

Feasibility Study of the Activator (FESTA)

FESTA Report

As per of the contract signed by IBEL SA & Dr. Jose Galvez:

This report presents the experimental results and the MonteCarlo calculations for the FESTA Experiment.

This document has been delivered in four parts (4 deliverables) and in total consists of 9 chapters and 3 annexes.

Prepared by:

Dr. Jose Galvez
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Approved by:

Date:

Feasibility Study of the Activator (FESTA) Report



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for a better life

July, 2015

*IBEL SA, Place du Molard 3, Geneve 1204, Switzerland
Tel.: +41 22 310 1214, info@i-bel.ch*

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1 Executive Summary

1.1 About IBEL

(Should be modified by IBEL SA. Put emphasis on scientific aspects and collaboration with scientific institutions)

IBeL SA is a Swiss company aiming to bring innovative products and services in the domain of radiopharmaceutical production using innovative methods and devices and also in other domains related to energy production.

The company is proposing cutting edge technologies and also services to implement specific business models, which allow to implement challenging projects by bringing together the institutions and the needed capital through a specialized business model and a detailed business.

The potential to bring innovative technologies together with business ideas and financial support is one of the main characteristics of the company composed of a group of highly competitive business professionals and scientists. It consists of people with long experience in their particular domains and young energetic specialists.

The company is operating in the fields of science aiming to bring real benefit to the society and will make peoples life better and longer.

1.2 Production of Radioisotopes

Radioisotopes are extensively used in medicine, industry, biology and agriculture. The medical uses of radioisotopes cover diagnosis and therapy as well as clinical research. With the use of radioisotopes in the diagnosis, ongoing biochemical processes can be displayed in living organs without damaging them. When injected in the body, they are called radiotracers, because they release energy from its spontaneous radioactive decay, which can be directly detected by instruments external to the body to obtain information about the physiological, cellular and molecular process of interest. The half-life of a radioisotope must be long enough to allow transport from production sites to end-use locations without excessive loss, and short enough to minimize the unwanted radiation dose to the patient after the procedure is completed.

Radioisotopes are produced by nuclear reactors or particle accelerators on so-called target materials, that can be natural substances or materials enriched with stable isotopes. Nearly 80% of the radioisotopes used in nuclear medicine are currently produced by nuclear reactors. Among them, ^{99m}Tc (6.01h half-life), a decay product of ^{99}Mo (65.94h half-life), is the most important and widely used. This amounts to 7 million examinations per year in Europe and 8 million per year in the USA (about 12 kCi per week). Among ten major nuclear reactors that produce ^{99m}Tc from crude isotope ^{99}Mo , European countries operate five - HFR (The Netherlands), OSIRIS (France), BR2 (Belgium), MARIA (Poland), and LVR-15 (Czech Republic), Canada operates - the NRU at Chalk River, South Africa - Safari-1, at Pelindaba Site (near Pretoria), Australia - OPAL in ANSTO Research facility, Indonesia - the MPR RSG-GA in Java, and Argentina - the RA-3 in Buenos Aires. This indicates that approximately 95% of ^{99}Mo has been produced by the fission reaction of highly enriched ^{235}U (HEU). Since the early 2000s, there has been a shortage of ^{99m}Tc , widely used to diagnose cardiac and respiratory diseases, and neurological conditions. There has been frequent maintenance shutdown of the NRU and HFR, leading to shortage of ^{99}Mo , the raw material required for production of ^{99m}Tc . In addition, a number of the producing reactors in the current ageing fleet are scheduled to be removed from the supply chain, with the earliest scheduled for 2015 (OSIRIS) and 2018 (NRU) due to technical setbacks. There is thus a need for exploring the large-scale cyclotron production of ^{99m}Tc (see Table 1) as an alternative to nuclear reactors, allowing for a local production of radioisotopes, for example in hospitals, industry and/or research centers, in order to facilitate and guarantee supply of such isotopes at competitive affordable market price (OECD, 2005), (NEA, OECD, April, 2014).

Researchers at TRIUMF in Canada have been testing the $^{100}\text{Mo}(p, 2n)^{99m}\text{Tc}$ reaction, (K. Gagnon et al., 2011) as a viable alternative to the reactor based $^{235}\text{U}(n, f)^{99}\text{Mo} \rightarrow ^{99m}\text{Tc}$ strategy for production of ^{99m}Tc . At INFN-Legnaro Laboratories in Italy, (J. Esposito et al.), reports on the feasibility of alternative $^{99}\text{Mo}/^{99m}\text{Tc}$ production routes based on accelerator-driven neutron sources.

Table 1: Alternative production routes

^{99}Mo	$^{99\text{m}}\text{Tc}$
$^{100}\text{Mo}(\text{p}, \text{pn})^{99}\text{Mo}$	$^{100}\text{Mo}(\text{p}, 2\text{n})^{99\text{m}}\text{Tc}$
$^{100}\text{Mo}(\gamma, \text{n})^{99}\text{Mo}$	$^{98}\text{Mo}(\text{d}, \text{n})^{99\text{m}}\text{Tc}$
$^{98}\text{Mo}(\text{n}, \gamma)^{99}\text{Mo}$	$^{98}\text{Mo}(\text{p}, \gamma)^{99\text{m}}\text{Tc}$
$^{100}\text{Mo}(\text{n}, 2\text{n})^{99}\text{Mo}$	

From the seven nuclear reactions available, only two reactions would make use of an accelerator-driven neutron source. One of them, the reaction $^{100}\text{Mo}(\text{n}, 2\text{n})^{99}\text{Mo}$ requires fast neutrons above 8 MeV and 15 MeV as maximum energy to produce a (n, 2n) reactions, however the production of ^{99}Mo via the $^{98}\text{Mo}(\text{n}, \gamma)^{99}\text{Mo}$ reaction is based on neutron radioactive capture and inelastic scattering reactions, which could be performed by using the ARC method.

Neutron radioactive capture is an efficient process to produce nuclear transmutations when combined with the Adiabatic Resonance Crossing (TARC) technique proposed by C. Rubbia (Rubbia, 1997) at the European Laboratory for Particle Physics (CERN). Perspective applications vary from the elimination of unwanted long-lived radioactive waste (Abánades, A et al., 2001) to short-lived radioactive element production for medical or industrial applications. The method was demonstrated with the TARC experiment at CERN in 1997.

The ARC principle is based on the nuclear properties of lead. Lead is a highly transparent material to neutrons (very low absorption cross section from energies of the order of MeV to very low energies), but with a moderate neutron elastic cross section and nearly independent of energy in a large energy range (mean free path about 3 cm) and with a high atomic mass limiting the maximum energy transfer in a neutron elastic collision (the fraction of the original energy of neutrons after a collision is $\alpha \equiv ((A-1) / (A + 1))^2 = 0.981$ for natural Lead, which gives a small average lethargy $\xi = 1 + \alpha \ln(\alpha) / (1 - \alpha) = 9.6 \times 10^{-3}$). Given these properties, it is easy to see that if a neutron source, producing neutrons in the MeV region is introduced into a large lead volume, a neutron flux $\phi(E, t)$, will be developed with a neutron energy spectrum almost iso-lethargic. In other words high-energy neutrons produced will moderate slowly their energy, moving over long distances without substantial change in their energies (high flux intensity) and, due to the low absorption, the total number of neutrons is conserved down to very low energies. The scattering cross-section being constant thus provides an almost iso-lethargic flux, when integrated over the whole volume covered by the neutron flux. For any particular position this distribution is modified slightly depending on the spatial distribution of the neutron source. The main element to achieve this high flux intensity is the very small energy loss per collision, for this reason, this process is described as adiabatic neutron moderation. The iso-lethargic energy distribution of the flux allows full advantage of resonances in the cross section of the process of activation. In the case of activation

by neutron capture, for example when ^{99}Mo , which has a very strong neutron capture resonance nearly to 20 eV, captures a neutron produces $^{99\text{m}}\text{Tc}$ that decays with a half live of 66 h to $^{99\text{m}}\text{Tc}$. Making this resonance capture in an adiabatic neutron flux, the use of ARC is a promising method to be applied for the activation of medical isotopes materials. This is made possible by the high neutron capture efficiency resulting from the ARC method, which allows the required amount of the radioisotope to be produced with a relatively modest neutron source.

The simplicity of this method is intended to promote 'local' production of short-lived radioisotopes, thus eliminating costly, swift transportation and the consequent need of larger initial inventories and thus extending their practical utilization.

The ARC method was validated experimentally by measuring in a consistent way the neutron flux energy and space distribution. However major disadvantages can be remarked in particular the notable low yield production, the use of enormous amount of lead and iron allocated to the device and finally the use of a too high energy accelerator. These limitations do not devalue the scientific purpose of the ARC method, but put this method far from any industrial purpose. However this can happen by using low energy particle accelerators (cyclotrons) with a fundamental change in the geometry and the materials that constitute the device. Precisely in order to reach an industrial use of this principle, after the TARC experiment were carried out various attempts which unfortunately could not be industrially implemented (I. Tilquin, 2005), (K. Abbas, et al., 2007), (K. Abbas, et al., 2009), (P. Froment, 2002).

With these motivations a detailed engineering design of a compact Activator was carried out by IBEL SA and proposed to be tested at Antoine Lacassagne (CAL) Center in Nice, which is one of 18 cancer centers in France. Equipped with a 65 MeV proton cyclotron (MEDICYC), CAL is specialized, inter alia, in proton therapy for eye cancer treatment. The objective of the experimental test is the measurement and simulation of the production of neutrons inside of the Activator device constructed by IBEL SA. In addition it was also relevant to study the neutron diffusion and moderation inside such a compact device and to validate the Adiabatic Resonance Crossing, ARC, method for the most important radioisotopes used at present in Nuclear Medicine, as for example $^{99\text{m}}\text{Tc}$. This method could be also used for a possible recovery of ^{98}Mo from spent generators.

1.3 FESTA Experimental programme

To carry out the experiment FESTA have conducted the following studies:

Neutron flux measurements, which was carried out by irradiating with neutrons produced by the impact of protons (deuterons) in a beryllium target, different activation foils of high purity (> 99%) such as Al, Au, Cu, Fe, In, Mg, Ni, Sc, Ti, V and Zr. The gamma radiation of these samples was measured with a germanium detector (HPGe). We were able to collect a lot of information by placing the samples in different positions of the activator as a function of the target distance. For this the activator was composed of 10 holes that allow the entry of samples holders and they could place up to three samples simultaneously for each irradiation. For this reason, technically speaking the activator could contain up to 30 positions simultaneously. The beam direction (Z axis) is perpendicular to hole directions (X axis). The use of these positions was made considering four layers A, B, -B and C in the plane (Y, Z) as shown in Figure 1. Layer A was allocated in line of the beam direction and layer -B, B and C at 15 cm, -15 cm and at 20 cm from Layer A respectively.

