Registered Office
Herrmann-Debrouxlaan 40
1160 Brussel – Belgium

Foundation of Public Utility
VAT BE 406.568.867

Research Centres
Boeretang 200
2400 Mol – Belgium

Chemin du Cyclotron 6
1348 Ottignies-Louvain-la-Neuve – Belgium

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Authors*
Alexey Stankovskiy

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Improvement of proton induced nuclear data for key isotopes needed to design MYRRHA Phase I (MINERVA)

Author:
Maureen Ciccarelli

Supervisor:
Dr. Alexey Stankovskiy

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Abstract

SCK CEN is currently designing phase I of the Multi-purpose hYbrid Research Reactor for High-tech Applications (MYRRHA). It will be an innovative nuclear reactor driven by an linear accelerator and able to operate in both sub-critical and critical modes. Such high-energy proton accelerators are sources of radiation along the whole length of the beam line due to beam interactions with materials at targeted positions and beam losses along the line. Shielding is then required to protect workers, public, environment and equipment against prompt radiation. In addition, accurate activation calculations are important in order to properly estimate the doses received by these workers, public, environment and equipment. Reliable nuclear data are then needed in order to perform these activation and shielding calculations.

This report will present analysis of existing proton-induced nuclear data for nuclides important for the project and their improvement in order to perform more accurate Monte Carlo transport code calculations for activation and shielding.

Acknowledgements

Performing my master thesis in SCK CEN on MYRRHA project was a pleasure and a great and rewarding experience.

I would like to especially express my gratitude to my mentor Alexey Stankovskiy for the guidance, useful comments, remarks and engagement through the learning process of the master thesis despite the special circumstances of this year 2020.

Furthermore, I would like to thank my co-mentor Yurdunaz Celik for introducing me to the topic as well for the support on the way. Also I would like to thank all Nuclear System Physics group for welcoming me on their service.

In the end, I am grateful to Lille University's professors, in particular the master's director Philippe Carrez for the sharing of their knowledge and for giving me the opportunity to perform a long-term internship on such an interesting topic during my academic studies.

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1 Introduction

MYRRHA (Multi-purpose hYbrid Research Reactor for Hightech Application) is a flexible experimental lead-bismuth cooled accelerator driven system currently being developed at SCK CEN [1]. The Accelerator-Driven System (ADS) concept (600MeV, 4mA) has been chosen as a basis for this irradiation facility. The first phase of the MYRRHA project focusing the construction of the 100MeV proton Linear Accelerator (linac) and the target facilities is underway. Primary purpose of this phase, called MYRRHA Isotopes productioN coupling the linEar acceleRator to the Versatile proton target fAcility (MINERVA), is to test the accelerator to provide the regime necessary to operate reactor.

During routine operation of the accelerator, there will occur beam losses on the beam line structures. The interactions of the primary protons with the structural materials will produce radiation field combining secondary particles (depending on the incident beam energy) along the whole length of the beam line. Radiation shielding is required in order to protect workers, public, environment and equipment against prompt and delayed radiation. Attenuation of radiation to acceptable levels (below dose rate limit) can be achieved using shielding layers of appropriate materials and thickness. In addition, activation calculations are required to correctly estimate the doses. More accurate shielding design and activation calculations are based on calculation codes simulating the particle transport in matter and reliable nuclear data.

The purpose of this Master thesis is to analyze then improve proton-induced nuclear data used for activation and transport calculations for MINERVA. This internship focused on carbon (a candidate material for the core structure of the dump) data (^{12}C and ^{13}C) and in the future, the work will need to be extended to other key isotopes needed to design MYRRHA phase I, MINERVA.

In this Master thesis, a presentation of the context is given in order to understand the stakes of the subject. A theoretical framework about proton interaction with matter is then given in section 3.

Numerical tools used to perform analysis and improvement of the nuclear data are the nuclear reaction simulator TALYS [2], MCNP radiation transport code [3], the data processing code NJOY [4] and utilities created to process data stored in ENDF-6 nuclear data format [5]. These numerical tools will be detailed in section 4 before presenting different database and experimental benchmarks needed to carry the work of the thesis.

In the section 7, results of experimental nuclei production cross section and neutron production analysis are presented and discussed.

At the end of the internship at SCK CEN, new data files for carbon isotopes have been created using adequate residual nuclei production cross section data for activation calculations and adjusted neutron spectra data for shielding calculations.

2 Context

2.1 SCK CEN

The Nuclear Research Center (SCK CEN) located in Mol was founded in 1952 to study the applications of nuclear energy. Since then, they have expanded their knowledge to a wide range of research fields and it is now one of the largest research centres in Belgium. SCK CEN focuses on different topics in nuclear physics [1]:

- Nuclear safety and radiation protection
- Medical and industrial applications of radiation
- The back end of the nuclear fuel cycle (nuclear reprocessing and management of radioactive waste)
- Nuclear decommissioning and decontamination on nuclear sites
- The fight against nuclear proliferation

I performed my Master thesis in the Institute of Advanced Nuclear Systems, in the expertise group "Nuclear Systems Physics" (NSP). This institute develops knowledge about the technological aspects of innovative nuclear reactors and designs, constructs and operates experimental assemblies for various projects. The project I worked on during the internship is MYRRHA phase I (MINERVA), detailed in the following section.

2.2 MYRRHA

The Multi-purpose hYbrid Research Reactor fir High-tech Applications (MYRRHA) is the world's first large scale Accelerator Driven System (ADS) project at power levels scalable to industrial system. MYRRHA offers unparalleled research opportunities in spent nuclear fuel, nuclear medicine and fundamental and applied physics. It consists of a subcritical nuclear reactor driven by a high power linear accelerator. MYRRHA consists of four major components:

- the Linear Accelerator (linac) that will provide protons at an energy of 600MeV to the reactor
- the lead-bismuth eutectic (LBE) cooled reactor
- the Proton Target Facility that will ensure the production of radioisotopes and research into several fields
- the Fusion Target Station, a flexible, nuclear fusion oriented research infrastructure designed to handle proton beam of 4mA and 100MeV

With the subcritical concentration of fission material, the nuclear reaction is sustained by the particle accelerator only. Turning off the proton beam results in an immediate and safe halt of the nuclear reaction. All these elements make the system more safe, easily controllable and allow an efficient use of the fissile material which will reduce radioactive waste. [6]

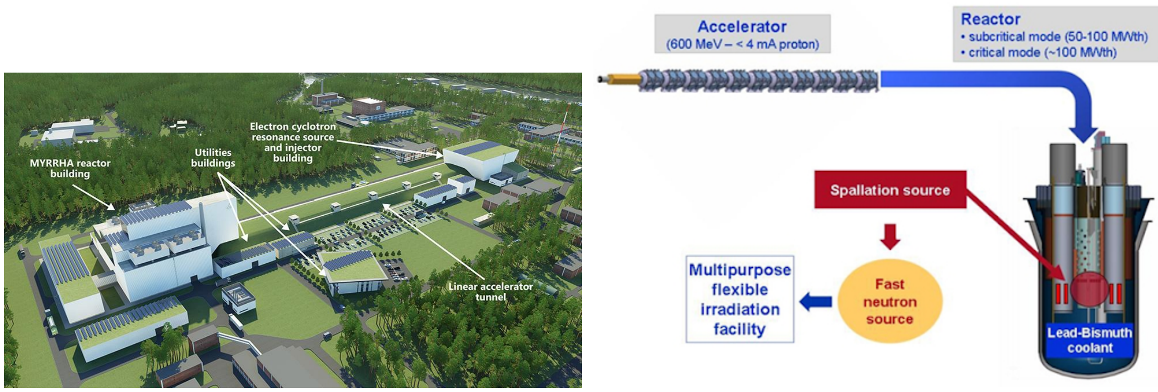


Figure 1: MYRRHA Reactor Project

MYRRHA will be constructed in three phases:

- **Phase I:** Design and construction of the first linac section (up to 100MeV), the related Proton Target Facility and the Fusion Target Station
- **Phase II:** Extension of the 100MeV linac to 600MeV required to drive the reactor
- **Phase III:** Construction of the reactor cooled by lead-bismuth eutectic (LBE) and with maximum thermal power output of 100MW

