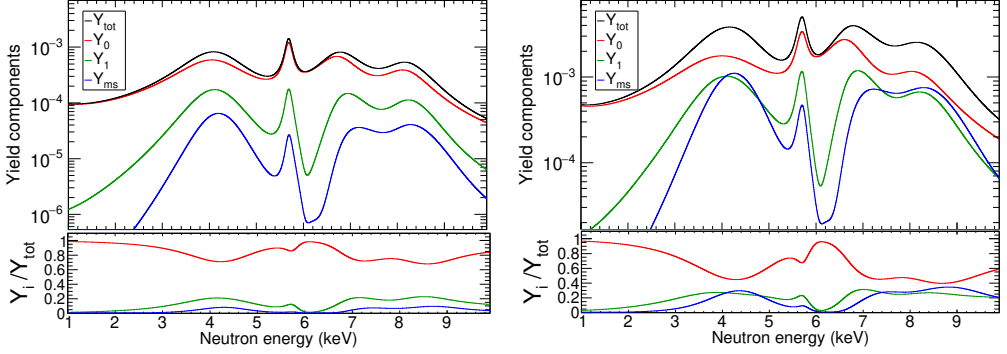


Figure 7.5: Individual yield components calculated with SAMMY for the ⁵³Cr-thin (left) and thick samples (right).

since it is receiving the background from the neutrons scattered in the three higher energy resonances, occurring at later times. The new resonance parameters obtained in this work are compared in Table 7.2 with the ones from the evaluations discussed here.

The individual components of the yield have been extracted with SAMMY and displayed in Fig. 7.5, showing clearly the neutron scattering problems associated to the thick sample. While the multiple-scattering component of the yield remains under 10% in the thin sample, its contribution is almost the same that Y_0 and even larger than Y_1 at the top of the s-wave resonances in the thick sample. This justifies the use of a very thin sample and not including the yield from the thick one in the study of this low energy region.

7.4.2. Resonances from 10 to 100 keV

The complete set of new resonance parameters proposed are listed in Table B.1 of Appendix B. For most resonances, by fixing one parameter (usually Γ_n because of the large neutron width of chromium) and fitting the other was enough to obtain a good result. Here we highlight the more interesting cases.

- **12.9 keV.** This resonance is an example in which both widths had to be fitted together. It is also a good example of the significant discrepancies between the evaluations. For instance, JEFF-3.3 and INDEN assign to this resonance a total spin of $J = 2$ with $s = 2$, and CENDL-3.2 $J = 1$ with $s = 1$, each one with different capture and neutron widths. The best result was obtained with $J = 2$ and $s = 2$.

Table 7.2: Resonance parameters of the ⁵³Cr main s-wave resonances obtained in this work compared with the values of the current evaluations. The uncertainty has been derived from the uncertainty of the widths provided by SAMMY.

	E (keV)	Γ_γ (eV)	Γ_n (eV)	Corr. (%)	K_γ (eV)
JEFF-3.3	4.248	2.076	1361	-	0.777
CENDL-3.2	4.210	3.410	1410	-	1.276
INDEN	4.139	5.038	1408	-	1.883
This work	4.033(4)	3.089(12)	1.332(8)	7	1.1558(17)
JEFF-3.3	5.671	0.616	231.6	-	0.384
CENDL-3.2	5.670	0.495	220.0	-	0.309
INDEN	5.675	0.787	201.9	-	0.490
This work	5.677(6)	0.541(7)	143(3)	29	0.337(3)
JEFF-3.3	6.788	1.711	1183	-	0.641
CENDL-3.2	6.750	4.130	1115	-	1.543
INDEN	6.779	4.144	956.6	-	1.547
This work	6.857(7)	3.31(4)	906(13)	53	1.236(5)
JEFF-3.3	8.207	1.911	1139	-	1.192
CENDL-3.2	8.180	1.570	1120	-	0.980
INDEN	8.191	3.353	1313	-	2.090
This work	8.210(8)	1.680(20)	1091(18)	35	1.049(8)

- **14.7 keV.** For this resonance CENDL-3.2 features a different energy, but according to our measurement, the correct one is 14.66 keV, in agreement with JEFF-3.3 and INDEN.
- **16 and 24 keV.** These two very small resonances are not present in CENDL-3.2, but they are in other evaluations. They are also visible in our data, confirming their existence.
- **28.8 keV doublet.** This couple of resonances appears as a doublet in JEFF-3.3 and INDEN but as a single one in CENDL-3.2. Our data confirms clearly that it is indeed a doublet, although the fit to the lower energy one is not as satisfactory as for other resonances.
- **41.9 keV.** According to JEFF-3.3 and INDEN there is a very small resonance at 41.9 keV. Unfortunately, our statistics are too limited to either confirm or deny its existence.