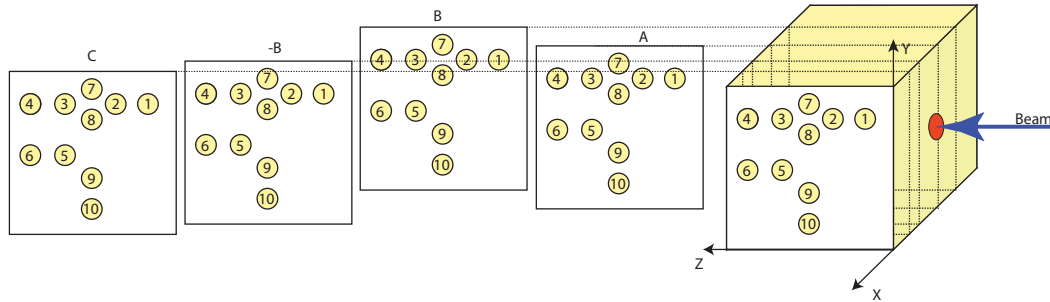


Figure 1: Activator holes & position layers

Then all these positions (Figure 1) give us the possibility to carry out a detailed study of the neutron flux in the Activator. The distance from the center of the target (0, 80, -6.9) to the center of the sample is also shown in Table 2.

Table 2: Layers and positions of the Activator samples

	Layer A					Layer B					Layer C				
Hole	x	y	z	d	x	y	z	d	x	y	z	d			
1	0.00	96.00	-30.00	28.10	-15.00	96.00	-30.00	31.90	20.00	96.00	-30.00	34.50			
2	0.00	96.00	-10.00	16.30	-15.00	96.00	-10.00	22.10	20.00	96.00	-10.00	25.80			
3	0.00	96.00	10.00	23.30	-15.00	96.00	10.00	27.70	20.00	96.00	10.00	30.70			
4	0.00	96.00	30.00	40.20	-15.00	96.00	30.00	42.90	20.00	96.00	30.00	44.90			
5	0.00	81.00	10.00	16.90	-15.00	81.00	10.00	22.60	20.00	81.00	10.00	26.20			
6	0.00	81.00	30.00	36.90	-15.00	81.00	30.00	39.80	20.00	81.00	30.00	42.00			
7	0.00	102.50	1.00	23.80	-15.00	102.50	1.00	28.20	20.00	102.50	1.00	31.10			
8	0.00	94.25	1.00	16.30	-15.00	94.25	1.00	22.10	20.00	94.25	1.00	25.80			
9	0.00	65.75	1.00	16.30	-15.00	65.75	1.00	22.10	20.00	65.75	1.00	25.80			
10	0.00	57.50	1.00	23.80	-15.00	57.50	1.00	28.20	20.00	57.50	1.00	31.10			

We have conducted some precise studies, for example, using Al foils, to check the presence of high-energy neutrons in the Activator, this going to be explained in another section. We have made measurements also of the neutron capture activity with different samples (activation foils) and with different thickness in order to study their influence on the increase or decrease in the rate of capture in the region of resonances.

Neutrons capture rate measurements. Apart from the irradiation of activation foils was conducted the study of the activation by neutron capture of a number of isotopes of particular interest in nuclear medicine as ^{98}Mo , $^{\text{Nat}}\text{Lu}$, $^{\text{Nat}}\text{Re}$ and $^{\text{Nat}}\text{Ho}$. The measurement of the irradiated samples, i.e. the gamma peaks was performed with a germanium detector.

The large number of measurements using a proton/deuteron beams carried out on samples in the whole volume of the activator provide self-consistency checks, including reproducibility of measurements and systematic control, and confirms the validity of the cross section data of the studied samples.

Monte Carlo Simulation. To estimate the experimental results, Monte Carlo simulation work was performed. Originally, only FLUKA was considered for FESTA, however a problem was detected in the deuteron beam case conducting us to consider other MC codes like PHITS and GEANT4. SimpleGeo program was used for converting from FLUKA geometry into PHITS (or MCNP) geometry.

In the case of proton beam, we have observed an excellent agreement with the entire FESTA experimental data set that validates in general the FLUKA MC simulation results for the different samples and different positions in the Activator.

Figure 2: MEDICYC Schematic view

MEDICYC can provide 65 MeV protons with a very low intensity beam, from few nA to 10 μ A (it could be reached also 40 μ A). In addition it is also possible for MEDICYC to provide a 30 MeV deuteron beam with low intensity beam until 10 μ A. In this context, it was agreed in the framework of the scientific and research collaboration that AD carries out with the magnetic field studies and MEDICYC settings for accelerating protons and deuterons and with the optical studies and definitions of the different elements in order to provide the new beam line for the installation of the IBEL activator (Figure 3).

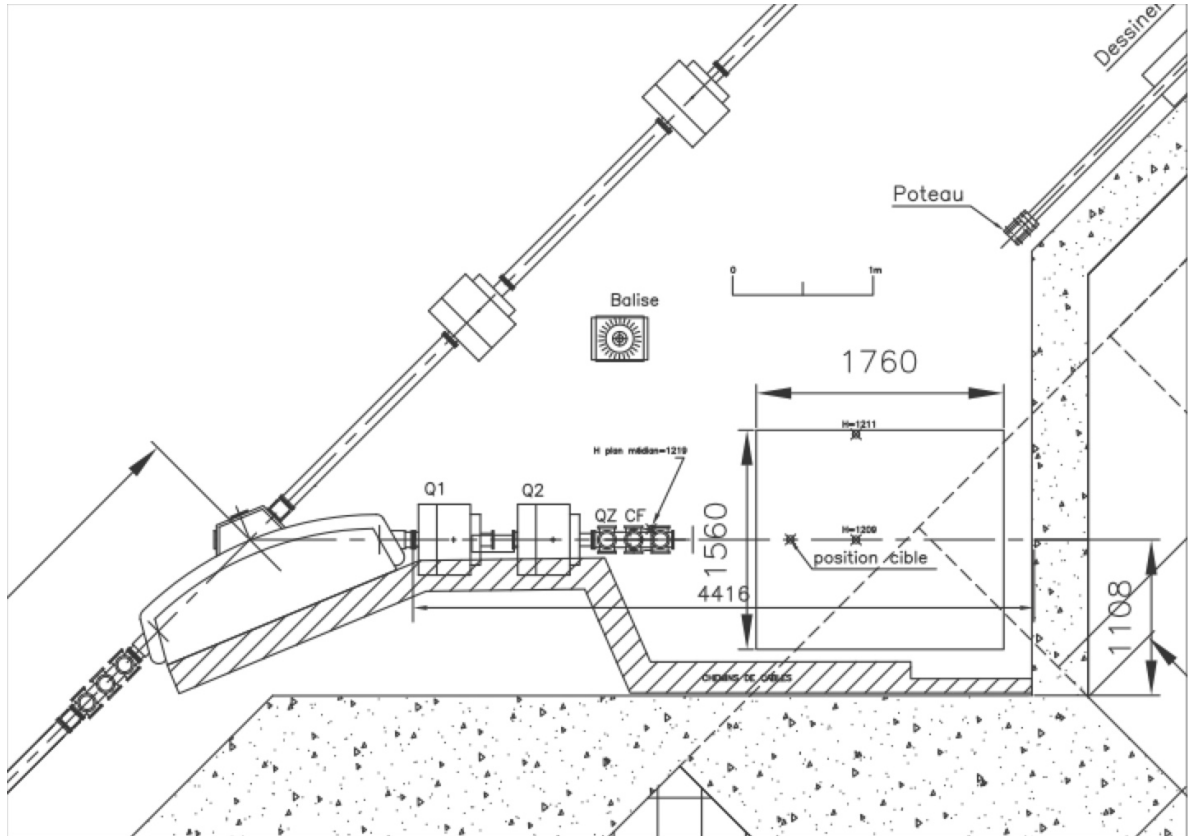


Figure 3: FESTA Experimental Setup at CAL

The main elements of the FESTA experiment are the Activator, the proton (deuteron) beam, the DAQ system and Monte Carlo Simulations.

The activator consists of a 3.27 tons lead assembly, with a cubic geometry of 0.6 x 0.6 x 0.8 m³. The design also includes inner graphite and outer polyethylene layers, which completely surround the lead assembly and are considered to be used as a reflector, moderator and shielding correspondingly (Figure 4). The diffuser block is made of 16 lead bricks with 200 Kg each. The purity of lead is very high and is about 99.99%, avoiding in this way unexpected activation of unwanted materials and to ensure that impurities have a negligible effect on the neutron flux.

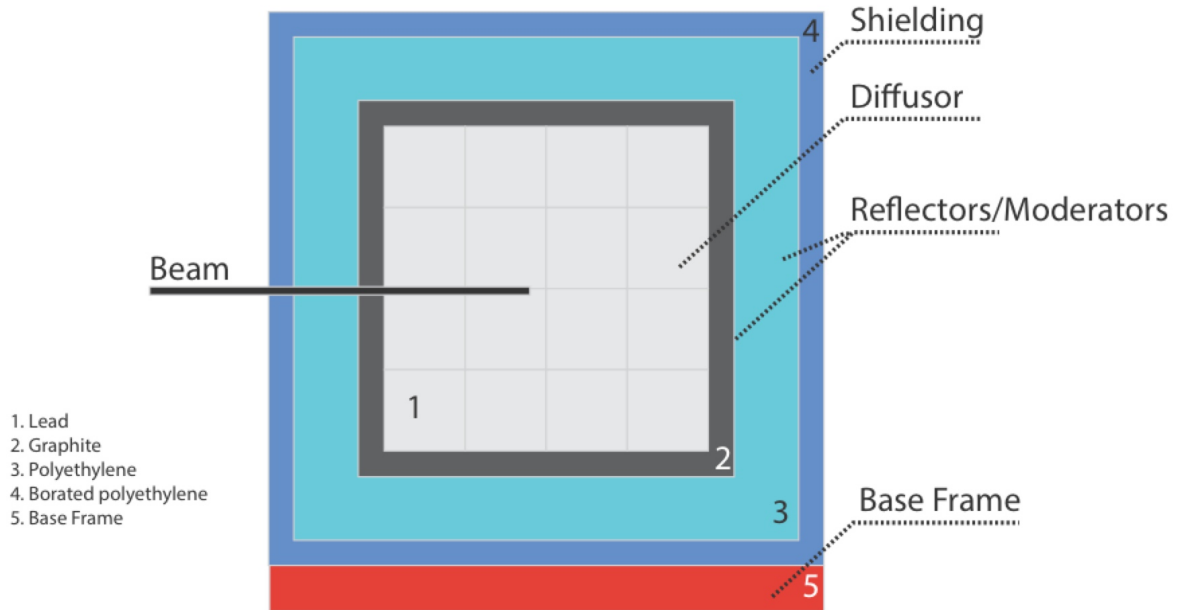


Figure 4: Side view of the Activator assembly

In practice, the experiment calls for high-purity lead devoid of such known impurities as silver, antimony, and cadmium. All the measurements were combined to obtain the best estimate of the impurity concentrations in lead (Table 3).

Table 3: Impurity concentrations in lead

Chemical elements	Max customer specifications	Measurement results
Sb	0.001	< 0.00002
As	0.001	< 0.00005
Sn	0.001	< 0.0001
Cu	0.003	< 0.0015
Zn	0.0005	< 0.0002
Bi	0.03	< 0.0115
Cd	0.001	< 0.00005
Ag	0.005	< 0.0022
Ni	0.001	< 0.0002
Impurities	0.03	< 0.0156
Pb		99.98%

In order to check how the purity of lead diffuser affects to the activity of the irradiated samples, a simulation test was conducted comparing two different types of lead (99.99% & 99.97%). The composition of the first lead (green lead) is in Table 3, and second lead material (violet lead), is shown in Table 4.