2.2.1 MINERVA

MINERVA is the first phase of the three-phase implementation plan of MYRRHA ongoing and scheduled for completion in 2026. It consists in the construction of the linear particle accelerator's first phase (up to 100MeV), the Proton Target Facility (PTF) and the Fusion Target Station. The final linac will be 400m-long and provide protons at energy of 600MeV to the reactor. At the end of the linac, the 4mA proton beam is injected into the reactor, generating a flux of fast neutrons through spallation. Simultaneously, protons are also fed to the multi-purpose Proton Target Facility and to the Fusion Target Station. First phase consists in a linac accelerating protons up to 100MeV and the confirmation of the linac's operational reliability required later to drive the reactor with the 600MeV proton beam. Proton Target Facility will ensure the production of medical radioisotopes and will provide resources for fundamental and applied research in physics as well as for material research. Fusion Target Station (FTS) is a flexible, nuclear fusion oriented research infrastructure designed to handle a proton beam of 4mA and 100MeV where materials for fusion reactors will be tested. [7]

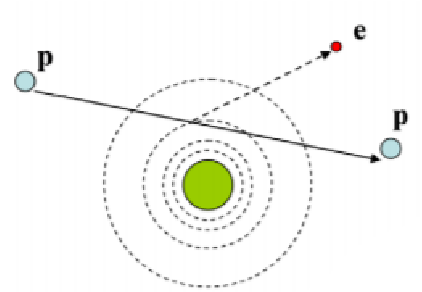
3 Theoretical framework

Proton interaction with matter

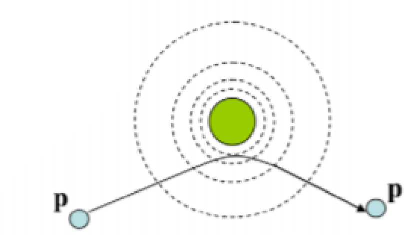
In order to understand why shielding and activation calculations are crucial for MYRRHA design, it is important to be aware of the physical phenomena occurring when accelerated protons interact with matter. [8]

The proton has three main types of interactions with matter:

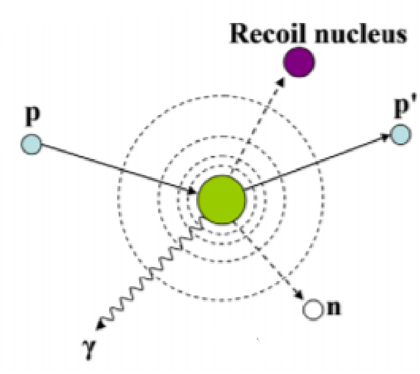
- Inelastic Coulomb scattering with atomic electrons
- Elastic Coulomb scattering with atomic nuclei
- Non-elastic nuclear interaction



(a) Inelastic Coulomb interaction with atomic electrons



(b) Elastic Coulomb scattering with atomic nuclei



(c) Non-elastic nuclear interaction

Figure 2: Main interactions of a proton with matter

When a charged particle penetrates in matter, it will interact with them through electromagnetic force. If the particle is a proton, an alpha particle or any other charged hadron, it can also undergo a nuclear interaction and this will also be discussed later on this section. Since the energy of accelerated proton used to pilot MYRRHA reactor is large compared to the binding energy of the electrons in the atom, matter can be approximated as a mixture of free electrons and nuclei at rest. Proton will then feel the electromagnetic fields of the electrons and the nuclei and in this way, will undergo elastic collisions with these objects. While colliding with the electrons and the nuclei, proton will transfer some of its energy and change directory.

Energy loss of charged particle due electromagnetic force

Since protons are way heavier then electrons (m_p= 1.0073 u.m.a , m_e= 0.0005 u.m.a), **inelastic Coulomb interaction** with atomic electrons won't change much the trajectory of the proton but it will transfer a large amount of energy to the electrons which cause the limited range of protons in matter. With this transfer, some of the molecules of the materials will be excited but the most important effect is ionization. Most of the electrons received a very small amount of energy but some of them will acquire sufficient energy to travel macroscopic distances in matter. The range of secondary electrons is less than 1mm and the most of the dose is absorbed locally. The continuous slowing down of the proton by inelastic Coulomb interaction with atomic electrons is quantified by the proton stopping power which is defined as the energy loss per unit track length of the proton (dE/dx) and is given by the Bethe-Bloch equation (1). Eventually the proton will have lost all its kinetic energy and will stop its trajectory. Proton has the specificity to lose most of its energy at the end of its trajectory in a region called "Bragg peak" before dropping to zero. This peak depends of the energy of the proton. It is well pronounced at 100MeV but for 600MeV proton this effect is really small because at higher energies more nuclear interactions occur where the energy is transferred via non-elastic interactions to recoil nuclei which are stopped locally.

$$\frac{dE}{dx} = \rho \frac{Z_{nucl}}{A_r}(0.307MeVcm^2/g)\frac{Z^2}{\beta^2}[\frac{1}{2}ln(\frac{2m_e c^{22}\gamma^2 T_{max}}{I^2} - \beta^2 - \frac{\delta(\beta)}{2}] \quad (1)$$

Where:

- dE/dx = energy loss of particle per unit length
- Z = charge of the particle divided by the proton charge
- c = velocity of light
- β, γ = relativistic parameters
- ρ = density of the material
- Z_{nucl} = dimensionless charge of the nuclei
- A_r = relative atomic weight
- I = mean excitation energy in eV (usually determined experimentally, typically around 10eV times Z_{nucl})
- T_{max} = maximum energy transfer to the electron
- $\delta(\beta)$ = density-dependent term that attenuates the logarithmic rise of the cross section at very high energy

In contrast, a proton is much lighter than most nuclei and will lose a little energy when colliding with atomic nuclei. However due to the large mass of the nucleus it will experience repulsive **elastic Coulomb interaction** which deflects the proton from its original trajectory and can even bounce backwards.

As a results, most of the energy loss of the proton penetrating matter is due to inelastic Coulomb interaction with atomic electrons while most of the change of trajectory is due to elastic Coulomb scattering with atomic nuclei.

Non-elastic nuclear reactions between protons and the atomic nucleus are less frequent but have a much more profound effect in terms of the fate of an individual proton. It occurs at higher energies and may produce secondary particles. Secondary nuclei usually stop close to the interaction point while other charged particles such as protons may go relatively far. The higher the mass of the charged particle is, the shorter its range will be. For a 100-MeV incident proton beam, non-elastic nuclear interaction will generate secondary charged particles such as proton (p,p), deuteron (p,d), alpha particle (p,α) or recoil protons (p,p'). These secondary products are absorbed locally due to their charges. However, their interaction may generate neutral particles such as neutron (p,n) or γ-rays (p,γ) (for example, on a ^{12}C target: 12C(p,γ)13N). These neutral particles have a longer range than charged particles and are more difficult to stop.

Interactions of particles in matter due to the Strong Force

At high energy all hadrons, on average, will undergo a nuclear interaction after a distance approximately equal to the hadronic interaction length. A very high-energy proton will lose a few MeV per cm due to ionisation in a solid, and the range of proton due to the energy loss will be larger than the hadronic interaction length (10-100cm in solids). The proton will most of the time undergo a nuclear interaction before it has lost all its energy in ionisation. If a high-energy proton collides with a very heavy nucleus (many more neutrons than protons) the nucleus will be broken up. Since a heavy nucleus have a lot more neutrons than protons, the fragments will quickly expel their excess neutrons and a large number of secondary neutrons are produced. This process called **spallation** is used to produce neutrons necessary to maintain fission reaction in MYRRHA.

4 Numerical tools

4.1 TALYS

TALYS is an open source software package for the simulation of nuclear reactions. Created in 1998 at NRG Petten (the Netherlands), CEA (France), University of Brussels (Belgium) and IAEA (Vienna). TALYS is a nuclear reaction program developed in order to regroup and provide a complete and accurate simulation of nuclear reactions from the neutron unresolved resonance range up to intermediate energies. This enables to evaluate nuclear reactions involving neutron, photons, protons, detuerons, tritons, ^{3}He- and alpha- particles in the energy range of 1keV to 200MeV for target nuclides of mass 12 and heavier.