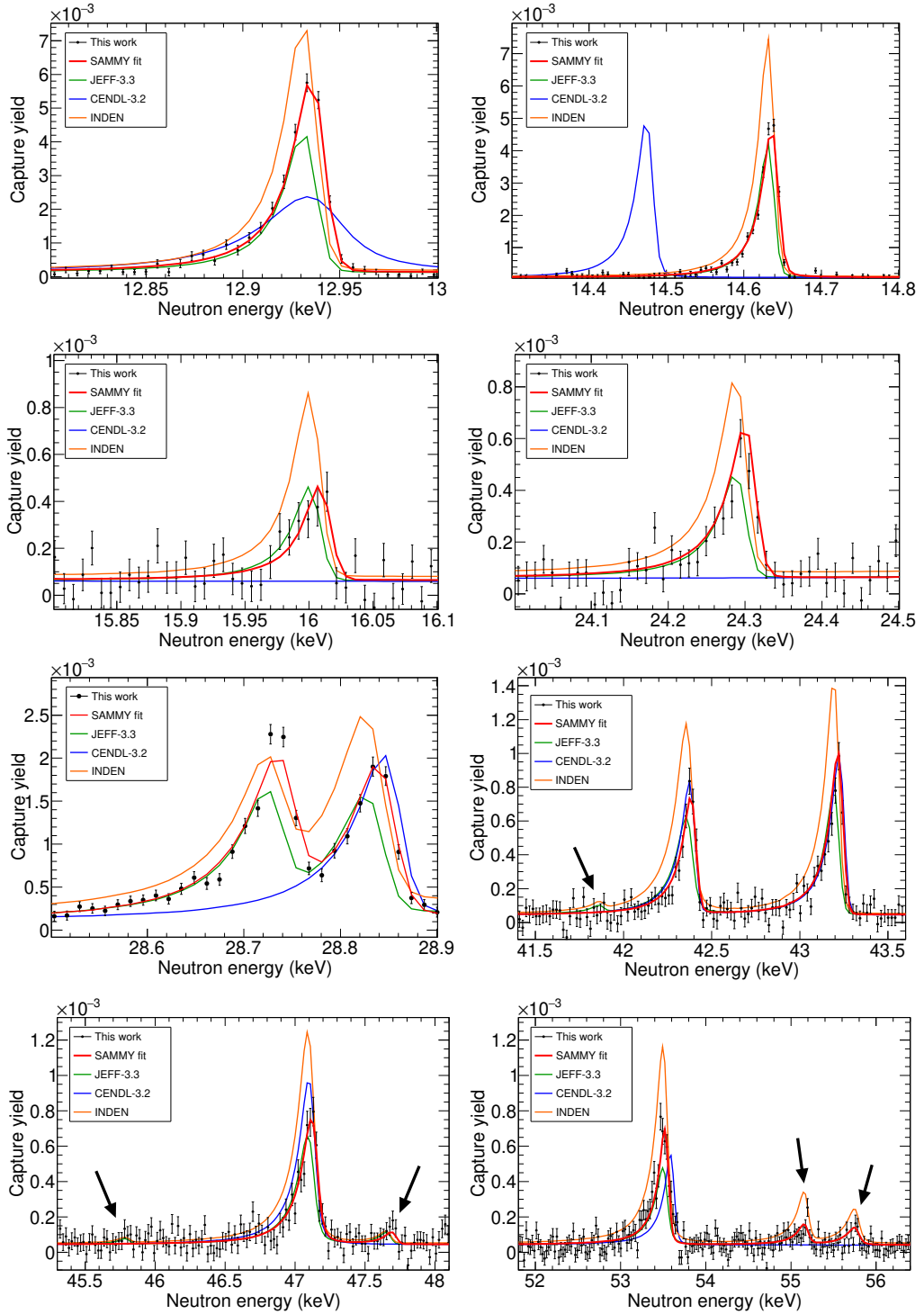


Figure 7.6: ^{53}Cr capture yield fitted with SAMMY, along with the one predicted using JEFF-3.3 and CENDL-3.2 for comparison. The arrows point to the smallest resonances, whose existence is arguable. See text for more details.

- **45-48 keV.** As in the previous case, there is a resonance at 45.9 keV included in the evaluations that we are not able to observe because of the limited statistics. On the other hand, an expected small resonance at 47.7 keV is visible above the background level. Since a good enough fitting was not possible, the resonance parameters from JEFF-3.3 are adopted.
- **55 and 56 keV.** Another two resonances which existence we can confirm but not extract its parameters. However, we can at least confirm that they are overestimated in INDEN.
- **62 keV doublet.** Thanks to the very good energy resolution of n_TOF-EAR1, we confirm that there is a doublet, as in JEFF-3.3 and INDEN. The best result is achieved by fixing the parameters of the first resonance to the ones of CENDL-3.2 and varying the second resonance.
- **66 and 67 keV.** These two resonances are also missing in CENDL-3.2, but visible in our data as well as in the other evaluations.
- **69.5 keV.** In this case, JEFF-3.3 and INDEN include one small resonance between the two that are visible in our data. Unfortunately, the resolution and statistics of our measurement is not enough to distinguish it in case it exists. Because the fit is the same whether the resonance is included or not, we decided not to.
- **70-80 keV.** In this region there are two p-waves resonances on top of two s-waves resonances. To find the best parameters, the whole cluster was fitted at once. A very small s-wave resonance at 72.4 keV is included in JEFF-3.3 and INDEN, but in our data is either hidden under the tail of the next resonance or not visible because of the statistics, hence we cannot confirm its presence. In addition, there are three small resonances at 75.7, 76.6 and 79.5 keV included in the same evaluations, but our data is too limited for them. We decided to keep the first two, because the fluctuations may indicate their presence, but the third one is definitively not visible, so we have not considered it.
- **80-100 keV.** This region is also characterized by a s-wave and p-wave resonance cluster. Because of the limited statistics, the resonance at 86.6 keV included in JEFF-3.3 and INDEN is not visible, and the presence of the small 93.1 keV resonance cannot be confirmed because it would be hidden under the tail of the next one.