Table 4: Violet lead composition

Lead	Sb (%)	Sn (%)	As (%)	Bi (%)	Cu (%)	Ag (%)	Cd (%)	Ni (%)	Zn (%)
Violet	0.0005	0.0005	0.0005	0.01	0.0005	0.0015	0.0002	0.0002	0.0002

The activities obtained with the two types of lead are shown in Figure 5, for a cooling period of one second. The deviations between the two configurations are very small, within the statistical uncertainties of the simulations. The "Green" lead (99.99% purity) should be preferred in this case, since it is less expensive and exhibits a similar performance, from the neutronics point of view.

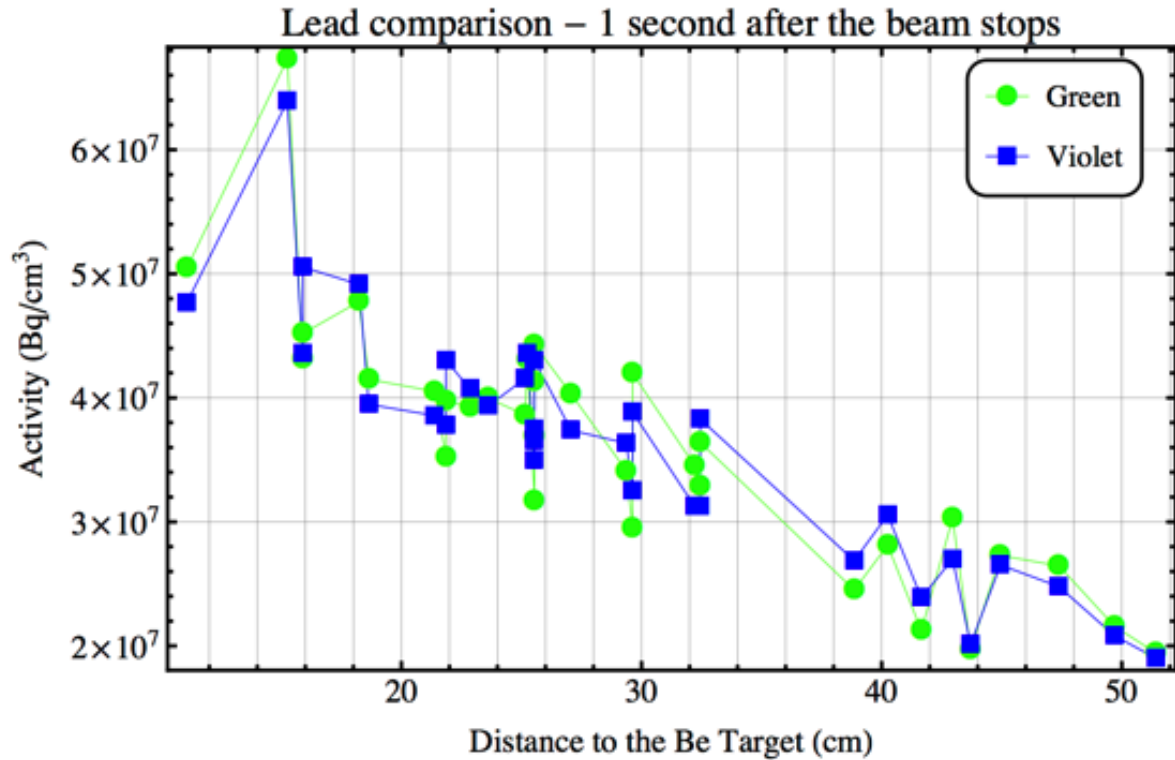


Figure 5: Comparison between the activities of ⁹⁹Mo using "green" and "violet" lead, 1 second after the beam stops.

Another analysis made with this configuration was the comparison of graphite and beryllium as neutron moderators/reflectors surrounding the ⁹⁸Mo samples. The results (Figure 6) show that there is no difference between 15 cm thick graphite and 5 cm thick Be reflectors, so the preference goes for the least expensive: graphite.

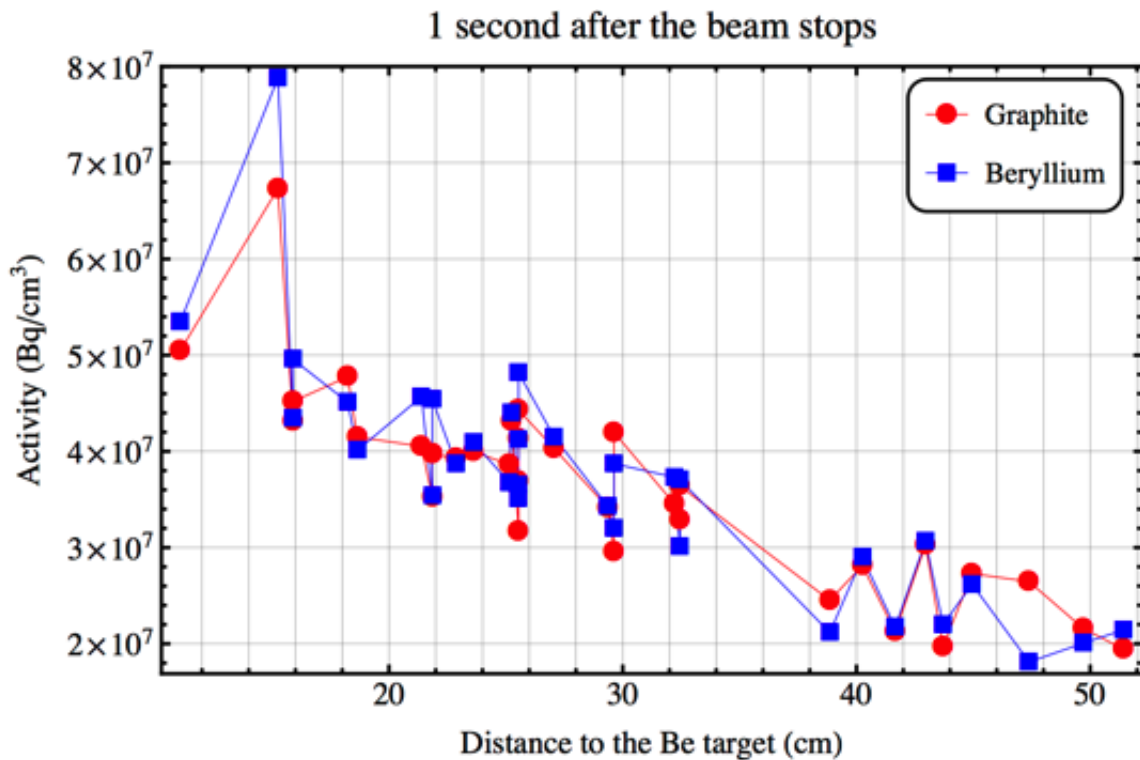


Figure 6: Comparison between the activities of ^{99}Mo using graphite and beryllium as neutron moderators/reflectors surrounding the ^{98}Mo targets.

2.2 Activator Assembly

The activator is composed of 3 main subsystems (DAES SA, 2015):

- The activator box
- The target assembly
- The sample holder

2.2.1 The activator box

The activator box is made with the following components:

- The base frame and the metallic table (all of iron)
- The lead bricks
- The graphite reflector
- The polyethylene and borated polyethylene shielding

The activator box was assembled in situ and starting with the base frame metallic support structure (iron). This base frame has several metal feet that allow an easy alignment of the whole structure, especially with the beam line direction. (Figure 7).

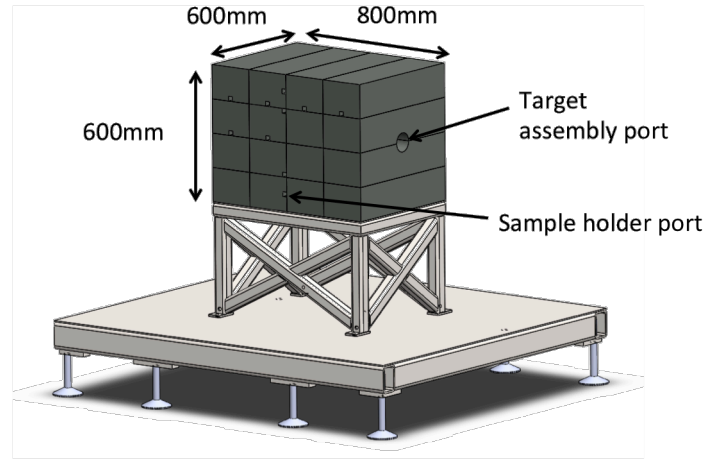


Figure 7: The base frame and lead table supporting the lead bricks

The metallic table holds the lead diffuser, consisting of 16 bricks, with a total weight of 3.27 tons, cubic geometry with a $0.6 \times 0.6 \times 0.8 \text{ m}^3$.

The lead structure has 10 holes perpendicular to the direction of the beam, allowing the insertion of sample holders and one more hole aligned with the beam line which holds the Berilium target.

A graphite reflector (15 cm thickness and $\rho = 1.79 \text{ g/cm}^3$) surrounding the lead diffuser is serving to reduce neutron leakage out of the diffuser by elastic scattering (Figure 8).

Finally, the outer polyethylene layers (3 x 10 cm thickness) acting as a moderator and at the same time as neutron shield, and borated polyethylene layer (30 cm thickness) is to further enhance radiation protection by capturing efficiently the remainder of the neutron radiation (Figure 9).