TALYS is a single code system using nuclear reaction models and it has two main purposes, strongly connected:

- Nuclear physics tool
- Nuclear data tool

Nuclear physics tool can be used for the analysis of nuclear reaction experiments. By crossing existing experimental database and known theory, TALYS gives insight in the fundamental interaction between particles and nuclei, and precise measurements enable to constrain models. These resulting nuclear models then permit to have an indication of the reliability of measurements, when they are considered to have sufficient predictive power. Second function of TALYS is to generate nuclear data for all open reaction channels beyond the neutron resonance region either in default mode, when no measurements are available, or after adjusting parameters of various reaction models by using experimental data when available. The resulting nuclear data library which consists of a set of files provides essential information for existing and new nuclear technologies and can be used for several applications such as conventional and innovative nuclear reactors. [9]

4.2 Monte Carlo N-Particle transport code

MCNP is a general-purpose Monte Carlo N-Particle Transport Code developed in 1957 by Los Alamos National Laboratory, USA. The code is designed to track many particle types over broad ranges of energies. It can be used for neutron, photon, electron or coupled neutron/photon/charged particles transport using evaluated nuclear data libraries for low-energy interaction probabilities.. MCNP is used in radiation protection and dosimetry, radiation shielding, radiography, medical physics, nuclear criticality safety, Detector Design and analysis, nuclear oil well logging, Accelerator target design, Fission and fusion reactor design, decontamination and decommissioning. The code treats an arbitrary three-dimensional configuration of materials in geometric cells bounded by first- and second-degree surfaces and fourth-degree elliptical tori. It uses a Monte Carlo method to simulate particles transportation through matter. This Monte Carlo method involves following source-particles and secondary particles generated since their "creation" until their "death", i.e. until they are absorbed by matter of leave the studied area. This approach enables to determine physical values, such as particle-flux or energy deposit. This is a statistical method using random walks and requires nuclear data. The more histories (number of source particles generated), the less statistical uncertainty.

All calculations presented in this work were performed using MCNP-6.2. [3]

4.3 NJOY

The NJOY nuclear data processing system is a modular computer code package used to convert evaluated nuclear data in the ENDF format (see section 4.4) into libraries useful for applications calculations. It processes nuclear cross sections and related quantities to serve applications such as continuous-energy Monte Carlo (MCNP, Serpent, PHITS), deterministic transport codes (PARTISN, ANISN, DORT) and reactor lattice codes (WIMS, EPRI). [4]

During the master thesis, NJOY2016, was used to convert ENDF-6 format into ACE, a compact ENDF format, for the Los Alamos continuous-energy Monte Carlo MCNP and MCNPX codes. All the cross sections are represented on a union grid for linear interpolation.

4.4 ENDF-6 format and useful utilities

4.4.1 ENDF-6 format

ENDF system was developed for the storage and retrieval of evaluated nuclear data to be used for applications of nuclear technology (originally for use in the US national nuclear data files). Format describes how the data are arranged in the libraries and give the formulas needed to reconstruct physical quantities from the parameters in the library.

The internationally agreed format for data files of evaluated nuclear reaction data is ENDF-6. The ENDF formats and libraries are maintained by the National Nuclear Data Center (NNDC) on behalf of the Cross Section Evaluation Working Group (CSEWG) that make the decisions.

ENDF format first provided representations for neutron reaction data, fission-product yield data, neutron thermal scattering data, photo-atomic interaction data and others. Version 6 (ENDF-6) now allows higher incident energies and permits the inclusion of data for nuclear reactions induced by photons and charged particles. This work will focus on reaction cross sections and product energy-angle distributions as detailed in section 5.

Reaction cross sections file (MF=3)

Cross section of a reaction is the probability that this reaction will occur. Residual nuclei production cross sections are tabulated in nuclear data files. These files are recorded in ENDF-6 format contains different quantities such as standard material charge and mass parameters, frame, interpolation scheme, reactions threshold, cross section for a particular reaction given as energy-cross section pairs, ...

ENDF-6 file consists of sections called "files" (MF) and sub-sections called "sections" (MT). Because originally format was developed for reactor applications until 20MeV, it does not contain enough number of section identifiers (MT) to store various residual products (for example, at 600MeV one can have around 1000 of different products). Therefore, the residual production cross section is a product of total non-elastic reaction cross section (MF=3 MT=5) multiplied by residual product yield per average incident particle stored in MF=6 MT=5.

Focus in this work will be residual nuclei production and nonelastic cross sections in order to perform activation calculation (section 5).

Product energy-angle distributions file (MF=6)

This file is provided to represent the distribution of reaction products in energy and angle. A section of File 6 is divided into one subsection by product. Every subsection for given product starts with yield then the energy-angle distribution follows. Every reaction is defined by giving the production cross section for each reaction product (in barns/steradian) assuming azimuthal symmetry:

$$\sigma_i(\mu, E, E') = \sigma(E) y_i(E) f_i(\mu, E, E')/2\pi \tag{2}$$

Where:

- i denotes one particular product
- E is the incident energy
- E' is the energy of the product emitted with cosine μ
- σ(E) is the interaction cross section (MF=3)
- y_i(E) is the product yield or multiplicity for the particle i
- f_i is the normalized distribution with units (eV.unit-cosine)$^{-1}$ where

$$\int dE' \int d\mu f_i(\mu, E, E') = 1 \tag{3}$$

The individual products are identified by their standard material charge and mass parameters. The different f_i representations are defined by a flag to distinguish different representations called LAW:

- LAW=0: unkown distribution
- LAW=1: continuum energy-angle distribution
- LAW=2: two-body reaction anglar distribution
- LAW=3: isotropic two-body distribution
- LAW=4: recoil distribution of a two-body reaction
- LAW=5: charged-particle elastic scattering
- LAW=6: n-boy particle elastic scattering

- LAW=7: laboratory angle-energy law

Nuclear data files presented in existing nuclear data libraries TENDL and JENDL-4.0/HE files analysed in this report have respectively LAW=1 and LAW=7 representation then we only will focus on these in the following paragraphs.

4.4.2 Continuum energy-angle distributions (LAW=1)

TENDL is given with LAW=1 representation providing a compact representation of coupled energy-angle distribution for neutrons and chaged particles developed by C. Kalbach and F. Mann based on systematics of experimental and evaluated data [10]. Kalbach-Mann systematics is an extended form and is a powerful representation of outgoing neutron and charged particle distribution in high energy reactions. The general shape of the f_i function was later shown to come directly out of preequilibrium theory by M. Chadwick[11]. Kalbach distribution for a reaction of the form:

$$A + a \rightarrow C \rightarrow B + b \tag{4}$$

can be represented by

$$f(\mu_b, E_a, E_b) = f_0(E_a, E_b) \left\{ \frac{a}{sinh(a)} \left[cosh(a\nu_b) + rsinh(a\mu_b) \right] \right\} \tag{5}$$

where

- Ea is the energy of the incident projectile in the laboratory system
- Eb is the energy of the emitted particle in the center-of-mass system
- μ_b is the cosine of scattering angle of the emitted particle b in the center-of-mass system

Parameters $r(E_a, E_b)$ and a(E_a, E_b) are respectively the precompound fraction as given by the evaluator, and a parameterized function that depends on emission energy, incident energy and particle type. f_0 is the total emission probability.

The Kalbach representation is used in many nuclear data libraries and is an important factor in the new high-energy libaries up to 200MeV now being constructed for accelerator applications.

4.4.3 Laboratory angle-energy law (LAW=7)

Since experiments normally give spectra at various fixed angles, some evaluators prefer to enter data sorted according to E, μ and E' in laboratory system where E is incident energy, μ is cosine of scattering angle of emitted particle and E' the energy of emitted particle. Both μ and E' must cover the entire angle-energy range open to the emitted particle.

4.4.4 Utilities

One of the stake to analyze residual cross section and neutron production data from nuclear data libraries was the processing of ENDF-6 format into readable format (and vice versa). For this purpose, several utilities have been developed at NSP:

- *breakdown.exe*
- *assembly.exe*
- *nspectra_differential.exe*
- *integrate_spectrum_law7.exe*
- *break_and_mix.exe*

breakdown.exe decomposes ENDF-6 file into individual subsections with residual nuclei cross sections and spectra.

assembly.exe reassembles decomposed files from *breakdwon.exe* into ENDF-6 file processable with NJOY to give MCNP readable files.

nspectra_differential.exe gives spectrum for given energy and angle by doing a two-dimensional interpolation between angles and energies.

integrate_spectrum_law7.exe reads spectra from ∗.spe file, integrates them and normalizes if integral differs from 1. This utility was needed when JENDL-4.0/HE smoothing spectra presented in section 7.2.2 to assure spectra normalization before re-assemble folder and test it with MCNP.

break_and_mix.exe converts JENDL-4.0/HE neutron spectra written in LAW=7 into LAW=1 data used by other libraries such as TENDL and ENDF/B.