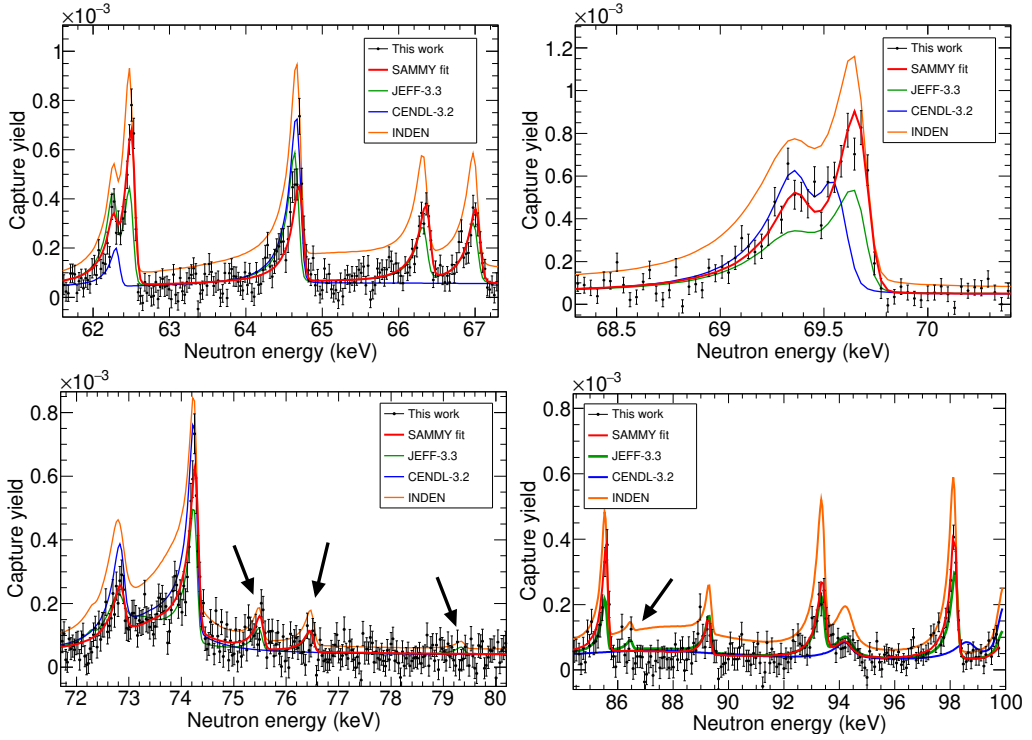


Figure 7.7: ^{53}Cr capture yield fitted with SAMMY, along with the one predicted using JEFF-3.3 and CENDL-3.2 for comparison. The arrows point to the smallest resonances, whose existence is arguable. See text for more details.

7.5. Comparison with previous data

The main goal of the measurement described here is to solve the discrepancies existing between the different $^{50,53}\text{Cr}(n,\gamma)$ evaluated cross section because they have a significant effect in criticality benchmarks, specially in the energy range from 1 to 10 keV. In this region, these discrepancies are more pronounced because previous measurements suffered from strong multiple-scattering and neutron background effects that have been minimized in this work.

Some general notes are given in the following, before the quantitative comparison is made of both resonance strength (through the radiative kernels) and Maxwellian Averaged Cross Section (MACS):

- Regarding the ^{50}Cr main resonance below 10 keV, our measurement is in better agreement with the JEFF-3.3 library, rather than with CENDL-3.2 or INDEN, the latter clearly overestimating the cross section. Above 10 keV, where JEFF-3.3

and INDEN adopt the same resonance parameters, we observe three resonances missing in the current libraries because they were eliminated in recent versions, but were also observed in old measurements.

- Concerning ⁵³Cr, the main s-wave resonance cluster below 10 keV is in better agreement with CENDL-3.2, although there are some differences. At higher energies, there are several resonances included in the most recent evaluations, JEFF-3.3 and INDEN, that are not visible in our data, due to lack of either statistics or resolution.

7.5.1. Radiative kernels

The capture or radiative kernel of a resonance, defined as $K_\gamma = g_J \Gamma_\gamma \Gamma_n / \Gamma$, allows for quantitative comparisons between cross sections in the RRR, since it is proportional to the area of the neutron capture resonance. We have obtained the ratio between the radiative kernel according to the new resonance parameters derived from the SAMMY analysis of the capture yield from the n_TOF measurement and the ones from the evaluations. The corresponding uncertainties are given by SAMMY as part of the fitting procedure, including the correlation between the widths when more than one is fitted.

The ratio between the K_γ from this work and the evaluations is represented in Fig. 7.8 as a function of the neutron energy (left) and also against the scattering-to-capture probability (right) of each resonance, i.e. Γ_n / Γ_γ . The latter serves to identify or discard trends related to neutron scattering issues. Some important aspects of the plots follow:

1. Only the resonances for which new parameters have been derived are represented. In addition, the two first p-waves resonances of ⁵⁰Cr, at 5.46 and 9.31 keV, are not included either. Their K_γ is so different between the evaluations, probably due because they are greatly influenced by the main s-wave resonance, that their ratios deviate almost a factor 4 from the mean.
2. The weighted mean values have been obtained using the uncertainty of the widths as the weighting factor. The corresponding weighted standard deviation is represented by the shaded lines.
3. The analysis of the ratio as a function of neutron energy has been divided below and above 10 keV, because as discussed already the lower energy region is dominated by a few broad s-waves while above it is by numerous narrow p-waves. Since for ⁵⁰Cr only one resonance has been included in the first region, the average cannot be calculated.