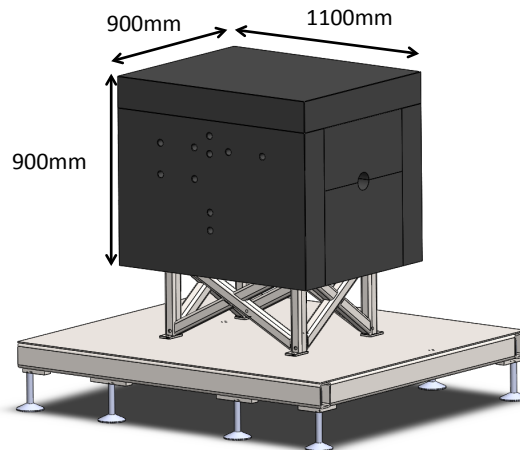


Figure 8: Graphite reflector covering the Lead diffuser

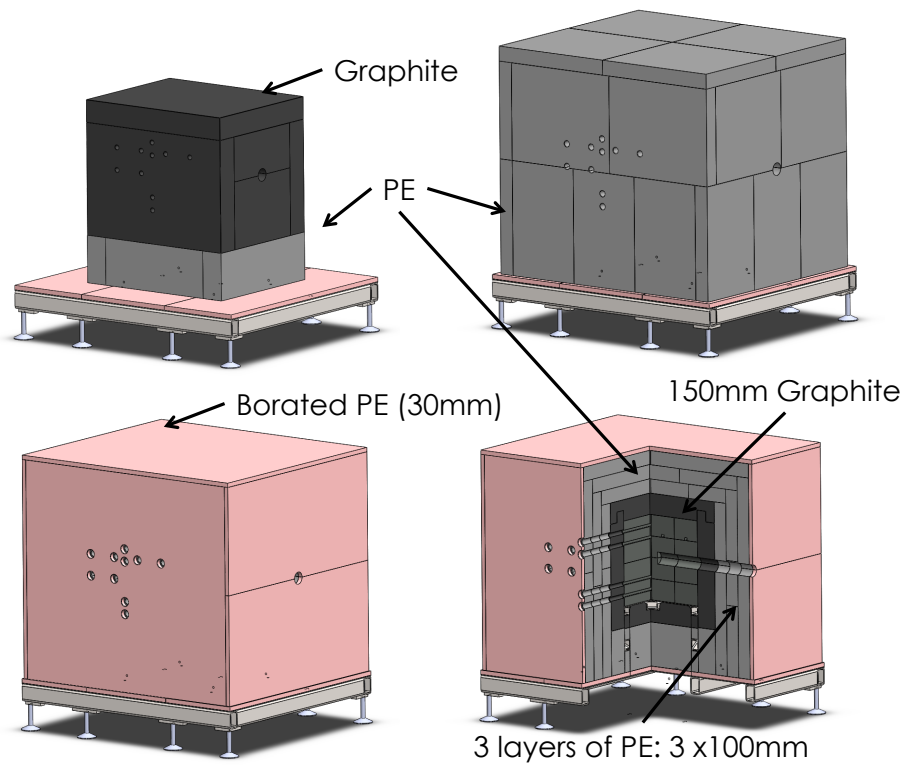


Figure 9: Polyethylene and borated polyethylene layers

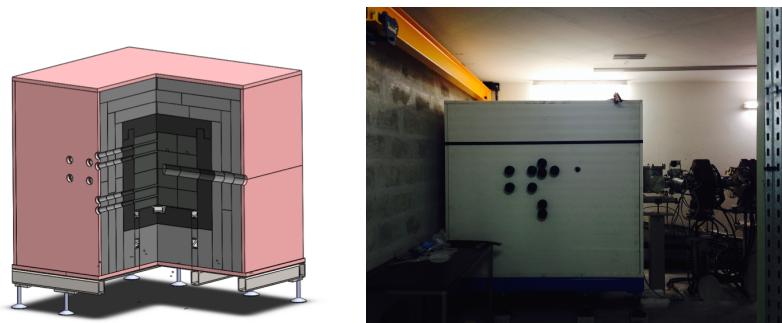


Figure 10: Schematic view of the Activator Assembly

2.2.2 The target assembly

The beryllium target (99.4% purity; $d = 4$ cm & thickness = 2 cm) was placed at the center of the assembly at the end of the proton beam transport tube, and is needed to produce a high neutron flux by bombarding it, with a proton/deuteron beams.

For thick targets the average neutron energy and the yield of neutrons with energies above 2 MeV produced at 0° from Be + p reaction can be calculated by the empirical expressions (M. A. Lone et al., 1981):

$$E_n = 0.47 E_p - 2.2 \text{ (MeV)}$$

$$Y_n = 1.6 \times 10^{13} E_p^{2.2} (n \times sr^{-1} \times A^{-1})$$

For deuteron energies above 10 MeV, the average neutron energy and the yield of neutrons with energies above 2 MeV produced at 0° from Be + d reaction can be calculated by the empirical expressions:

$$E_n = 0.4 E_d - 0.3 \text{ (MeV)}$$

$$Y_n = 3.4 \times 10^{13} E_d^{2.5} (n \times sr^{-1} \times A^{-1})$$

The average energy in case of Be + p is of the order of 27 MeV and in case of Be + d reaction is of the order of 12 MeV. Neutron yields from Lone formula and the experimental data (M. Majerle, S. Simakov) are shown in Figure 11.

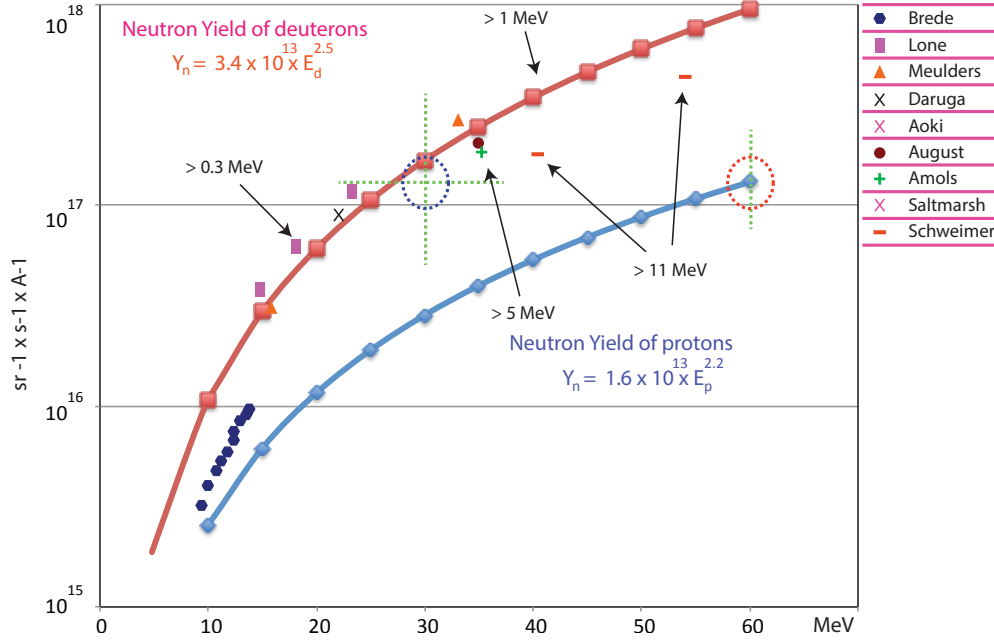


Figure 11: Neutron yield production on the Be target at different energies

Neutron emission spectra in the (d,n) reactions on thick ^9Be target were measured for 25 MeV deuterons at the Tohoku University AVF cyclotron facility. The beryllium target was $\sim 3\text{mm}$ thick to stop the incident beam within the targets. Target area size was $2\text{ cm} \times 2\text{ cm}$. The spectra were measured with the time-of-flight method at 10 laboratory angles between 0° and 90° using two NE213 detector system (T. Aoki et al.)

As a result of the bombardment of proton (deuteron) beams on the Be target with an intensity from 10 nA to 40 μA it is important to estimate the heat produced since the target could melt during a long period of irradiation and thus a cooling system becomes inevitable (Figure 12).

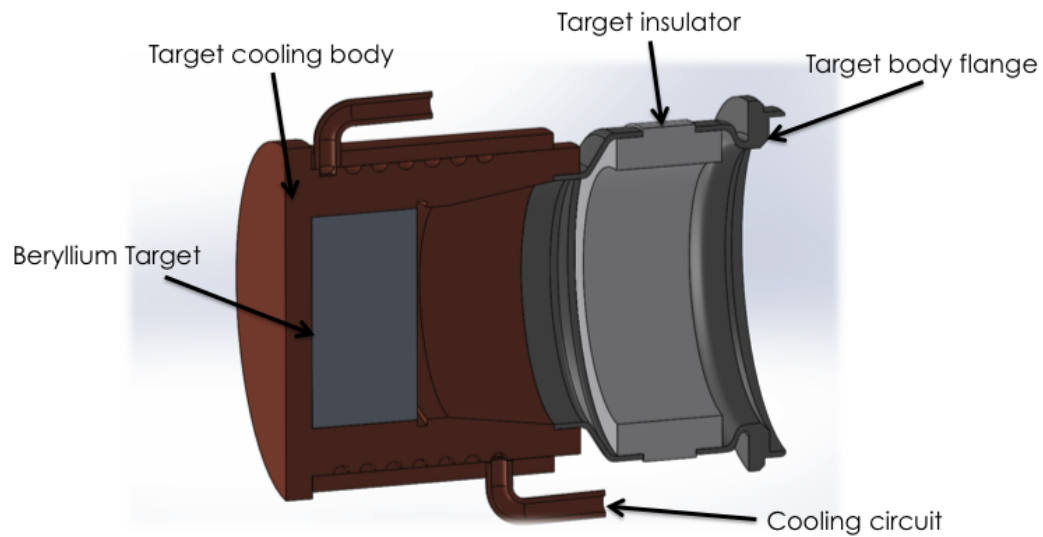


Figure 12: Target cooling system

The temperature distribution calculated in the target assembly for a proton beam current of $0.01\mu\text{A}$ and 0.01kg/s mass flow of water coolant is shown in Figure 13 & Figure 14.

H: Copy of Degradeur13.45
 Temperature
 Type: Temperature
 Unit: °C
 Time: 1
 26/07/2014 16:21