In addition, *checkr.exe* utility [12], provided by ENDF utility collection suite was used: it checks if an evaluated data file conforms to the ENDF format. It is used after reassemble a library and before processing it with NJOY.

5 Nuclear data libraries and experimental data

In particle transport codes, nuclear reactions can be handled either by nuclear models or by data libraries. So MCNP requires nuclear data to run calculations such as photoreaction data or interaction cross section data.

Activation calculations are simulations of the nuclear interactions of radiation with material. In MYRRHA phase I, activation calculations are needed in order to determine resulting radioactive waste produced during process and to estimate residual activity. At this purpose, reliable cross section data is needed.

Second type of calculations needed for MYRRHA design is radiation transport calculations. In order to protect workers, public, environment and equipment against prompts and delayed radiation, shielding is needed for MYRRHA. Attenuation of radiation to acceptable levels (below dose rate limit) can be achieved using shielding layers of appropriate material thickness. More accurate shielding design is based on transport calculation codes such as MCNP. These calculations need reliable data regarding neutron production from protons for accelerator applications.

MYRRHA is an Accelerator Driven System (ADS) coupling a proton accelerator to a spallation target. As accelerator protons trigger all further nuclear interactions in the system, accurate data for incident protons are extremely important. Proton-induced library will be the object of this study. The work will then focus on proton-induced library residual nuclei cross section data for activation calculations and neutron production (and spectra) for transport calculations. Carbon is candidate considered as a material for the beam dump of MYRRHA to stop protons and is a material for various carbide targets to produce medical isotopes. So study will at first be performed for carbon data but will also have to be reproduced for other key isotopes needed to design MINERVA.

To perform analysis of residual nuclei cross section and neutron production, two types of data are required: evaluated nuclear data libraries and experimental data.

Nuclear data are taken from evaluated nuclear data files contained in nuclear data libraries. The libraries are developed using prediction of nuclear models and experimental measurements. Several libraries exist and can present differences due to different evaluated nuclear data sources or different setup arrangements (for example, same experiment performed at different temperatures).

During the internship, three nuclear data libraries have been used: JENDL-4.0/HE [13], TENDL-2017 [14] and TENDL-2019 [15]. JENDL-4.0/HE is an extended version of JENDL-4.0 [16] up to 200MeV for a number of nuclei which are relevant to high-energy (HE) applications. It also includes evaluated cross sections for incident protons up to the same energy. This library provides double-differential cross sections for emitted neutrons, protons, deuterons, tritons, helium-3 nuclei, alpha-particles and gamma-rays. It also includes production cross sections of residual nuclei (total, total-reaction and elastic scattering). TENDL [2] is a nuclear data library which provides the output of the TALYS nuclear model code system for direct use in both basic physics and applications. TENDL-2017 and TENDL-2019 are respectively 9^{th} and 10^{th} version of TENDL, based on both default and adjusted TALYS calculations and data from other source.

Experimental data for residual nuclei cross section production and energy-angle distribution (spectra) are taken from EXFOR library, the most comprehensive compilation of experimental nuclear reaction data. It contains cross sections and other nuclear reaction quantities induced by neutron, charged-particle and photon beams. [17]

6 Experimental benchmark

Experimental benchmarks have been used to analyse MCNP calculations results. Since the focus of this work is proton-induced library data files for stable carbon isotopes, benchmark experiments were all using proton-beam on carbon target.
Adustment of neutron spectra data have been tested with a thin carbon target (5mm) hit by a 140-MeV proton beam [18] and with two thick targets, a 52-MeV proton beam on a 2.14cm carbon target [19] and 113-MeV proton beam on a 5.83cm carbon target [20].

7 Results and discussion

7.1 Experimental residual nuclei production cross sections data analysis

Activation calculations need residual production knowledge. To improve proton induced nuclear data, analysis of residual nuclei cross sections is necessary to chose best fit with experimental data or rescale existing evaluated data if necessary. Experimental data have been compared with existing nuclear data cross sections and with TALYS calculation for ^{12}C, ^{13}C and ^{nat}C. JANIS database[21] also provides cross section data for JENDL-4.0/HE and TENDL-2017. These data will also be compared with experimental data. Results are presented in the form of energy dependence of residual production cross sections, called excitation functions.

For a better visibility, Figure 3 shows legend of nuclear data libraries and TALYS calculations of the following graphs of the section. Grey line and grey zone represent TALYS calculations average performed on 500 run with error bars.

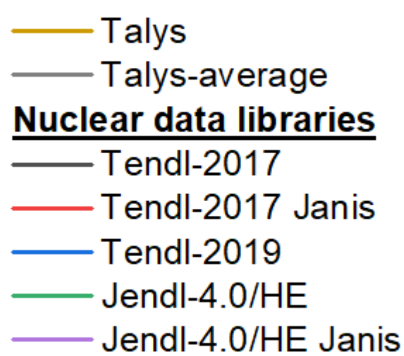


Figure 3: Nuclear data library and TALYS calculation legend for excitation functions

7.1.1 Carbon 12

Figure 4 shows total nonelastic production cross sections for proton beam on a ^{12}C target. Excitation functions for residual nuclei production for a proton beam on a ^{12}C target are presented in Figures 5 to 9.

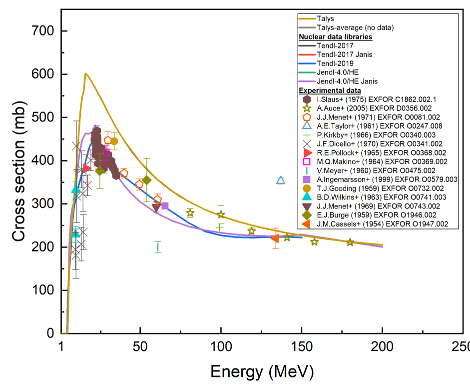
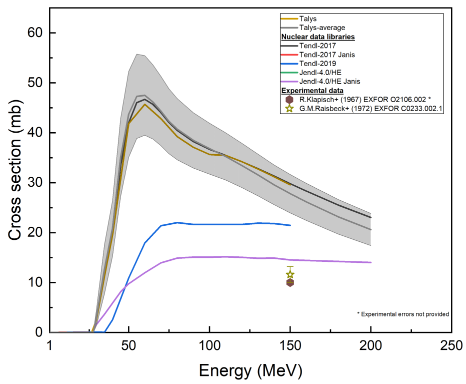
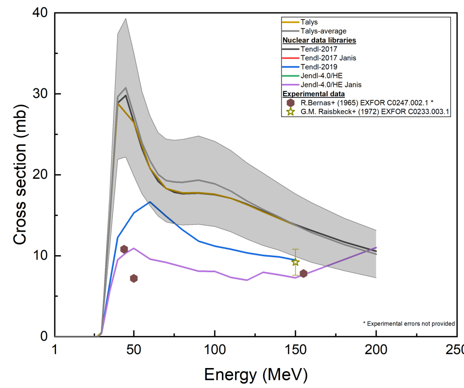


Figure 4: Total nonelastic production cross sections for proton beam on a ^{12}C target

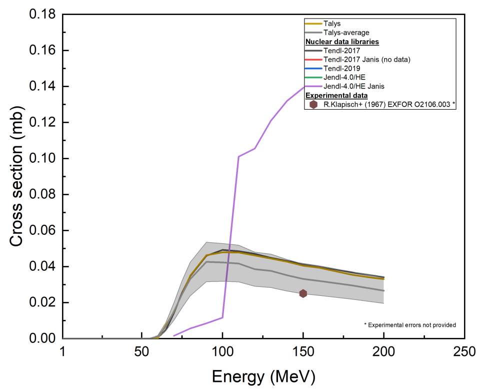
It is noted that JENDL-4.0/HE data taken directly on JAEA website is same that JENDL-4.0/HE data from JANIS database. However, TENDL-2017 from JANIS has same data that TENDL-2019 on website, not TENDL-2017. Analysis of TENDL-2019 original file shows that data are actually borrowed from ENDF/B library, an old evaluation originated from 1996. In TENDL-2017 website, 3 files are available: first one is also file taken from ENDF/B, second and third ones are results of TALYS calculations stored in different formats. JANIS database is using the first file explaining that TENDL-2019 data and TENDL-2017 from JANIS are same.