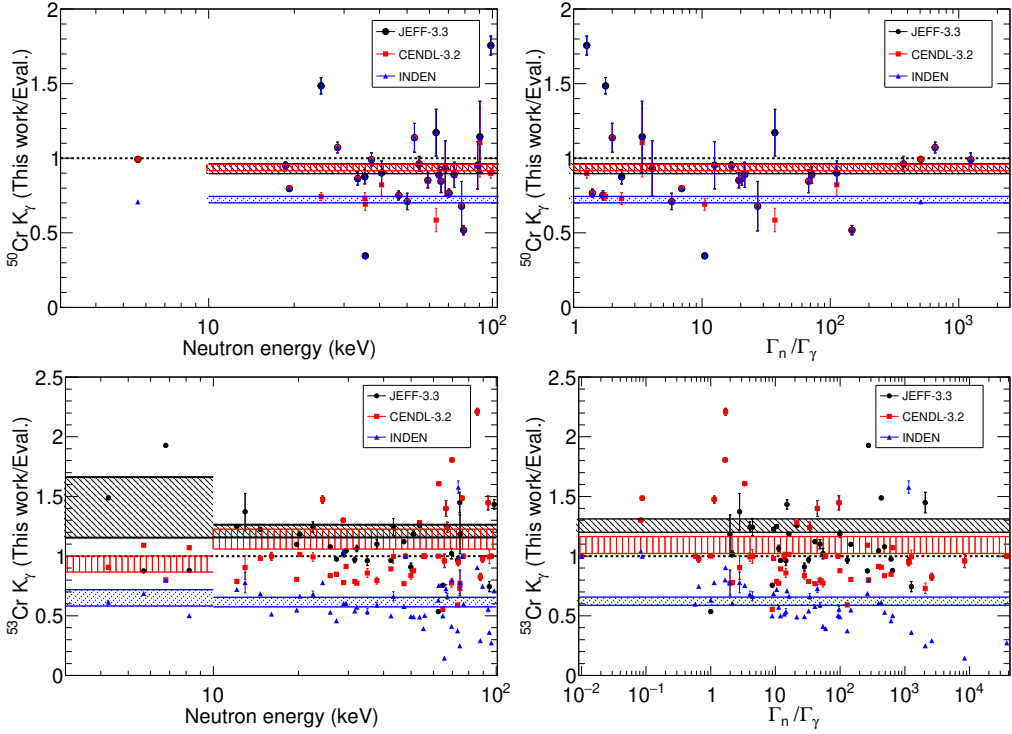


Figure 7.8: Ratio of radiative kernels obtained in this work and reported in the different evaluations as function of neutron energy (left) and scattering-to-capture ratio (right). The solid lines represent the standard deviation of the weighted mean value. See text for more details.

The results for ⁵⁰Cr is represented in the top panels of Fig. 7.8. From 10 keV onwards INDEN adopts the parameters from JEFF-3.3, and the ones from CENDL-3.2 are also very similar to these. Overall, there is an overestimation of the radiative kernels in the previous evaluations with respect to this work, being just $\sim 7\%$ for JEFF-3.3 and CENDL-3.2 but almost 40% for INDEN. Regarding the dominant s-wave resonance at 5.64 keV, its kernel is a remarkable 40% lower than that of INDEN, but is in perfect agreement with JEFF-3.3 and CENDL-3.2, even though the resonance shape is very different compared to CENDL-3.2 (see Fig. 7.1). Regarding the behaviour with respect to the scattering-to-capture ratio, there does not seem to be any clear trend.

The same kind of comparison is displayed for ⁵³Cr in the bottom panels of Fig. 7.8. In this case, the three evaluations considered feature very different resonance parameters. Compared to the result of our measurement and analysis, JEFF-3.3 and CENDL-3.2 are on average 25% and 9% lower, respectively; while the highest discrepancy is observed again when comparing to INDEN, which overestimates the ⁵³Cr capture cross section

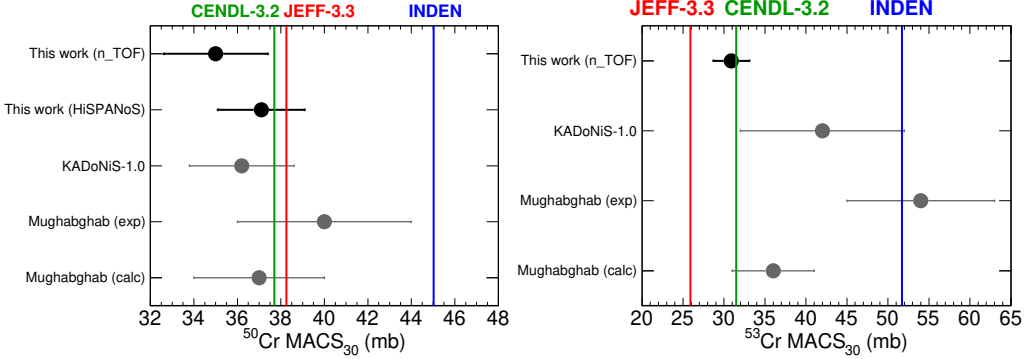


Figure 7.9: MACS₃₀ of $^{50,53}\text{Cr}$ obtained in the measurements described in this work, along with the values from the data bases and the ones derived from the main evaluations.

by nearly 60%. Focusing on the main s-wave resonances below 10 keV, it is clear that our data is in better agreement with CENDL-3.2 ($\pm 10\%$ differences), and that the comparison with JEFF-3.3 shows a very large dispersion from resonance to resonance. Regarding neutron-scattering effects, an apparent decreasing trend in the kernel ratio with respect to Γ_n/Γ_γ is visible for the INDEN case. This could indicate that the estimation of neutron scattering effects in the recent INDEN evaluation is overestimated, but this would have to be confirmed or revised by the evaluators.

7.5.2. Maxwellian Averaged Cross Sections (MACS)

In contrast to the previous comparison of K_γ on a resonance by resonance basis, the comparison in terms of the Maxwellian Averaged Cross Section (see Eq. 2.25) at a temperature of $kT = 30$ keV (MACS₃₀) provides an integral comparison precisely in the energy range of interest in this work. It is, therefore, a usual way to compare cross sections in the epithermal energy region (see Tables IV to VII in Ref. [26]). Therefore, the MACS₃₀ values of $^{50,53}\text{Cr}$ have been derived from the new cross section obtained in this work using Eq. 2.25. However, the new resonance parameters obtained in this work cover the region only up to 100 keV, thus the higher energy range (which has a marginal effect but still needs to be considered) has been completed with the different evaluations, obtaining MACS values for each of them. For this we have considered only JEFF-3.3 and CENDL-3.2, because INDEN has been shown to be clearly incompatible with our measurement.

When completing the region above 100 keV with JEFF-3.3, the results are $\text{MACS}_{30}(^{50}\text{Cr}) = 34.3$ mb and $\text{MACS}_{30}(^{53}\text{Cr}) = 30.9$ mb, and when using CENDL-3.2, $\text{MACS}_{30}(^{50}\text{Cr}) = 35.6$ mb and $\text{MACS}_{30}(^{53}\text{Cr}) = 30.8$ mb; in all cases in agreement with each other within

Table 7.3: MACS₃₀ values of ⁵⁰Cr and ⁵³Cr obtained in this work, compared to the values available in the evaluations and data bases.