22.045	Max
22.041	
22.037	
22.033	
22.029	
22.025	
22.021	
22.017	
22.013	
22.009	Min

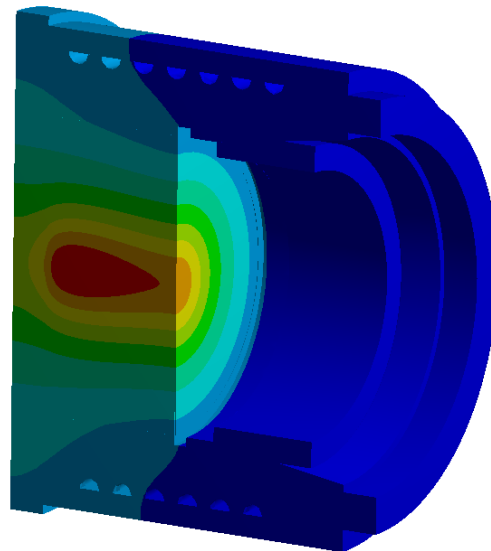


Figure 13: Temperature distribution in the target assembly for a proton beam

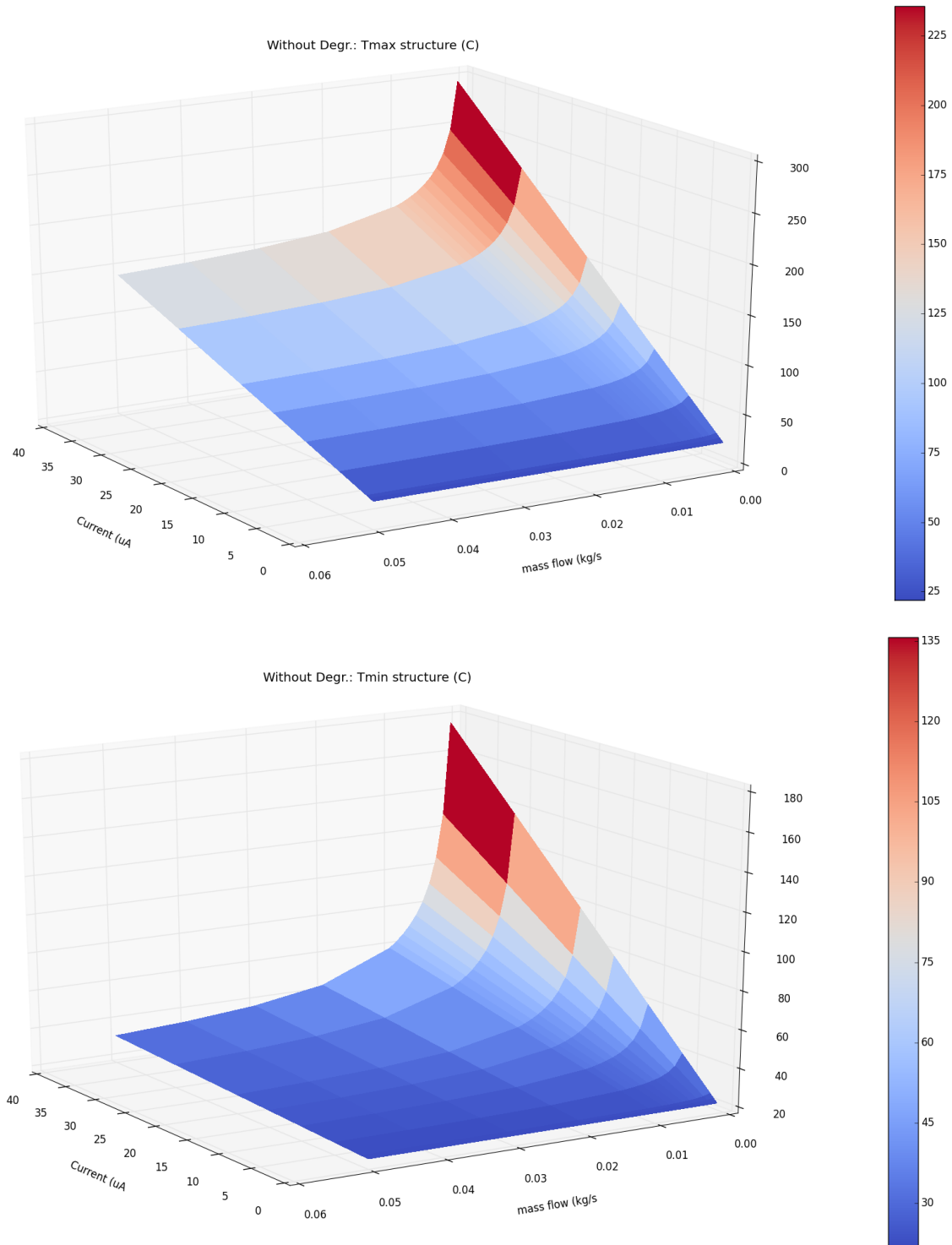


Figure 14: Maximum (top) and minimum (bottom) temperature in the structure against different mass flow rates and different proton beam intensities.

For the purpose of having different energies, eg 30 MeV proton beam or 15 MeV deuteron beam, a degrader was adapted to the target unit (Figure 15 & Figure 16). A degrader is made of Graphite and with the beam interaction also generates heat which is also necessary to evacuate.

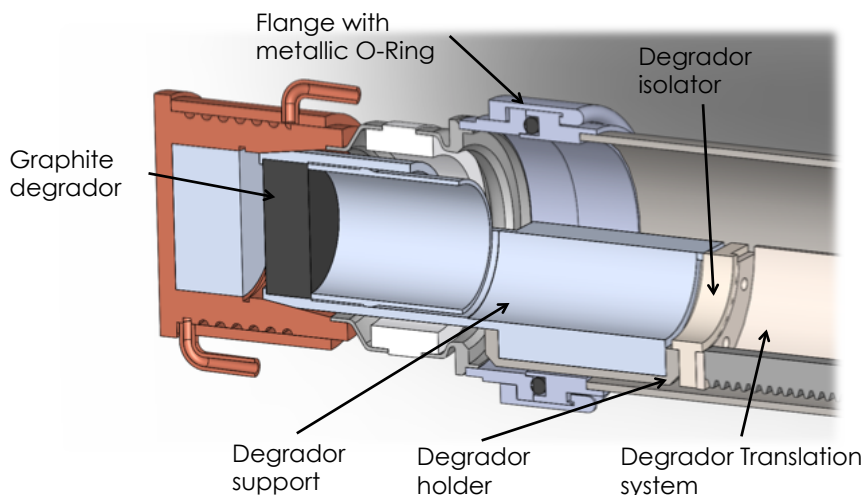


Figure 15: Target assembly with the degrader and the different components

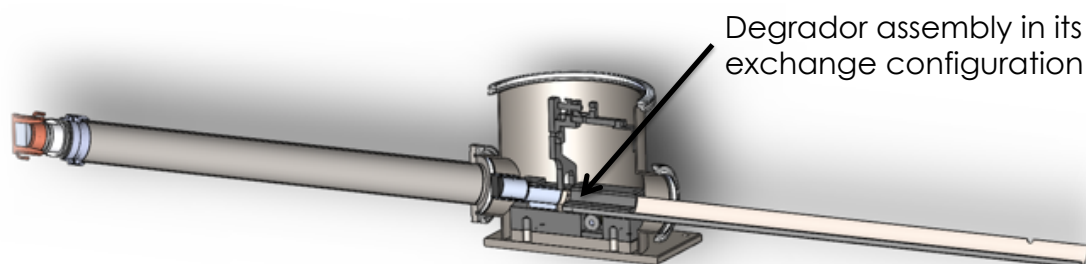


Figure 16: Target assembly with the degrader in its inserted position

2.2.3 The sample holder

The sample holder is a thin structure capable of holding the sample (Figure 17). The sample was inserted into a graphite capsule.

The sample capsule is surrounded by graphite and/or lead blocks. These blocks are acting as local reflector/moderator in case of graphite or acting as an extension of the diffuser box in case of lead.

Samples are properly located at various positions, in total 10 positions, in the lead assembly where the resonance neutron capture probability is maximized.

Sample holders made with Al were used to allocate the samples inside of the activator. The empty space in the sample holder was filled with graphite and lead bricks (Figure 18).

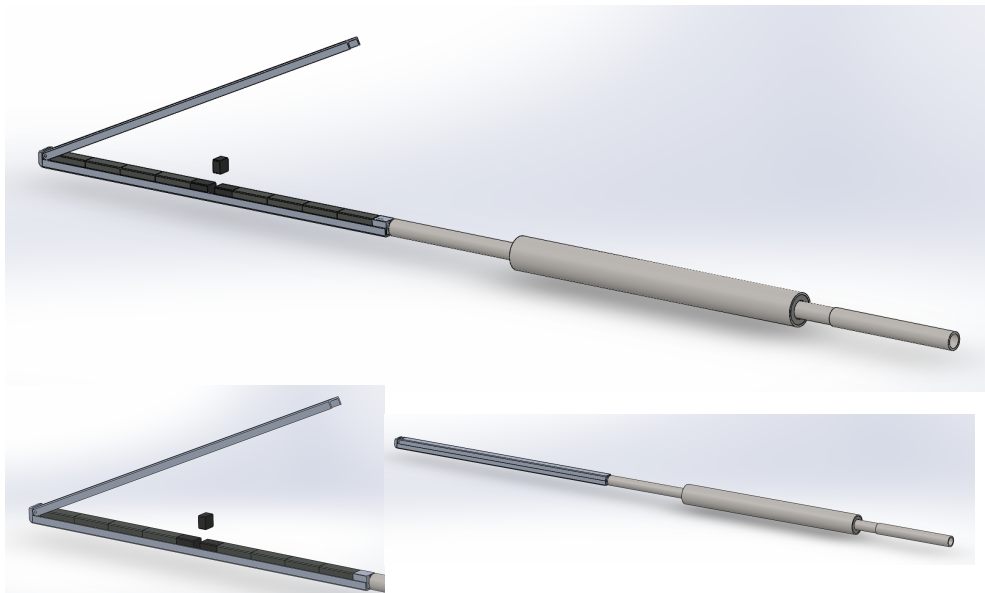


Figure 17: The sample holder, the graphite capsule and the handling tooling

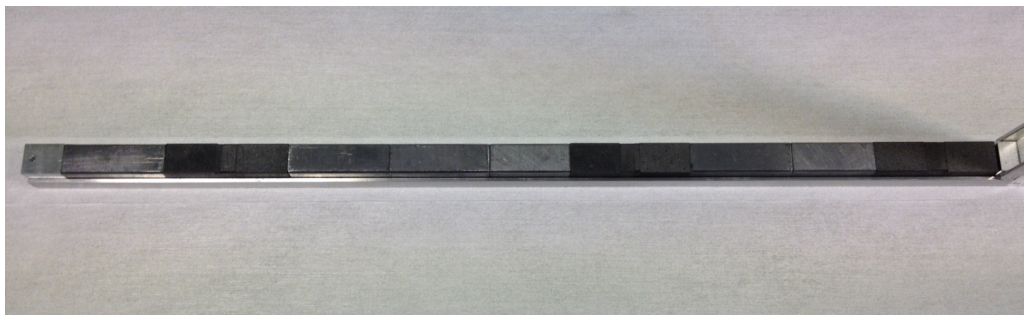


Figure 18: Sample holders filled with lead and graphite bricks

2.3 Activation foils

High purity (> 99%) Al, Au, Cu, Fe, In, Mg, Ni, Sc, Ti, V and Zr foils were used to study the neutron flux in the Activator (Table 5). All foils are 0.5" (12.7 mm) diameter. The Kit from SWX-1553 from Shieldwerx, include a spectrographic analysis of each material and an Activation Foil Manual. In addition to the listed foils, each Combination Kit contains 10 Cadmium Cover Sets for eliminating thermal neutrons.

^{nat}Al , ^{nat}Zr , and ^{nat}Cu foils have a circular shape with a diameter of 1.27 cm. The mass of these samples was with a 10^{-4} g precision.

^{nat}In , ^{nat}Au and ^{nat}Sc are quarter-circle shape with a radius of about 0.064 cm. These foils were measured by 10^{-5} g precision scale balance.

^{nat}W foils are square shape. The length of one side is about 0.9 cm. ^{nat}W foils were measured by 10^{-3} g precision scale balance.

The activation foils were reused several times after the decay more than 10 half-lives.

Table 5: Activation foils used in FESTA (E_{res} the energy of the resonance, σ_{peak} the cross-section at the resonance and Γ the width of the resonance)

	Purity (%)	Thickness (mm)	Avg mass (g)	E_{Res} (eV)	$\sigma_{peak}(b)$	Γ (eV)	Company
Au	99.950	0.051	0.0312	4.906	27405	0.1502	Shieldwerx
In	99.990	0.127	0.0315	1.460	29230	0.0830	
Al	99.989	0.762	0.2600				
Zr	99.739	0.127	0.1120				
Cu	99.993	0.127	0.1460				
Sc	99.900	0.127	0.0109				
W	99.900	0.250	0.3640	18.810	15426	0.3690	SIGMA-ALDRICH

2.4 Medical isotopes

Five kinds of isotope samples were activated in the activator (Table 6). The ^{nat}Mo , ^{nat}Ho , ^{nat}Lu , and ^{nat}Re metal foils were divided into square shapes. The length of one side is about 0.9 cm. The weight of the samples was measured by 10^{-3} g precision scale balance. Thin natural molybdenum foils (0.025 mm thickness) were measured by 10^{-5} g precision scale balance.

The enriched ^{98}Mo metal powder was irradiated in a graphite sample capsule. The diameter of the inner space is about 1.4 cm. The thicknesses of the inner space for different graphite capsules are 1 mm, 0.5 mm and 0.1mm. In general, 1mm thickness mostly was used. The mass was determined by 10^{-3} precision scale balance.