(a) $^{12}C(P,X)^{6}Li$ reaction



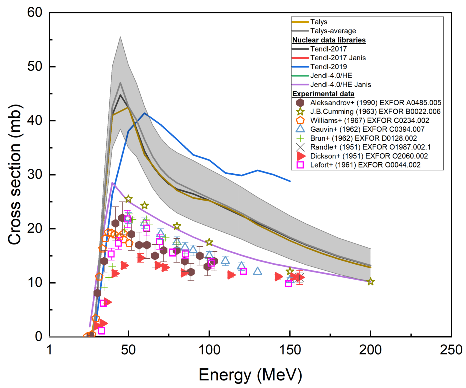
(b) $^{12}C(P,X)^{7}Li$ reaction



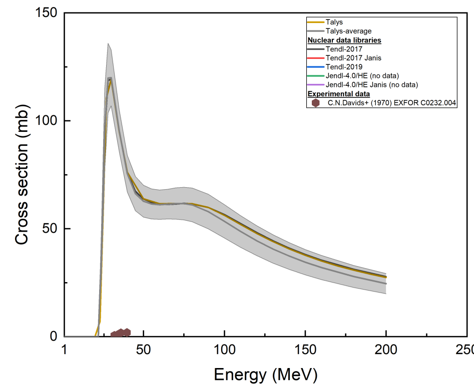
(c) $^{12}C(P,X)^{9}Li$ reaction

Figure 5: Experimental residual Lithium production cross-section with nuclear data libraries TENDL and JENDL-4.0/HE, and Talys calculations

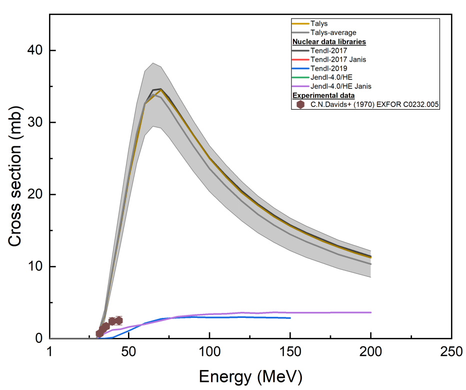
Comparison between experimental data, existing nuclear data libraries and TALYS calculation show a better fit with JENDL4.0/HE for ^{6}Li (Figure 5a), ^{7}Be (Figure 6a), ^{9}Be (Figure 6c), ^{11}B (Figure 7b), ^{3}H (Figure 9b) and proton production (Figure 9a). Figure 5b shows a good match between experimental data and TENDL-2019 for ^{7}Li production and TALYS calculation will be taken for ^{9}Li (Figure 5c) and ^{10}Be (Figure 6d).



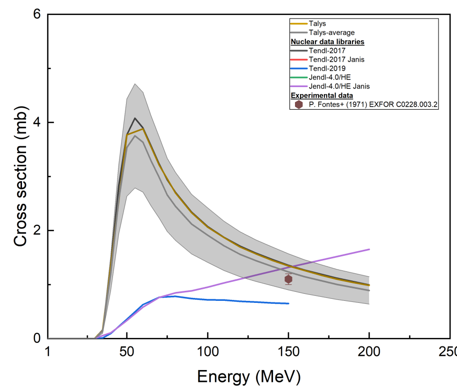
(a) $^{12}C(P,X)^{7}Be$ reaction



(b) $^{12}C(P,X)^{8}Be$ reaction



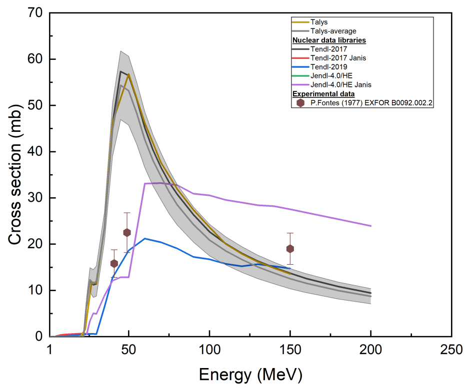
(c) $^{12}C(P,X)^{9}Be$ reaction



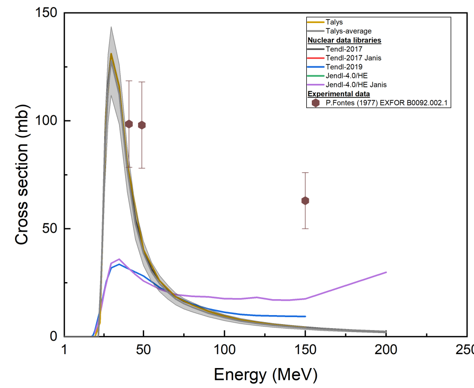
(d) $^{12}C(P,X)^{10}Be$ reaction

Figure 6: Experimental residual Beryllium production cross-section with nuclear data libraries TENDL and JENDL-4.0/HE, and Talys calculations

Regarding ^{10}B (Figure 7a), ^{11}B (Figure 7b), ^{10}C (Figure 8a) and ^{11}C (Figure 8b) a rescale of existing nuclear data library is necessary. The rescale has two purposes: first is not underestimating experimental data. By underestimate nuclei production cross section data, doses from these nuclides when using data in activation/dose calculation will also be underestimated which is not acceptable for safety reasons. Second goal of rescaling is having a data closer to experiment. Rescale factors have been calculated taking account of experimental errors by using chi-square test. Data taken for ^{10}B, ^{11}B, ^{10}C and ^{11}C are respectively 1.23853 $*$ TENDL-2019, 3.43815 $*$ JENDL-4.0/HE, 2.55537 $*$ TALYS and 1.09169 $*$ JENDL-4.0/HE.

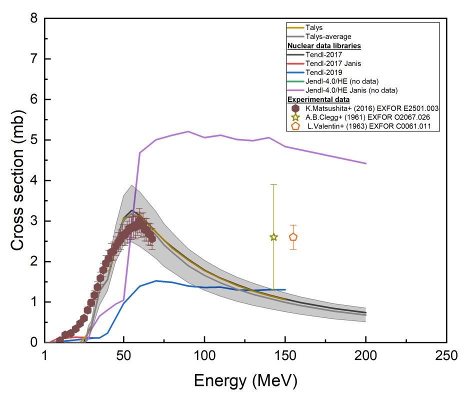


(a) $^{12}C(P,X)^{10}B$ reaction

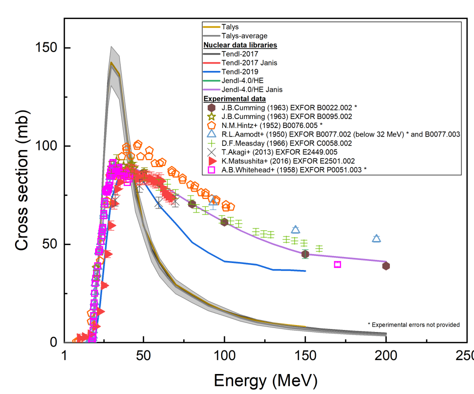


(b) $^{12}C(P,X)^{11}B$ reaction

Figure 7: Experimental residual Boron production cross-section with nuclear data libraries TENDL and JENDL-4.0/HE, and Talys calculations



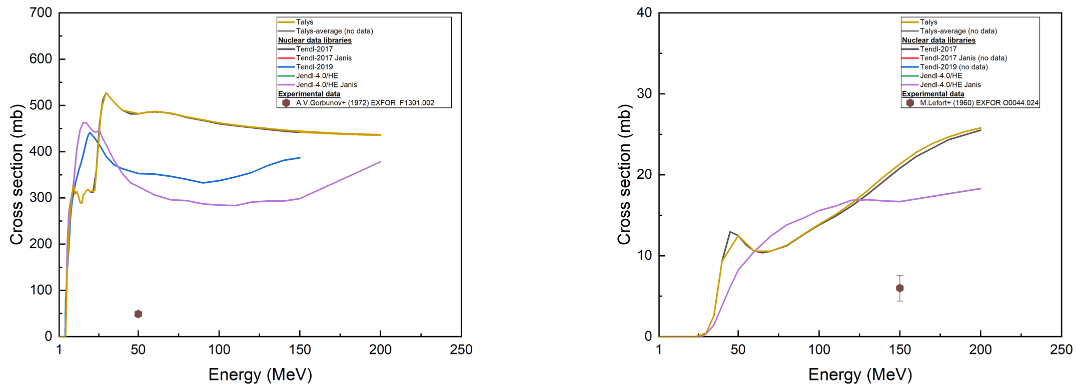
(a) $^{12}C(P,X)^{10}C$ reaction



(b) $^{12}C(P,X)^{11}C$ reaction

Figure 8: Experimental residual Carbon production cross-section with nuclear data libraries TENDL and JENDL-4.0/HE, and Talys calculations

JENDL-4.0/HE doesn't contain data for ^{8}Be production. Without JENDL-4.0/HE data, it seems that experimental data is wrong so no useful information can be obtained from Figure 6b.