MACS ₃₀ (mb)	⁵⁰ Cr	⁵³ Cr
<u>Compilations:</u>		
KADoNiS-1.0 (2014)	36.2(24)	42(10)
Mughabghab (exp) (2018)	40(4)	54(9)
Mughabghab (calc) (2018)	37(3)	36(5)
<u>Evaluations:</u>		
JEFF-3.3 (2017)	38.2	25.9
CENDL-3.2 (2020)	37.7	31.5
INDEN (2023)	45.0	51.7
<u>This work:</u>		
n_TOF	35.0(24)	30.9(22)
HiSPANoS	37.1(20)	-

just a few percent. The uncertainty of the MACS is difficult to estimate because is not only related to that of the capture yields but also of the resonance analysis. Furthermore, the uncertainty in the yield varies between 5% and 9% depending in the neutron energy. The choice is thus to adopt the average MACS value from the calculations extrapolated with the two evaluations mentioned above, to which a conservative uncertainty of 7% is assigned, resulting in:

$$\text{MACS}_{30}(^{50}\text{Cr}) = 35.0(24) \text{ mb}, \quad (7.5)$$

$$\text{MACS}_{30}(^{53}\text{Cr}) = 30.9(22) \text{ mb}. \quad (7.6)$$

These values are compared to those from the evaluations in Table 7.3. Regarding ⁵⁰Cr, our MACS₃₀ is lower than JEFF-3.3 and CENDL-3.2 by just 9% and 8% respectively; but, as it happened in the comparison of the K_γ , the largest difference (a very significant 30%) is found with respect to INDEN. This is remarkable, considering that it is the more recent and was specially developed to solve the issues observed in criticality safety experiments and calculations associated to these particular cross sections. The results from this work are in very good agreement with those from neutron activation at HiSPANoS reported in Section 4.3), serving as validation of both results because they are complementary and independent from each other, except for the sample. The comparison between all the different values is also displayed in Fig. 7.9.

Regarding ⁵³Cr, the values summarized in Table 7.3 illustrate a much more important disagreement, up to a factor of 2 just between the MACS₃₀ from the evaluations. Because an activation measurement is not possible with this isotope, unfortunately we cannot compare the new n_TOF value with any other alternative measuring technique. When

compared to the evaluations, our MACS₃₀ is clearly in agreement with CENDL-3.2 but 20% larger than JEFF-3.3 and 70% lower than INDEN. This is specially relevant for the mentioned issues in criticality safety, because when compared to ⁵⁰Cr, ⁵³Cr has a comparable capture cross section but its abundance is more than twice, and so is its role in the absorption of neutrons in a nuclear reactor environment.

Part IV

Summary and Conclusions

8 | Summary and conclusions

The role of nuclear energy has been recognized crucial in the fight to reduce CO₂ emissions in a global context of increasing energy demand. The development of more sustainable and safer nuclear reactors relies on nuclear data that should be more accurate than ever. In this context, criticality safety benchmarks help finding the specific nuclear data that should be improved because the calculations with the current data libraries do not reproduce as accurately as they should some specific research reactor configuration performances. For instance, it was recently found out that the uncertainties related to the modeling of neutron capture in some isotopes of chromium (mainly ⁵⁰Cr and ⁵³Cr), a major component of stainless steel, are behind the problems to calculate accurately the k_{eff} in several benchmarks. Indeed, the current capture cross section evaluations (like JEFF-3.3, BROND-3.1 or CENDL-3.2) of ⁵⁰Cr and ⁵³Cr show discrepancies with each other in the order of 30%, which gives an idea of the associated uncertainty. Such high uncertainty is related to the fact that the old chromium measurements may have suffered from neutron scattering effects because of the unavailability of low neutron sensitivity scintillators; while the most recent measurement by Guber et al. (2011) seems to have samples related issues. This is important, because the 30% uncertainty in these capture cross sections has an impact of about 1% on the k_{eff} in some of these benchmarks.

In order to improve this situation, the Nuclear Energy Agency (NEA) called for new measurements of the ^{50,53}Cr capture cross section between 1 and 100 keV with an accuracy of 8 to 10%, with emphasis on the 1 to 10 keV region where a broad s-wave resonance array is found. Very recently, the INDEN project carried out a re-evaluation of the chromium cross section combining the Guber et al. data with some of the old ones, proposing an important increase of the cross sections to improve the performance of some benchmarks, but calling for new measurements because the evaluation required many ad-hoc corrections.

In response to these requests by the NEA and the INDEN project, two new experiments have been designed and performed in this work: a time-of-flight measurement of both isotopes at the n_TOF facility of CERN (Switzerland) and a ⁵⁰Cr neutron activation measurement at the HiSPANoS facility of CNA (Spain). Both have been designed to

overcome the limitations of previous measurements and, moreover, it is the first time that the INDEN evaluation is tested against new experimental data.

One of the keys of both experiments is the highly enriched chromium samples. The physical characteristics of the chromium oxide powder made the fabrication of a self-sustained sample with the desired mass a big challenge by itself. The final process of compressing the powder inside a PEEK capsule and subsequent X-ray TC analysis proved to be the best options, offering a solution for future measurements of similar materials.

8.1. Neutron activation measurement at HiSPANoS

A neutron activation measurement of ^{50}Cr was performed at HiSPANoS using a high enriched chromium oxide sample. A quasi-maxwellian neutron spectrum at a temperature of $kT \sim 30$ keV was produced via the $^7\text{Li}(p,n)$ reaction with 1912 keV protons, which was previously characterized by time-of-flight. The neutron production target consisted on a thick metallic lithium disk (instead of the alternative of using LiF) in order to maximize the intensity of the neutron flux and achieve a large enough number of captures in the chromium sample. Two ^{197}Au foils were glued at each side of the sample holder, forming a sandwich. Since gold capture cross section is a standard among neutron reactions, the chromium capture cross section was measured relative to the one of gold.

The proton beam current impinging on the Li target was registered during the full duration of the experiment in order to be estimate the decay rate during the irradiation. The activation of the Cr and Au samples was determined through γ -ray spectroscopy using a 1.5"x1.5" LaBr_3 scintillator carefully calibrated, to take into account the geometrical differences between the samples. In relation to the neutron irradiation, a series of very detailed GEANT4 simulations of the sample assembly and the irradiation set-up were a key to estimate the several experimental effects such as neutron-scattering, inhomogeneities of the Cr sample, and differences between the real and the ideal Maxwell-Boltzmann neutron energy distributions.