Activation yields (^{nat}Ho , ^{nat}Re , ^{nat}Lu , and ^{98}Mo) after irradiation in different positions and different beam energies inside the activator, were measured by γ -ray spectrometry using calibrated HPGe detector:

- The isotope ^{99m}Tc ($T_{1/2} = 6\text{h}$) is produced in the nuclear reaction $^{98}\text{Mo}(n, \gamma)^{99}\text{Mo}$ followed by disintegration to ^{99m}Tc . (Major ^{99}Mo γ at 739.5 KeV)

- The isotope ^{186}Re ($T_{1/2} = 90.64\text{h}$) is produced in the nuclear reaction $^{185}\text{Re}(n, \gamma)^{186}\text{Re}$ (Major γ at 137 KeV)
- The isotope ^{188}Re ($T_{1/2} = 17\text{ h}$) is produced in the nuclear reaction $^{187}\text{Re}(n, \gamma)^{188}\text{Re}$ (Major γ at 59 KeV)
- The isotope ^{166}Ho ($T_{1/2} = 1.2\text{ d}$) is produced in the nuclear reaction $^{165}\text{Ho}(n, \gamma)^{166}\text{Ho}$ (Major γ at 6.95 & 80.6 KeV)
- The isotope ^{177}Lu ($T_{1/2} = 6.6\text{ d}$) is produced in the nuclear reaction $^{176}\text{Lu}(n, \gamma)^{177}\text{Lu}$ (Major γ at 208.4 KeV).

Table 6: Descriptions of the medical isotope samples

	Purity (%)	Thickness (mm)	Average mass of 1 sample (g)	Company
$^{\text{nat}}\text{Mo}$	99.9	1	0.854	SIGMA-ALDRICH
$^{\text{nat}}\text{Mo}$	-	~0.025	0.0228	-
^{98}Mo	98.61	powder	0.4	ISOFLEX USA
$^{\text{nat}}\text{Ho}$	99.9	1	0.587	Alfa Aesar
$^{\text{nat}}\text{Lu}$	99.9	1	0.727	SIGMA-ALDRICH
$^{\text{nat}}\text{Re}$	99.98	1	1.46	SIGMA-ALDRICH

Table 7: Isotopic distribution of enriched ^{98}Mo

Isotope	^{92}Mo	^{94}Mo	^{95}Mo	^{96}Mo	^{97}Mo	^{98}Mo	^{100}Mo
Content (%)	0.02	0.02	0.03	0.04	0.61	98.61	0.67

3 Characteristic of the beam

The MEDICYC cyclotron is operated by the "Antoine Lacassagne Centre" Laboratory, one of the 20 leading cancer centers in France, where AD is the company responsible for its development and in particular its latest upgrade to implement the deuteron acceleration mode

This is a multiple-particle cyclotron accelerator (protons and deuterons) specially designed for medical applications, that is, radio-therapy by using protons and neutrons, as well as for the production of a large range of isotopes for nuclear medicine.

MEDICYC is operational since June 1991. It is the first historical proton therapy center installed in France, which has treated over 5300 patients with proton therapy of ocular pathologies, representing approximately 5% of all proton therapy treatments that have been made in the world. MEDICYC also provided over 6000 Ci (EOB Activity) of ^{18}F FDG for PET examinations.

The neutron therapy program is no longer active, the beam line called "neutron Way" was rebuilt to meet the specifications of the FESTA Experiment. AD then has provided for this experiment a proton beam of 60 MeV and a deuteron beam of 29.4 MeV.

The major concept of MEDICYC consists in the choice of the acceleration of negative ions to enable:

- A very high performance ion extraction (> 99.9%)
- Fast switch of two beam extractions on each side of the cyclotron.
- Extraction of different ions with variable energy.
- A very accurate monitoring of the extracted beam intensity.
- High stability of intensity.
- Great flexibility and quick adjustment of the intensity of the beams to secure the two operation modes, that is, move quickly and very reliable to proton therapy mode (typically 50 nA extracted) and to isotope-production mode (typically 20 μA proton) i.e. a factor of about 500 for the dynamic in intensity.

To meet this specification, MEDICYC designers have chosen (Table 8):

- An isochronous cyclotron with 4 spiral magnetic sectors,
- A fixed frequency accelerator system (simple and stable),
- An external ion injection from an high performance ion source, where were installed 3 devices in series and independent (adjusting the current source, diaphragms for both modes of operation and Groupers RF cavities, or bunchers) to quickly ensure safe and reliable intensity dynamic.

Table 8: Main parameters of MEDICYC

Extracted beams	Energy Range	20-65 MeV protons 10-30 MeV deuterons
	Range of intensities	10 nA – 50 μ A
Axial injection of ions	External ion source	Negative ions H ⁻ & D ⁻
	Voltage Injection	33 KV
	Groupers HF	Double buncher
Electromagnet	Polar diameter	1.60 m
	Sectors	4 spiral magnetic sectors
	Mass of the magnet	100 tones
	Resistive coils	160000 A*tours
Accelerating cavities	Frequency	25 MHz
	Delta cavities	2 in phase opposition (harmonic 1) or in phase (harmonic 2)
	Peak Voltage	50 KV
Extraction	2 Strippers	- 1 high energy stripper for protontherapy and isotopes production at high energies - 1 lower energy stripper for the production of PET isotopes (F18)
Beam lines	3	2 high-energy lines 1 line at low energy

*Figure 19: MEDICYC*

Figure 19 shows the cyclotron and two extraction lines. On the left is the high-energy line and on the right is the low energy line for PET isotopes. Above the electromagnet, is the injection axial line of MEDICYC.

Figure 20 shows in details the injection axial line: the ion source is a source of multicusp type in a high voltage cage at 33 KV. The elements below the source cage are the focusing lenses and the Groupers RF cavities (bunchers). To bend the vertical trajectory in the horizontal midplane of acceleration, a spiral inflector (Figure 21) is housed in the central region of the cyclotron.



Figure 20: MEDICYC injection axial line



Figure 21: MEDICYC spiral inflector

Figure 22 shows a schematic view of MEDICYC. On the top right, is the high-energy extraction used for FESTA and bottom left is the 20 MeV protons extraction for the production of PET isotopes (^{18}F).

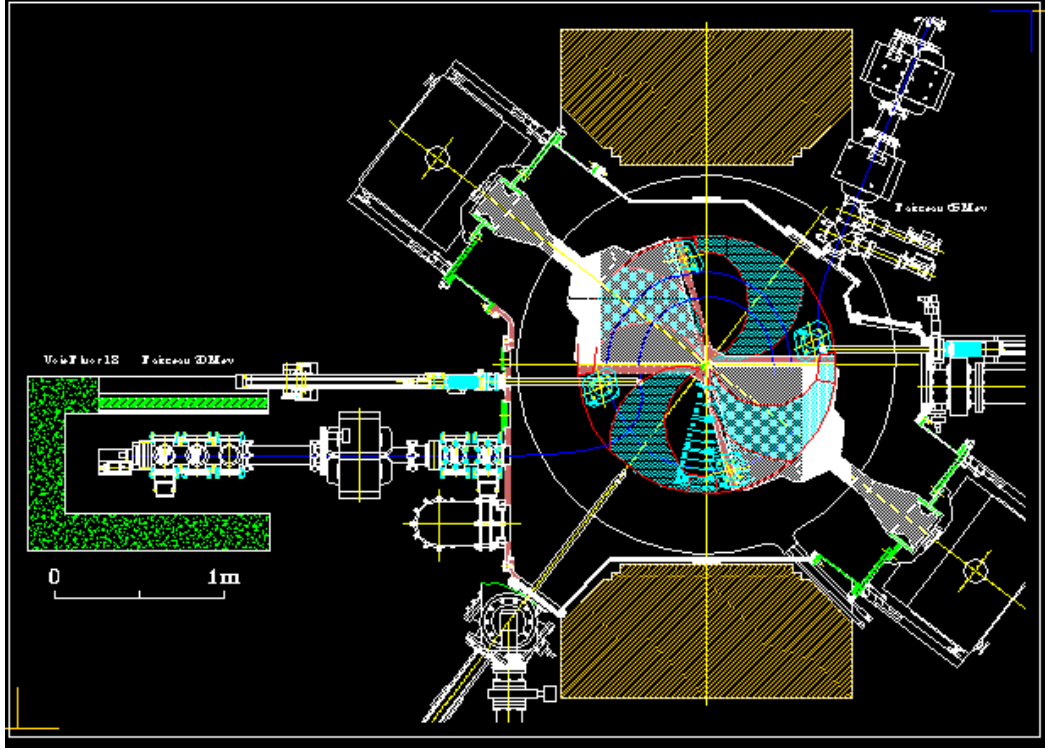


Figure 22: MEDICYC schematic view

Figure 23 shows the overall plan of MEDICYC beam lines. The cyclotron room is on the left with the two extraction beam lines. The energy line (65 MeV proton or 30 MeV deuterons) goes along over 20 meters to reach the magnet M2, which provides power to the third beam line needed in the area of the Activator (green box on the right of the plane). Proton region (not visible in this plane) is located downstream of the beam line to the right of the magnet Q9. In the center of the figure is shown the image of the beam at the diagnostic station 2 indicated by the blue arrow.

The experimental area (in green in Figure 23) of the Activator is downstream of the beam channel powered by magnet M2 (see the details of the optics in Figure 24). This approach was used in early MEDICYC for neutron therapy program. The optics of this pathway has been completely calculated and defined by AD in the framework of the collaboration CAL/AD/IBEL. In this context, CAL has completed the installation of the beam line to the Activator. CAL has provided the design of the Beryllium target for neutron production.

During the FESTA experiment, two types of beams were sent on the Beryllium target, which is allocated in the center of the Activator:

- A 60 MeV proton beam,
- A 29.4 MeV deuteron beam.