(a) $^{12}C(P,X)^{1}H$ reaction: proton production **(b)** $^{12}C(P,X)^{3}H$ reaction: tritium production

Figure 9: Experimental residual Hydrogen production cross-section with nuclear data libraries TENDL and JENDL-4.0/HE, and Talys calculations

7.1.2 Carbon 13

Less experimental data is available for ^{13}C isotope than for ^{12}C and there is no experimental data for total nonelastic production.

TENDL-2019 and TENDL-2017 on JANIS doesn't have a file for ^{13}C either so comparison will be done only with JENDL-4.0/HE (on website and on JANIS), TENDL-2017 file and TALYS calculation. As for ^{12}C isotope, JENDL-4.0/HE taken directly from website and on JANIS present same results.

Excitation functions for residual nuclei production for a proton beam on a ^{13}C target are presented Figures 10 and 11. Best fit for both ^{7}Be (Figure 10) and ^{11}C (Figure 11) production is TALYS calculation.

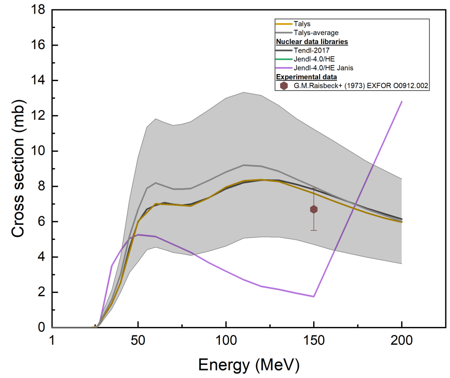


Figure 10: Experimental residual Beryllium production cross-section with nuclear data libraries TENDL and JENDL-4.0/HE, and Talys calculations: $^{13}C(P,X)^{7}Be$ reaction

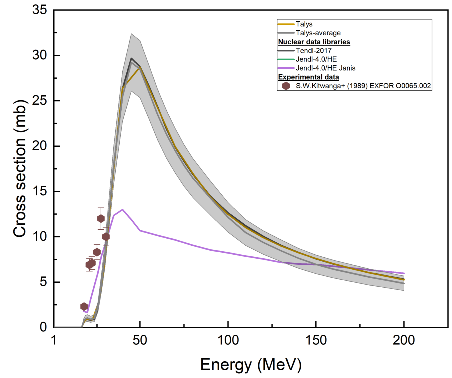


Figure 11: Experimental residual Carbon production cross-section with nuclear data libraries TENDL and JENDL-4.0/HE, and Talys calculations: ^{13}C(P,X)^{11}C reaction

7.1.3 Natural carbon

Natural carbon data aren't directly available on nuclear data libraries and are obtained using carbon isotopes abundances such as $^{nat}C(P,X)Y = 0.9893 *^{12} C(P,X)Y + 0.0107 *^{13} C(P,X)Y$. Since there isn't data available on TENDL-2017 (JANIS) and TENDL-2019 for ^{13}C, no data can be extracted either for ^{nat}C for this libraries.

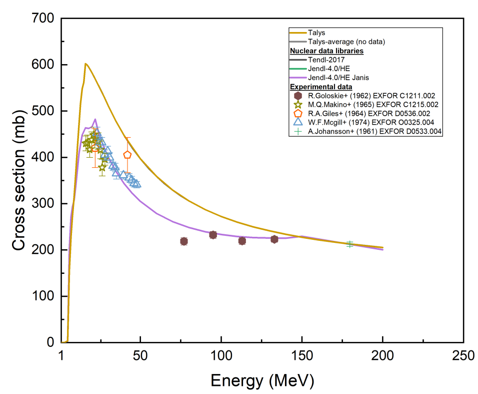
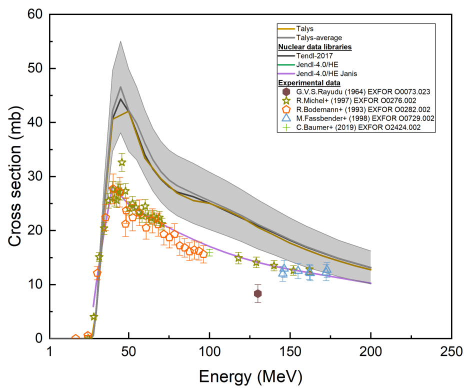


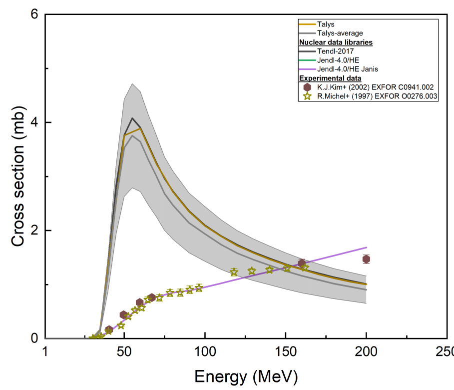
Figure 12: Total nonelastic production cross sections for proton beam on a ^{nat}C target

Figure 12 shows total nonelastic production cross sections for proton beam on a ^{nat}C target. Residual nuclei cross sections analysis for a proton beam on a ^{nat}C target are presented in Figures 13 to 15.

Best fit for Beryllium production seems to be JENDL-4.0/HE for both ^{7}Be and ^{10}Be nuclides (Figures 13a and 13b). However, behaviour for ^{10}Be experiment seems odd and papers would need to be studied. Still JENDL-4.0/HE fits best experimental data for both tritium (Figure 9b) and ^{11}C production (Figure 15) and TALYS results will be used for ^{9}Li (Figure 16).



(a) $^{nat}C(P,X)^{7}Be$ reaction



(b) $^{nat}C(P,X)^{10}Be$ reaction

Figure 13: Experimental residual Berrylium production cross-section with nuclear data libraries TENDL and JENDL-4.0/HE, and Talys calculations

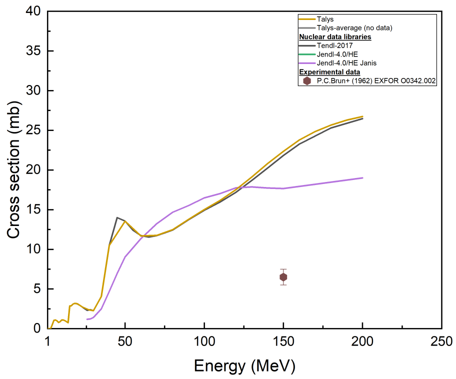


Figure 14: Experimental residual Tritium production cross-section with nuclear data libraries TENDL and JENDL-4.0/HE, and Talys calculations: $^{nat}C(P,X)^3H$ reaction

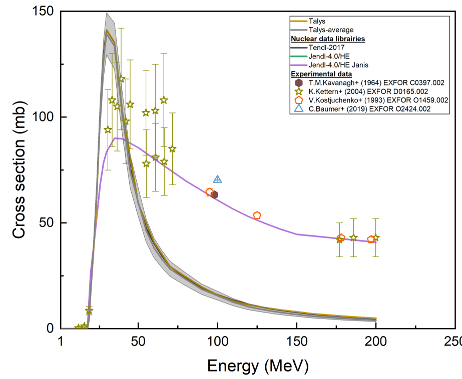


Figure 15: Experimental residual Carbon production cross-section with nuclear data libraries TENDL and JENDL-4.0/HE, and Talys calculations: $^{nat}C(P,X)^{11}C$ reaction