8.2. Time-of-flight measurement at n_TOF-EAR1

The neutron capture cross section of $^{50,53}\text{Cr}$ was measured at the 185 meters flight path station of the n_TOF facility, known as EAR1. The capture set-up consisted on an array of four C_6D_6 Total Energy detectors, characterized by their low neutron sensitivity, that registered the γ -rays produced in the de-excitation cascade following the neutron capture. Two enriched chromium oxide samples were measured for each isotope:

Table 8.1: Summary of the $^{50,53}\text{Cr}$ neutron capture cross section measurements performed using the time-of-flight technique.

Measurement	Stieglitz	Beer	Kenny	Brusegan	Guber	This work
Detector	Sc.tank	Sc.tank	C_6F_6	C_6F_6	C_6D_6	C_6D_6
^{50}Cr n(10^{-3}at/b)	8	18	5	7	-	0.6/1.9
^{53}Cr n(10^{-3}at/b)	14	14	8	12	14	1.2/5.9

a thin one to determine the capture yield below 10 keV with very low multiple-scattering associated corrections, and a thick one for which these corrections were larger but allowed accumulating enough statistics up to the aimed upper limit of 100 keV. As summarized in Table 8.1, both samples were significantly thinner than any of those used in previous measurements, thus ensuring a better control of the multiple-scattering effects that have been pointed as the cause of the discrepancies between previous measurements.

The capture yield as function of the neutron energy was determined following a dedicated process described throughout this manuscript:

- Regular energy and resolution calibrations of the detectors using a combination of radioactive sources measurements and Monte Carlo simulations.
- Discrimination of signals not coming from capture events and rebounds, by means of a dedicated PSA and cuts in energy deposition and in rise time.
- Monitoring of the number of protons and neutrons to control the performance of the detectors and the normalization between measurements.
- Time-to-energy calibration using the well-known ^{197}Au resonances.
- Background measurements including, among others, a Dummy and a $^{\text{nat}}\text{C}$ samples.
- Simulations of the capture cascades to apply accurately the Pulse Height Weighting Technique (PHWT).
- Normalization of the capture yield by means of the Saturated Resonance Method (SRM) with the 4.9 eV resonance of ^{197}Au .
- Development of an innovative method to apply the PHWT for individual resonances with low statistics, avoiding artificial fluctuations (RWF).

The resonances observed in the measured capture yield have been analyzed using the code SAMMY, which provided a new set of resonance parameters, with their uncertainties and correlations.

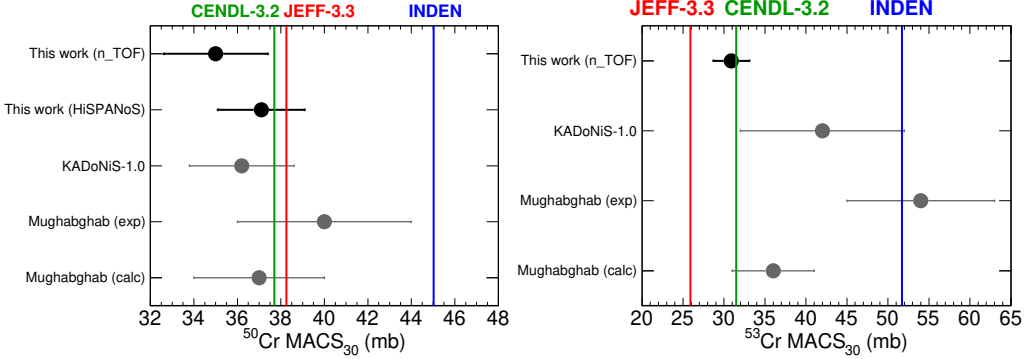


Figure 8.1: MACS₃₀ comparison between the values obtained in this work for $^{50,53}\text{Cr}$, the values derived from the evaluations and the ones included in the nuclear data bases.

8.3. Results and discussion

The successful measurements have provided high quality data on $^{50,53}\text{Cr}$ consisting on:

- The Maxwellian Averaged Cross Section (MACS) of ^{50}Cr at 30 keV with an accuracy of 5%, thus fulfilling the requirements of the NEA request.
- Neutron capture yields of ^{50}Cr and ^{53}Cr with an accuracy between 5 and 9% covering the full energy region of interest between 1 and 100 keV, thus fulfilling the requirements of the NEA request.
- A set of resonance parameters (E_n , J , Γ_γ , Γ_n) for ^{50}Cr (33 resonances) and ^{53}Cr (51 resonances), from which also the corresponding radiative kernels K_γ and MACS₃₀ have been calculated.

The MACS₃₀ of ^{50}Cr obtained in the activation measurement is 37.1(20) mb, being this the first time that it has been directly determined experimentally. The result is in good agreement with the libraries JEFF-3.3 (and, therefore, also ENDF/B-VIII.0, JENDL-5 and BROND-3.1) and CENDL-3.2, as displayed in Fig. 8.1; all of them in contradiction with the INDEN evaluation that overestimate the MACS by $\sim 20\%$. Such higher cross section value is not supported either by the values provided in Mughabghab nor by KADoNIS-1.0, both in relatively good agreement with our results.

Regarding the time-of-flight measurements, the accuracy of the measured capture yields and the subsequent resonance analysis with SAMMY allowed for a comparison against the different cross section evaluations.