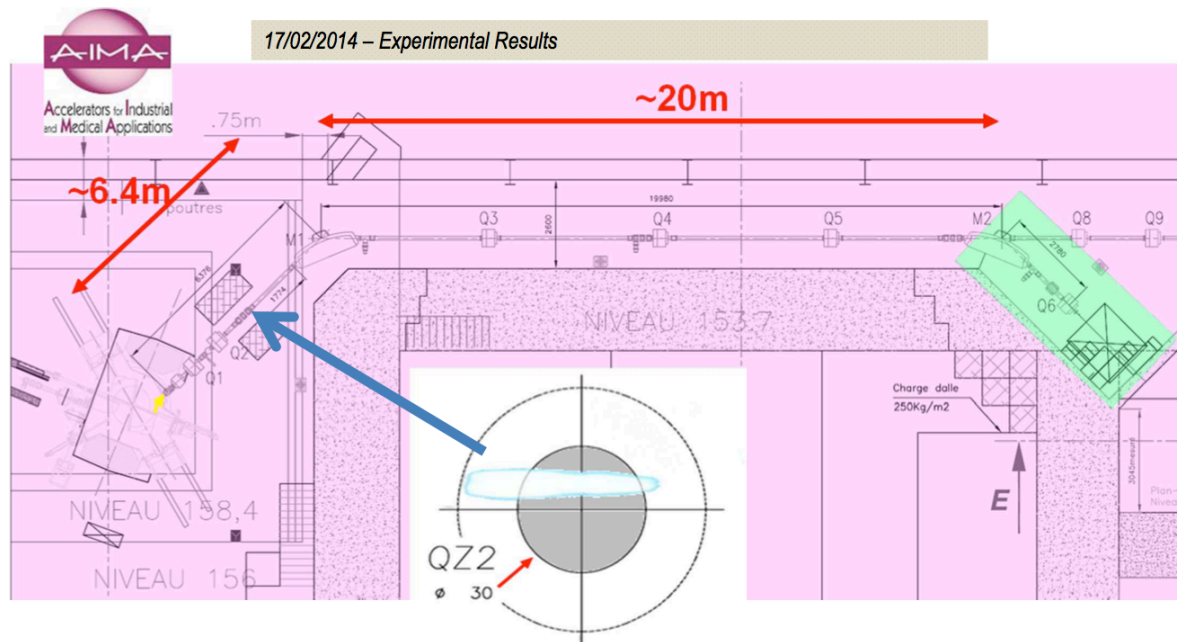


Figure 23: MEDICYC beam lines

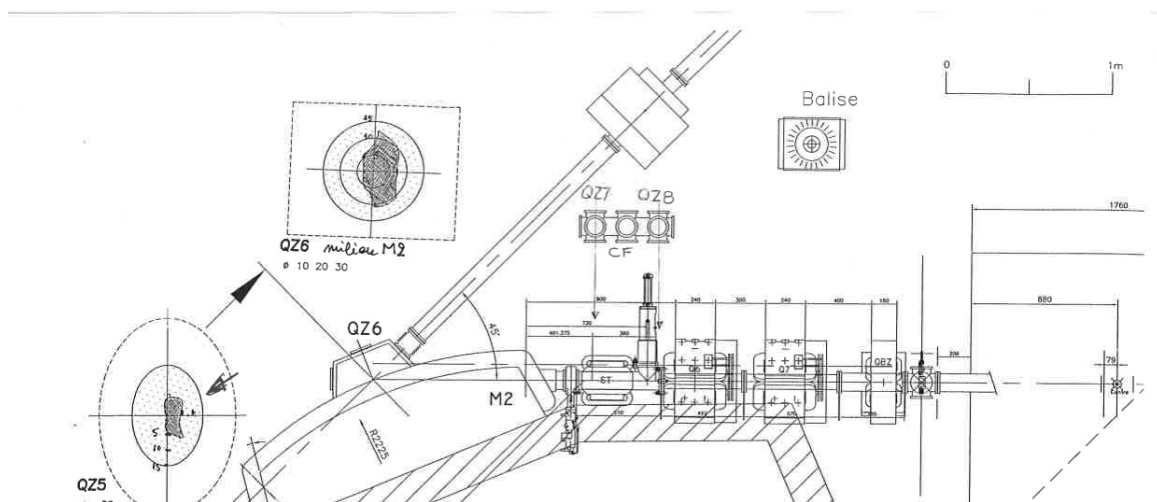


Figure 24: FESTA experimental area at MEDICYC

4 Data Acquisition System

In FESTA, measurements of the neutron flux in the Activator, covering a range from thermal energies to a few MeV, together with the activation measurements, were carried out using a high-purity germanium (HPGe) coaxial detector.

4.1 HPGe Detector setup

In FESTA a High-resolution gamma spectrometer with an HPGe semiconductor detector type SPEC-100, Ortec, U.S.A., was used to measure and identify the radionuclides produced after irradiating the samples inside the activator (Figure 25). The relative efficiency of the detector is 40% and its energy resolution is 2.15 KeV at 1332 KeV. All samples were contained in a graphite capsule (Figure 26), which was designed especially for the protection of the samples, to hold samples with different dimensions and for their possible re-uses (Table 9).



Figure 25: The configuration of the detector and the sample in a Graphite capsule.

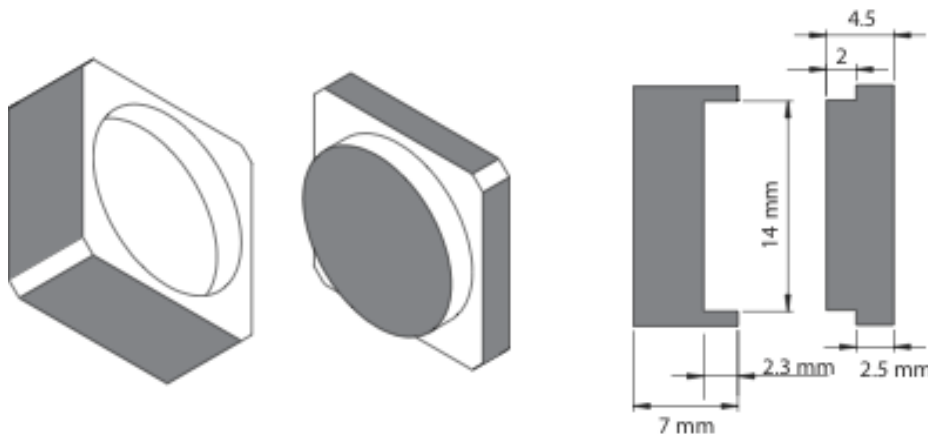


Figure 26: Graphite Capsule schematic view.

Table 9: Thicknesses of the graphite capsule (radius = 7 mm)

Thickness (mm)	Samples	#
0.1	^{nat} Au, ^{nat} Mo, ⁹⁸ Mo	30
0.127	^{nat} Cu, ^{nat} Sc, ^{nat} In, ^{nat} Zr	20
0.25	^{nat} W, ^{nat} Mo, ⁹⁸ Mo	30
0.3		20
0.5	^{nat} W, ⁹⁸ Mo	30
0.62		20
1	⁹⁸ Mo, ^{nat} Mo, ^{nat} Al, ^{nat} W, ^{nat} Re, ^{nat} Lu, ^{nat} Ho	30
2		10
3		10

Random summing (pile-up) effect and high dead times are produced by high-count rates in the HPGe Detector from the irradiated samples. To avoid these problems, the dead time was kept below ~3% by storing the high activity samples in the lead box to slowdown its activities for an adequate measurement. The distance between the sample and end-cap of the detector was kept 10 cm for all measurements and for most of the irradiated samples; the measurement process started 1h after the end of the bombardment (EOB) and lasts few days. Each sample was measured 1~3 times.

4.1.1 Specifications of HPGe detector

The trans-SPEC-DX-100 HPGe detector is an ORTEC GEM Series P-type high purity germanium crystal.

The crystal is housed in an all-metal-sealed, ruggedized cryostat, and cooled by a highly reliable miniature Stirling-cycle cooler. This cooler is capable of approximately 1 W of heat lift at 100°K, and draws less than 25 W when operating.

Detector signal processing is achieved by advanced digital electronics. An active digital noise reduction filter implemented in the digital spectrometer reduces the effect of mechanically generated microphonic noise on the detector output signal, resulting in good energy resolution.

More specifications of the HPGe Detector are as follow:

1. Dimensions 65 mm diameter x 50 mm length nominal. Coaxial construction. P-type high-purity germanium. Weight: 10.6kg
2. Relative Efficiency ≥40% typical (ANSI/IEEE 325-1996)
3. Resolution ≤ 1600 eV @ 122 keV and ≤ 2.3 keV @ 1332 keV (FWHM Warranted at optimum settings)
4. Peak Shape 1.9 typical (FWTM/FWHM)

5. Cryostat and Cooler “Hardened” cryostat with high reliability, low power Stirling cooler. The cryostat design is such that the trans-SPEC-DX-100T may be switched off at any time and power subsequently re-applied without waiting for a full thermal cycle (complete warm up before cool down), as is normal practice with a HPGe detector system.
6. Digital Noise Suppression "LFR Filter."
7. End cap and shield thickness : 1 mm (0.5 mm + 0.5 mm, Aluminum)
8. Operating temperature: 120 K
9. Energy range: ~ 40 keV ~ 3 MeV (Max. 7 MeV)
10. Dead-Time Correction Extended live-time correction according to Gedcke-Hale method. Accuracy: Area of reference peak changes $\pm 3\%$ from 0 to 50000 counts per second.

4.1.2 Specifications of software detector

- The latest version MAESTRO MCA software is included to for identifying peaks, editing libraries, and creating, printing and saving Regions of Interest (ROI), performing energy calibrations, automating tasks via using simple “Job Streams”.
- Gamma-Vision v6.08, a gamma spectroscopy application for high resolution spectrometer systems, was used to analysis the peak area. The core purpose of GammaVision is the accurate identification and quantification of radioactive material using high resolution gamma spectroscopy. This process requires accurate system calibrations and analysis settings, robust peak search and fit algorithms, and a variety of corrections for background, peak interferences, attenuation, reporting units, and various other factors. It is also important to comply with regulations and standards for reporting measurement uncertainty and detection limits.

4.2 Calibration of the HPGe Detector

The efficiency calibration of a channel analyzer using standard sources is a common practice for the HPGe detector, establishing a precise connection between the energy of the gamma radiation and the channel number. Once the energy is identified using the calibration source, the detector efficiency can be also calculated for any energy.

4.2.1 Calibration sources

For the energy efficiency calibration of the Germanium detector it is very important to use of standard radioactive sources, which can be either point, or extended sources. In FESTA two standard sources were used to calibrate the HPGe detector: ^{152}Eu (point source) and a Multi standard source (extended source).

The maximum gamma energy observable by the HPGe detector is about 3 MeV. In FESTA gamma radiation energies emitted from the irradiated samples can vary from tens of KeV to 2 MeV. However some of the samples, in particular ^{186}Re and ^{177}Lu can emit low energy gamma rays, which forces us to calibrate the detector from a range of energies of a few KeV to 2 MeV.

4.2.1.1 Eu-152 Source

The ^{152}Eu calibration source allowed us to calibrate the detector for energies greater than 200 KeV.

- Activity: 4 KBq
- Reference date: 1 April 2007 at 12:00 UTC
- Uncertainty: 3% (95% confidence level)
- Radioactive impurities: $^{154}\text{Eu} < 0.4\%$

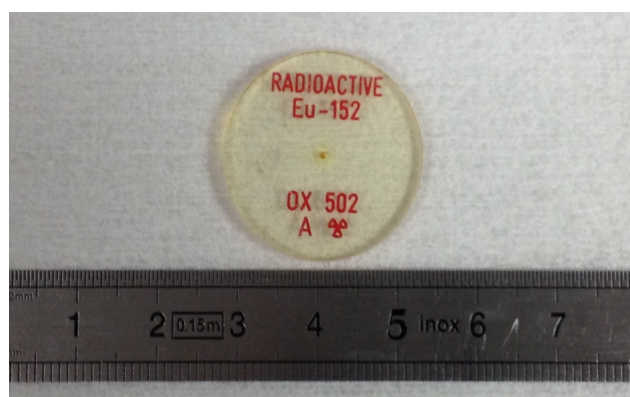


Figure 27: ^{152}Eu Standard Source.

4.2.1.2 Extended Multi Standard Source

The importance of the Extended Multi-Standard Source, with a total activity of 74.595 KBq, is to complete the Full Calibration in Energy of the HPGe Detector and to obtain the calibration curve covering from low 60 KeV to 1.8 MeV.

The dimensions of the source (Figure 28) were made according to the dimensions of the sample capsule holders used in FESTA. Thus the geometric correction for calculating the activity of an irradiated sample is no longer necessary.

- Producer: Korea Research Institute of Standards and Science, Republic of Korea
- Reference Date: 31 October 2007 at 15:00 UTC,
- Source size: 8mm in diameter,
- Source material: Filter paper.

In Table 10 is shown the exact contents of the radioisotopes that compose the source, which emit gamma radiation with energies from almost 60 KeV to 2 MeV. Thus, a full range of energies is covered.



Figure 28: Extended Multi-Standard Source.