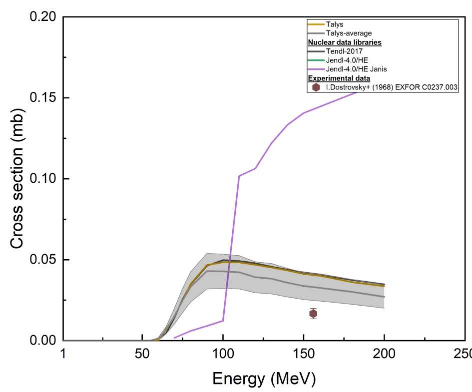


Figure 16: Experimental residual Lithium production cross-section with nuclear data libraries TENDL and JENDL-4.0/HE, and Talys calculations: $^{nat}C(P,X)^9Li$ reaction

7.2 Experimental data for neutron production analysis

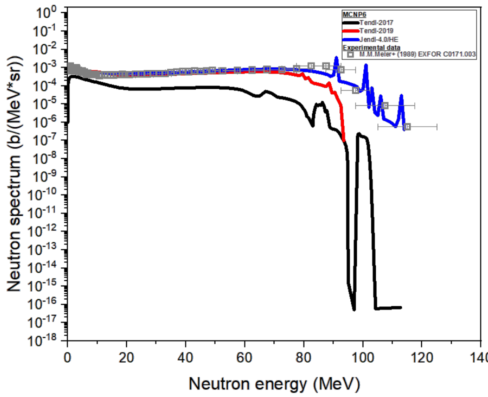
Neutron production and their spectra are important for shielding calculation and will be studied in this section. To analyse neutron spectra given by existing nuclear data libraries JENDL-4.0/HE, TENDL-2017 and TENDL-2019, several experimental data are available. 113MeV-proton beam on thick target experiment [20] by Meier et al. will be used so protons are fully stopped in the target. Secondary neutrons are produced at different proton energies although the probability of produced neutron decreases with decreasing energy.

Figure 17 shows a better fit for JENDL-4.0/HE with experimental data. However, while analyzing comparison between neutron spectra given by nuclear data libraries and Meier experiment (Figure 17), peaks at high energies on JENDL-4.0/HE spectra have been observed, mostly for low angles. These peaks are called discrete components of the spectrum. Continuum part of the spectrum is due to emission of neutrons from compound nucleus ($p +^{12} C =^{13} N$) while its discrete part is present when a single neutron is immediately kicked out before compound nucleus formation. Although presence of these peaks is then expected, their magnitude for JENDL-4.0/HE seems too high and therefore they need to be studied.

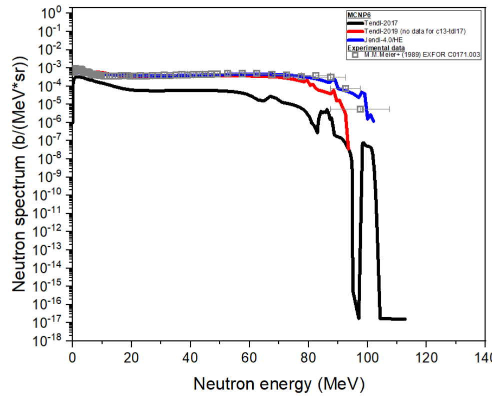
7.2.1 *break_and_mix.exe* test

As explained in section 4.4, JENDL-4.0/HE has old style representing secondary particles spectra by giving lab-frame energy spectra in defined angular bins (LAW=7) while other libraries (TENDL, ENDF/B) uses center-of-mass energy distribution integrated over angles (LAW=1). It was then necessary to develop an utility converting LAW=7 into LAW=1 data so NJOY could process JENDL-4.0/HE spectra: *break_and_mix.exe* This utility uses ENDF/B (or, in this case TENDL) file as template where secondary neutron spectra (fully or partially) are replaced by JENDL-4.0/HE data converted into LAW=1 representation for a specific energy range. *break_and_mix.exe* has been tested by using JENDL-4.0/HE data between 20- to 150-MeV data on TENDL-2017 template, processing this new library with NJOY and then running MCNP calculation.

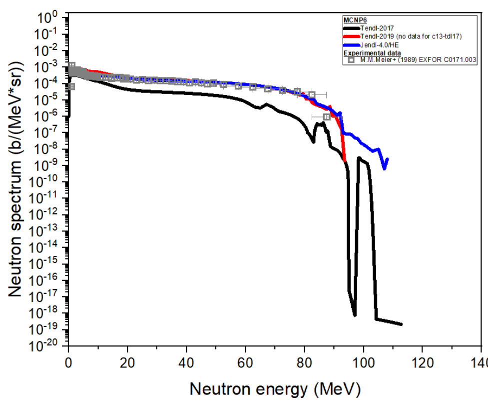
Results have been compared with TENDL-2017 (blue lines on the graphs) and JENDL-4.0/HE (red lines on the graphs) nuclear data and with experimental data for four different experiments and are presented in black lines in Figures 18 to 21. Even if break_and_mix could still be improved, a good enough match is observed with JENDL-4.0/HE processed by *break_and_mix.exe* utility to move to the next step.



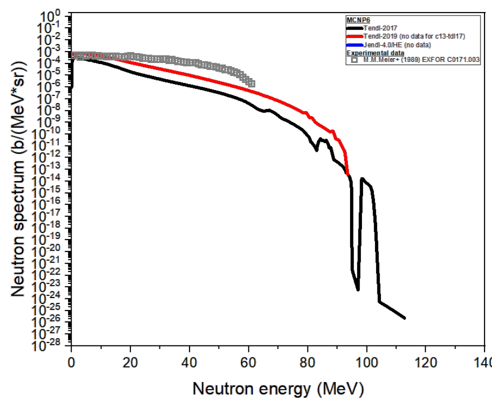
(a) at 7.5°



(b) at 30°



(c) at 60°



(d) at 150°

Figure 17: Neutron spectra comparison between existing nuclear data libraries and Meier experiment

7.2.2 Peaks impact on JENDL-4.0/HE

To study biggest impact of the peaks, they have been removed then the effect of the peaks removing has been studied.

Figure 22a and 22b show neutron production spectra from JENDL-4.0/HE for a 90 MeV proton beam on ^{12}C target in several directions (0° to 180° by ten degree angle bin) before and after spectra smoothing. While Figure 23a and 23b present neutron production spectra at 0° for several incident proton beam on ^{12}C target.

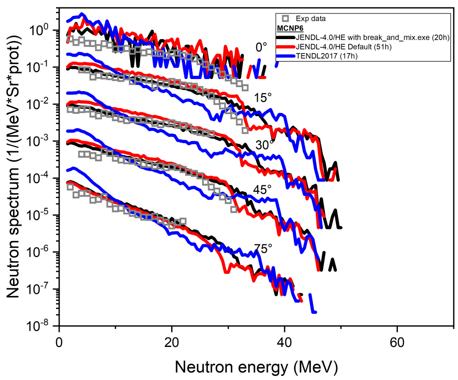


Figure 18: Neutron energy spectra at different angles from 52-MeV proton incident reactions on a 2.14cm-thick carbon target

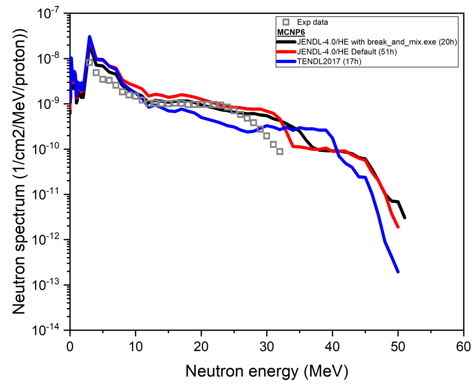


Figure 19: Neutron yield behind 46cm-thick concrete shielding (2.14cm-thick carbon target hit with a 52-MeV proton beam)

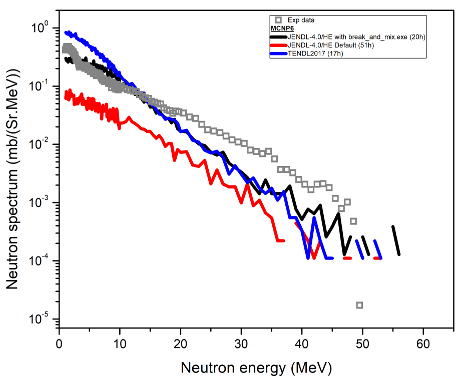


Figure 20: Neutron energy spectrum at 180 from 140-MeV proton incident on a 5mm-thick carbon target