- For ^{50}Cr , we report 3 resonances not present in the most recent evaluations but that were included in previous versions like JEFF-3.1. Regarding the main s-wave

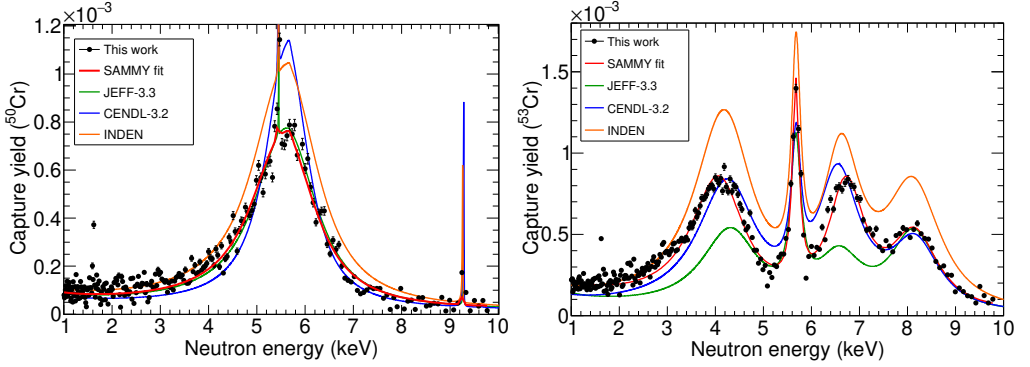


Figure 8.2: Capture yield of ^{50}Cr (left) and ^{53}Cr (right) measured at n_TOF and fitted with SAMMY, compared with the previous evaluations.

resonance at 5.6 keV, which dominates the 1 to 10 keV region, it is in good agreement with JEFF-3.3 but significantly different than CENDL-3.2 and INDEN, as illustrated in Fig. 8.2 (left panel). At higher energies, the comparison of the K_γ shows that, on average, our results are not far ($\sim 7\%$ lower) from JEFF-3.3 and CENDL-3.2, but again almost 40% lower than INDEN. This is reflected on the reconstructed MACS₃₀ value of 35.0(24) mb (see left panel of Fig. 8.1), compatible with uncertainties with the one from HiSPANoS (6% lower), and thus again significantly lower than the recently released INDEN evaluation.

- For ^{53}Cr , the evaluations show much larger discrepancies. In our measurement, although a few resonances in the evaluation could not be observed, the quality of our data is enough to resolve the discrepancies between evaluations. Regarding the cluster of s-wave resonances that dominates the cross section below 10 keV, our data is not very different from CENDL-3.2 but very far from JEFF-3.3 and INDEN evaluations (see right panel of Fig. 8.2), the latter again overestimating the cross section by 30-40% in this region. Above 10 keV, the new radiative kernels are, on average, 25% larger than JEFF-3.3, 9% larger than CENDL-3.2 and 60% lower than INDEN. Looking at the radiative kernels as function of the scattering-to-capture ratio, INDEN shows a decrease that could indicate an overestimation of the multiple-scattering and neutron background correction implemented in the evaluation, but it cannot be confirmed by this work. Last, the reconstructed MACS₃₀ using the new data is 30.9(22) mb, in agreement with the one from CENDL-3.2 and KADoNiS-1.0 (with this one because of its large uncertainty), 20% larger than JEFF-3.3, and 70% lower than INDEN (see right panel of Fig. 8.1). As in ^{50}Cr , the increase in the chromium cross sections proposed in the INDEN evaluation is not supported with this new experimental data.

Overall, the new experimental data are of high accuracy and meet the conditions of the NEA request. Thus, they serve to test the current evaluations, indicating that the increase in both cross section proposed by the new evaluation INDEN is incompatible with the experimental data presented herein. A new evaluation should be made using the data obtained in this work, maybe combined with previous ones of transmission and/or using natural samples. Hopefully this will contribute to a better understanding of the current issues in criticality safety benchmarks sensitive to stainless steel, and in particular to chromium.

Part V

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Part VI

Appendices

A |

Resonance parameters of
 ^{50}Cr between 1 and 100 keV

Table A.1: Resonance parameters of ^{50}Cr obtained in this work. The uncertainty of the resonance energy has been considered as 0.1% according to the time-to-energy calibration discussed in Section 6.1.4. The uncertainty of the resonances widths is the one obtained in the SAMMY fitting. If a value has no uncertainty it means that it has not been fitted, and that has been extracted directly from some evaluated library (see Section 7.3.2).

E (keV)	J	ℓ	Γ_γ (eV)	Γ_n (eV)	Corr. (%)	K_γ (eV)
5.464(5)	1.5	1	0.9(3)	0.0315(6)	-20	0.61(3)
5.581(6)	0.5	0	3.226(15)	1636(11)	21	3.220(16)
9.306(9)	0.5	1	1.2(7)	0.0539(18)	-28	0.0516(18)
18.63(2)	1.5	1	0.318(5)	5.400	-	0.601(16)
19.23(2)	0.5	1	0.394(8)	2.740	-	0.344(5)
21.85(2)*	0.5	1	0.370	0.039(7)	-	0.035(5)
24.08(2) ⁺ @	1.5	1	0.170	0.049	-	0.076
24.88(2)•	1.5	1	0.203(9)	0.360	-	0.260(10)
28.48(3)	0.5	1	0.598(20)	392	-	1.85(8)
33.49(3)	1.5	1	0.454(13)	9.300	-	0.87(5)
35.48(3)	1.5	1	0.53(3)	1.250	-	0.74(4)
35.65(4)	0.5	1	0.48(3)	5.000	-	0.436(25)
37.57(4)	0.5	0	1.85(8)	2300	-	0.75(4)
40.65(4)	1.5	1	0.348(16)	39(7)	-	0.69(6)
46.74(5)	1.5	1	0.53(3)	0.900	-	0.67(3)
50.07(5)	1.5	1	0.271(14)	1.570	-	0.462(8)
53.62(5)•	1.5	1	0.59(6)	1.180	-	0.79(7)
55.13(5)	0.5	0	0.76(4)	281	-	0.75(4)
59.21(6)	1.5	1	0.578(19)	11.200	-	0.110(6)
63.29(6)	1.5	1	0.270(19)	10.000	-	0.53(7)
64.08(6) ⁺ †	0.5	1	0.370	0.250	-	0.149
64.85(6)	0.5	0	0.60(4)	43.000	-	0.59(4)
65.87(7)	1.5	1	0.490(23)	33.100	-	0.96(9)
68.24(7)	0.5	1	0.44(13)	1.800	-	0.35(7)
70.28(7)	0.5	1	0.88(9)	1.230	-	0.513(18)
73.35(7)	1.5	1	0.442(23)	9.500	-	0.84(8)
75.38(7)*	0.5	1	0.46(5)	4.200	-	0.41(4)
77.86(8)	0.5	1	0.52(14)	14.200	-	0.50(12)
79.08(8)	0.5	1	0.68(4)	100	-	0.67(4)
88.69(9)	1.5	1	0.43(4)	5.340	-	0.79(13)
90.33(9)	1.5	1	0.64(11)	2.170	-	0.98(20)
94.95(9) [@]	0.5	0	0.967	2200	-	0.967
98.71(10)	0.5	1	1.82(21)	2.300	-	1.02(4)