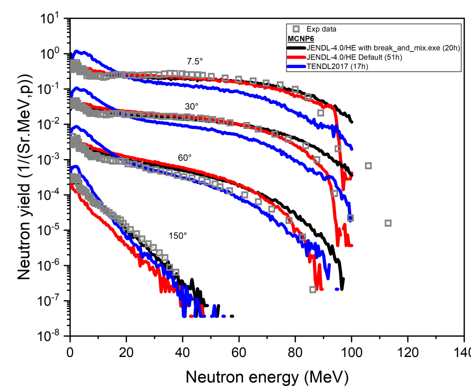
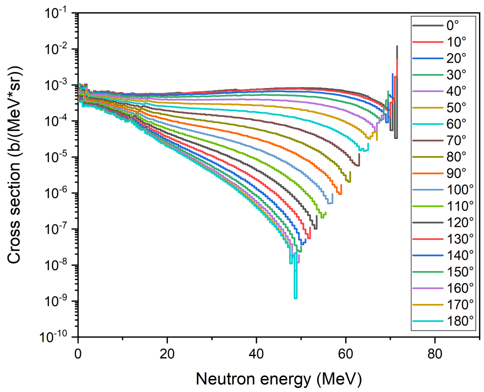
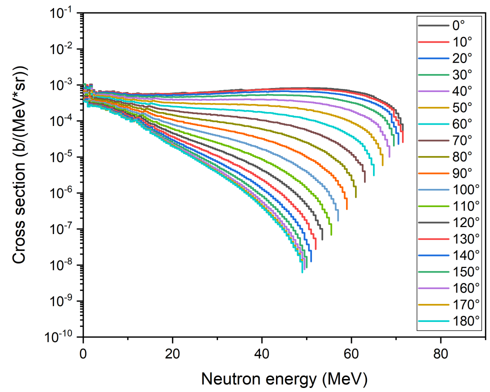


Figure 21: Neutron energy spectrum at different angles from 113-MeV proton incident on a 5.83cm-thick carbon target

At this purpose, JENDL-4.0/HE has been processed with *break_and_mix.exe utility* (as detailed earlier), neutron spectra have been smoothed, re-assembled with *assembly.exe,* processed with NJOY then tested with MCNP calculations. Figure 24a shows that removing peaks has negligible impact on MCNP simulation.

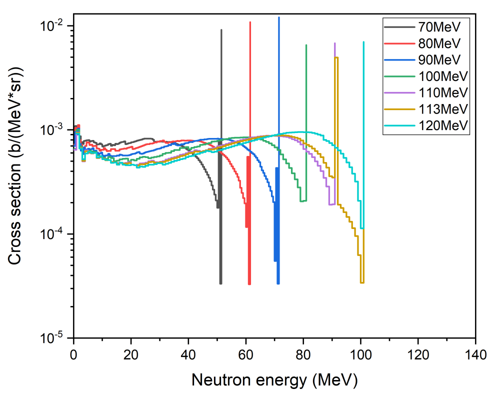


(a) before removing peaks

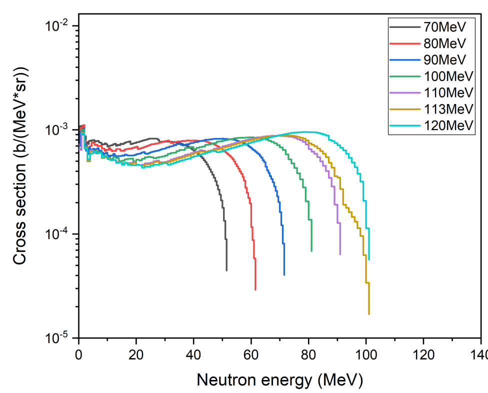


(b) after removing peaks

Figure 22: Neutron production cross-section in every direction from JENDL-4.0/HE library for a 90-MeV incident proton beam

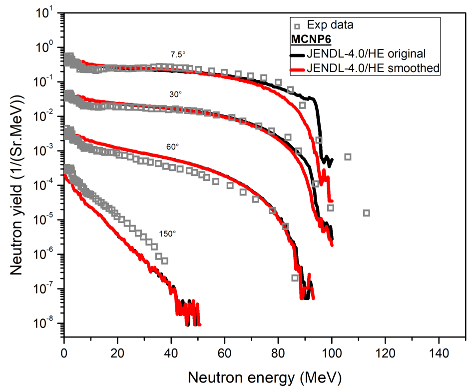


(a) before removing peaks

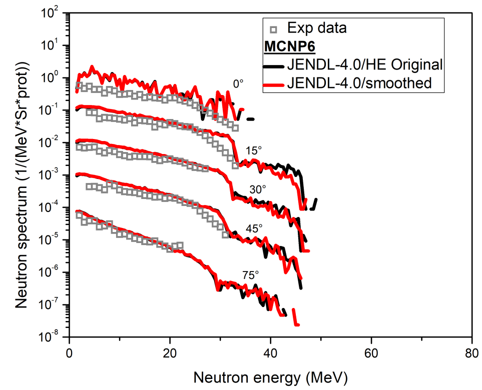


(b) after removing peaks

Figure 23: Neutron production cross-section at 0 from JENDL-4.0/HE library several energies incident proton beam



(a) 113-MeV proton beam on 5.18cm-thick carbon target



(b) 52-MeV proton beam on 2.14cm-thick carbon target

Figure 24: JENDL-4.0/HE neutron spectra smoothing effect on MCNP calculations for proton beam on a thin and thick carbon target

These results have been communicated to JAEA that developed JENDL-4.0/HE library so they can have a look at it and study the problem further. However, since it does not have significant impact on MCNP simulations performed during this internship, JENDL-4.0/HE can be used for the rest of this study. For consistency, smoothing effect has also been studied for the 52MeV experiment and no significant impact has been noticed either (Figure 24b)

8 Conclusion

The main objective of this internship was to analyse and improve proton-induced nuclear data for carbon isotopes.

Reliable nuclear data are needed to perform activation and shielding calculations in order to correctly estimate the doses to public, workers, environment and equipment due to activated materials and to design shielding needed to attenuate radiation to acceptable level.

A preliminary study of existing proton-induced libraries TENDL-2017, TENDL-2019, JENDL-4.0/HE, the libraries in JANIS and TALYS calculations has been carried out regarding residual nuclei production cross sections. These data have been compared with experimental data available in literature using EXFOR database. Analysis doesn't show a best fit with experimental data for one specific library and conclusion was different depending on the reaction considered. For some reactions, rescale of existing library was necessary to match experimental data and never underestimate cross section. After selecting best match with experimental data and rescaling existing nuclear data files when necessary, a new library with improved residual nuclei production cross section has been created for activation calculations purposes.

Second stake was the adjustment of neutron spectra data to be used in transport calculations. Nuclear data libraries are presented in ENDF-6 format needed to be converted into readable format to extract neutron spectra data. Utilities have then been created to process this ENDF-6 files. Since JENDL-4.0/HE is written in an old style representation of secondary particles spectra, by giving laboratory-frame energy spectra defined in angular bins, it couldn't be processed by NJOY that convert into format used in MCNP calculations. At this purpose, *break_and_mix.exe* has been created.

After testing it and validating it, neutron spectra for neutron production data given by the libraries have been compared with experimental work performed by Meier in 1989. While analyzing data, elevated peaks at high energies have been observed on neutron spectra extracted from JENDL-4.0/HE. Before moving forward, effect of this peaks has been studied. Indeed, even though such peaks should be present because its the discrete component of the spectrum, they should not be that high. Best way to study the biggest impact of the peak was to remove it completely and study this effect by comparing MCNP simulations performed with and without peaks. Simulations show no significant impact of the peaks on Meier experiment [20], it has also be tested on 52-MeV experiment from Uwamino (1981) [19] for consistency. While no significant impact have been shown, peaks observation has been reported to JAEA that develops JENDL-4.0/HE library to study the issue further.

By the end of this work, adjustment of residual production cross sections to experimental data for activation calculations purposes and adjustment of neutron spectra data for transport calculations have been made. Yet, lot of work still need to be done before MINERVA can take place. Next step will be reproduction of this work to other key isotopes for MINERVA design.

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