+) Both parameters from JEFF-3.3

@) Both parameters from CENDL-3.2

†) Both parameters from JEFF-3.1

•) Energy discrepancy

*) Removed in JEFF-3.2

B | Resonance parameters of ^{53}Cr between 1 and 100 keV

Table B.1: Resonance parameters of ^{53}Cr obtained in this work. The uncertainty of the resonance energy has been considered as 0.1% according to the time-to-energy calibration discussed in Section 6.1.4. The uncertainty of the resonances widths is the one obtained in the SAMMY fitting. If a value has no uncertainty it means that it has not been fitted, and that has been extracted directly from some evaluated library (see Section 7.4.2).

E (keV)	J	ℓ	Γ_γ (eV)	Γ_n (eV)	Corr. (%)	K_γ (eV)
4.033(4)	1	0	3.089(12)	1332(8)	7	1.1558(17)
5.677(7)	2	0	0.541(7)	143(3)	29	0.337(3)
6.857(7)	1	0	3.31(3)	906(13)	53	1.236(5)
8.210(8)	2	0	1.680(20)	1091(18)	35	1.049(8)
12.11(1)	2	1	0.430(6)	4.500	-	0.2452(17)
12.96(1)	2	1	0.212(7)	0.59(6)	-64	0.097(11)
14.66(1) [•]	1	1	0.343(7)	3.200	-	0.1161(7)
16.04(2) [*]	1	1	0.029(3)	0.393	-	0.0100(3)
19.71(2)	2	0	0.708(11)	104	-	0.439(4)
20.23(2)	3	1	0.406(6)	6.600	-	0.335(4)
22.49(2)	2	1	0.208(19)	0.840	-	0.104(4)
24.34(2) [*]	0	1	0.58(5)	0.66(6)	-44	0.0388(8)
25.90(3)	2	0	0.40(3)	195	-	0.250(5)
27.28(3)	1	0	1.11(3)	680	-	0.416(4)
28.79(3) [*]	1	1	5.7(6)	0.478(15)	-13	0.1656(19)
28.89(3)	0	1	1.85(10)	4.0(4)	-64	0.158(4)
29.49(3)	2	0	0.894(19)	350	-	0.557(7)
31.60(3)	3	1	0.268(8)	8.748	-	0.227(5)
32.17(3)	3	1	0.241(8)	2.700	-	0.194(5)
35.04(3)	3	1	0.221(12)	3.210	-	0.181(8)
37.84(4)	3	1	0.245(11)	12.000	-	0.210(8)
42.47(4)	1	1	0.521(25)	6.210	-	0.180(3)
43.30(4)	3	1	0.34(3)	1.500	-	0.242(14)
47.22(5)	1	1	0.86(4)	34.900	-	0.313(5)
47.79(5) ^{*+}	3	1	2.427	0.025	-	0.021
49.97(5)	3	1	0.257(11)	7.200	-	0.217(8)

+) Parameters from JEFF-3.3

@) Parameters from CENDL-3.2

†) Parameters from INDEN

•) Energy discrepancy

*) Not included in CENDL-3.2

E (keV)	J	ℓ	Γ_γ (eV)	Γ_n (eV)	Corr. (%)	K_γ (eV)
51.07(5)	1	1	0.46(3)	45.100	-	0.169(4)
53.63(5)	1	1	0.93(4)	20.000	-	0.334(5)
55.27(5) ⁺	0	1	0.520	31.127	-	0.064
55.87(6) ⁺	0	1	0.520	48.510	-	0.064
62.39(6) [@]	2	1	0.300	0.300	-	0.097
62.63(6)	1	1	1.43(7)	4.817	-	0.414(5)
64.83(6)	2	1	0.53(3)	4.657	-	0.294(8)
65.51(7)	2	0	0.48(4)	4017	-	0.300(17)
66.50(7)	3	0	0.308(17)	13.698	-	0.263(12)
67.15(7)	2	1	0.44(3)	15.029	-	0.269(9)
69.51(7)	1	1	0.92(6)	50.100	-	0.340(14)
69.81(7)	2	1	1.86(11)	3.112	-	0.728(6)
73.01(7)	1	1	0.59(4)	76.000	-	0.219(6)
73.60(7)	1	0	1.04(9)	1200	-	0.391(12)
74.43(7)	3	1	0.89(6)	1.76(18)	-49	0.52(7)
74.53(7)	2	0	0.48(5)	1000	-	0.301(18)
75.67(8) [†]	2	1	2.102	0.184	-	0.106
76.62(8) ⁺	1	1	0.494	0.287	-	0.681
85.80(9)	1	1	0.193(16)	3.272	-	0.455(6)
86.99(9)	1	0	2.48(24)	6500	-	0.93(3)
89.50(9)	2	1	0.65(6)	0.42(4)	-5	0.160(4)
93.64(9)	2	0	0.82(5)	78.533	-	0.507(21)
94.56(9)	2	0	0.40(4)	501	-	0.248(14)
95.98(10) ⁺ @	0	1	0.890	33446	-	0.111
98.40(10)	2	1	1.30(6)	19.570	-	0.759(21)

+) Parameters from JEFF-3.3

@) Parameters from CENDL-3.2

†) Parameters from INDEN

●) Energy discrepancy

*) Not included in CENDL-3.2