Table 10: Certified Values and Uncertainties of the Extended Multi-standard source

	Half-life (d) (1)		Main γ -ray (KeV)			Radioactivity (Bq)	
		Uncertainty	Energy	Intensity (%)			Uncertainty (2)
²⁴¹ Am	158004	219	59.54	35.92	0.17	6419	0.80%
¹⁰⁹ Cd	461.4	1.2	88.03	3.626	0.026	27206	0.90%
⁵⁷ Co	271.8	0.05	122.06	85.51	0.06	403	1.20%
			136.47	10.71	0.15		
¹³⁹ Ce	137.641	0.02	165.86	79.9	0.04	1105	0.80%
⁵¹ Cr	27.703	0.003	320.08	9.87	0.05	23331	0.90%
¹¹³ Sn	115.09	0.03	391.70	64.97	0.17	2740	0.90%
⁸⁵ Sr	64.85	0.007	514.00	98.5	0.4	2304	1.10%
¹³⁷ Cs	10976	29	661.66	84.99	0.2	2585	1.40%
⁶⁰ Co	1925.23	0.29	1173.23	99.85	0.03	3820	1.10%
			1332.49	99.32	0.006		
⁸⁸ Y	106.626	0.021	898.04	93.9	0.23	4682	1.30%
			1836.05	99.32	0.03		

(1) M. –M. Be et al., Monographie BIPM-5. The half-life and intensity of each nuclide are data for reference. Uncertainty means standard uncertainty. (2) About 95% confidence level, $k = 2$.

Figure 29 shows the typical spectrum for the Extended Multi Standard Source, which clearly we can remark, energy lines from 59 KeV to 1836 KeV. (^{241}Am , ^{109}Cd , ^{57}Co , ^{139}Ce , ^{51}Cr , ^{113}Sn , ^{85}Sr , ^{137}Cs , ^{60}Co , ^{88}Y)

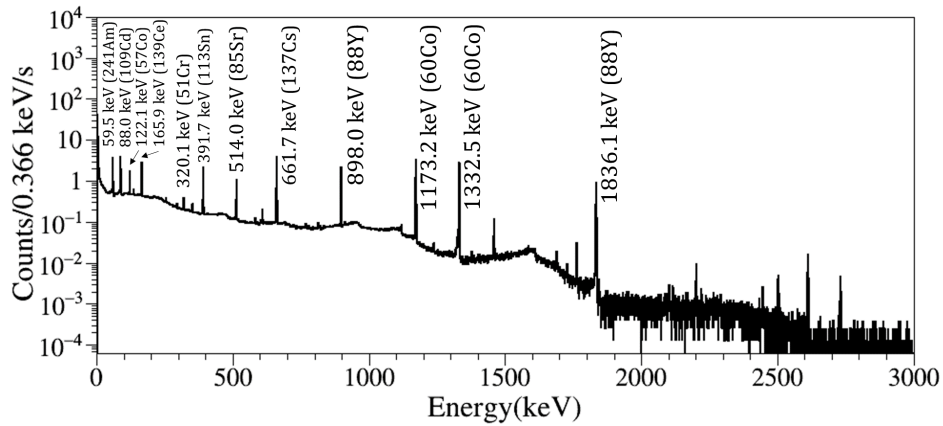


Figure 29: γ -ray spectrum of the multi standard source.

4.2.2 Energy calibration

The energy calibration aims to derive a relationship between the position of the peak in the spectrum and the corresponding γ -ray energy.

Energy calibration is performed:

- by measuring gamma rays from standard source,
- by identifying gamma rays peaks in the spectrum,
- by assigning γ -ray energies to the corresponding channel number in the spectrum.

After matching channel numbers and γ -ray energies, a quadratic fit is applied in order to obtain the energy calibration:

$$\text{Energy} = a_1 + a_2 \times \text{Channel} + a_3 \times \text{Channel}^2$$

where a_1 , a_2 and a_3 are fitting coefficients. Figure 5 shows the calculated energy calibration of the HPGe with the extended multistandard source.

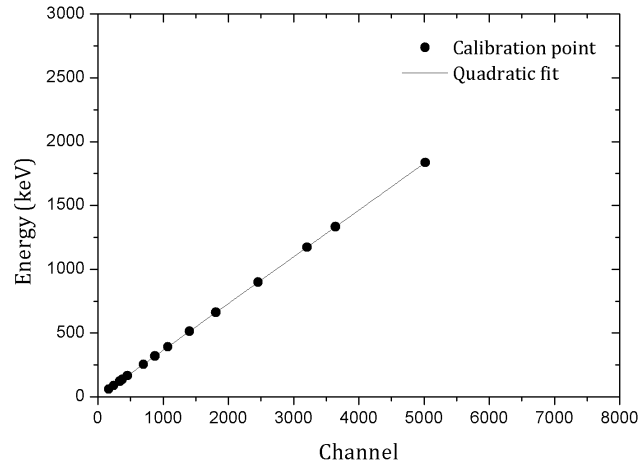


Figure 30: Energy calibration (extended multi standard source)

The gamma ray energies from specific radionuclides are taken from NuDat 2.6 database (NuDat2).

4.2.3 Peak width calibration

Immediately after, the FWHM (Full Width Half Maximum) is obtained for each peak from the spectrum by using gamma vision software. After the matching the FWHM with the channel numbers, a quadratic fit model is applied:

$$FWHM(ch) = b_1 + b_2 \times Channel + b_3 \times Channel^2$$

where b_1 , b_2 and b_3 are fitting coefficients (Figure 31).

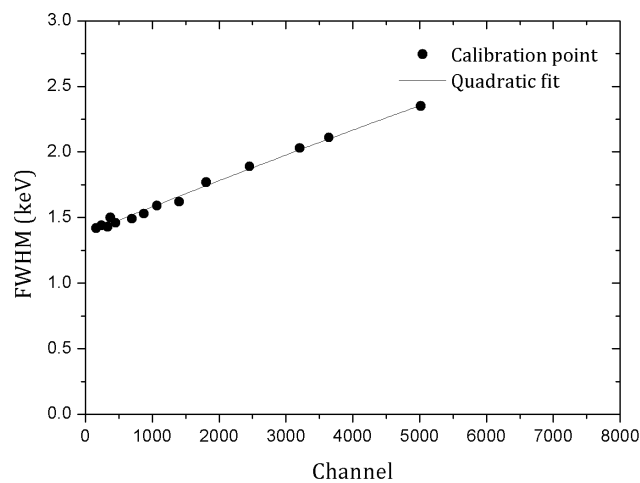


Figure 31: FWHM calibration (extended multi standard source)

Accurate FWHM calibration is important for analyzing γ -ray peaks (WANG Yiming, 2013) in order to precisely distinguish single peaks, overlapped peaks and meaningless fluctuations.

4.2.4 Efficiency calibration

Efficiency calibration is necessary to quantify the activity of radioisotopes. The efficiency is calculated from the following equation:

$$\varepsilon(E) = \frac{C_{net}}{A_N \cdot I_\gamma \cdot t_{live}}$$

where ε is the efficiency at energy E . C_{net} is the net area of the peak at energy E , $A_N(E)$ is the activity of nuclide N , I_γ is the intensity of the gamma ray of the nuclide N at energy E , and t_{live} is the live time.

After the calculation of the efficiency, a polynomial fit function optimized for p-type detectors (ORTEC) is applied:

$$\varepsilon(E) = \exp \left[a_1 E^2 + a_2 E + a_3 + \frac{a_4}{E} + \frac{a_5}{E^2} + \frac{a_6}{E^3} \right]$$

where $\varepsilon(E)$ is the efficiency at energy E (in MeV) and a_i are fitting coefficients.

A detection efficiency curve obtained is shown in Figure 32. The detection efficiency curve depends not only on a detection system, but also of the sample shape and its dimensions. So, the full energy peak efficiency depends on the geometrical conditions and on the energy.

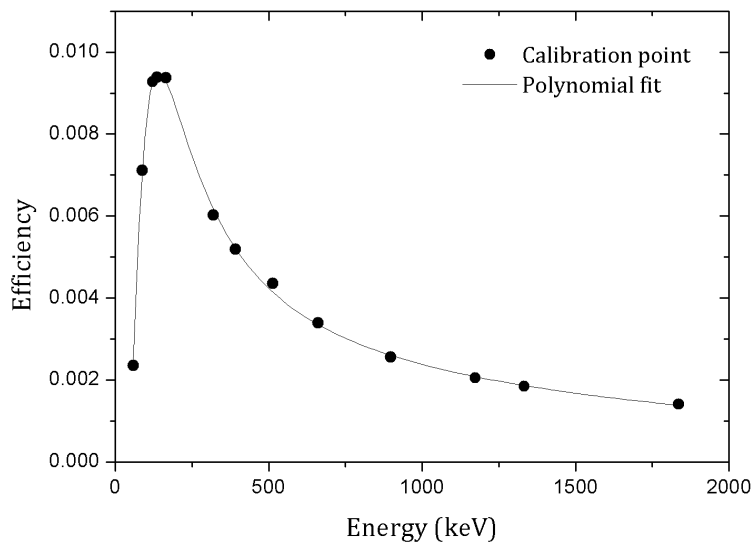


Figure 32: Efficiency calibration curve.

4.3 Background analysis

4.3.1 Background

Background measurements were performed without any special shielding structure. In the gamma spectrum without a radioactive source is observed about 50 counts/s. Figure 33 shows the natural radionuclides (^{40}K , ^{208}Tl , ^{212}Pb , ^{214}Pb , ^{214}Bi , ^{226}Ra and ^{228}Ac) identified in the spectrum. Figure 34 shows the measurement area at CAL Center en Nice, France.

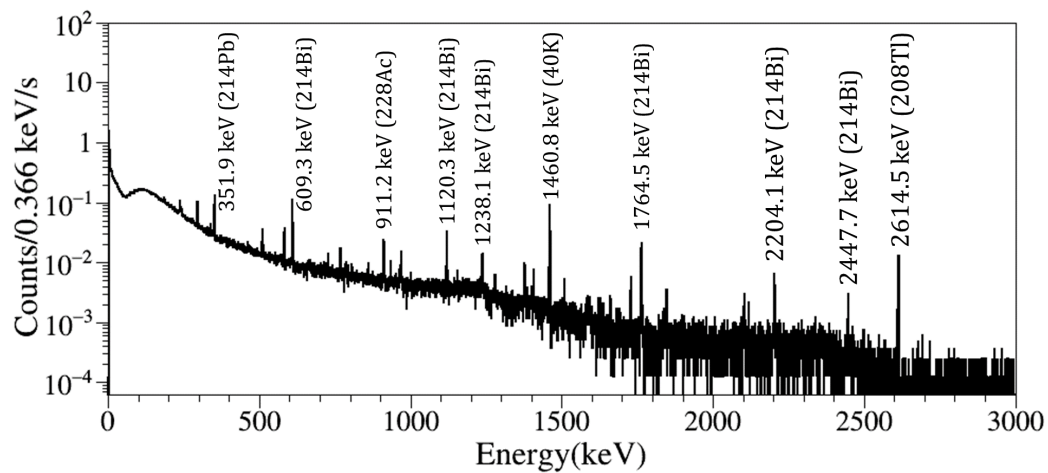


Figure 33: γ -ray spectrum from background.



Figure 34: Measurement area at CAL Center.

4.3.2 Empty graphite capsule

All irradiated samples were irradiated contained in a graphite capsule. The gamma rays of the irradiated samples are attenuated by crossing the graphite capsule, however because the HPGe detector was calibrated with a standard source with the same configuration as the samples, the attenuation effect of the graphite capsule is irrelevant.

Gamma-rays peaks at 511 KeV (^{11}C , $T_{1/2} = 20.334$ m), which emits only annihilation γ -rays ($I_\gamma = 199.534$ %) are not used in analysis (Figure 35).

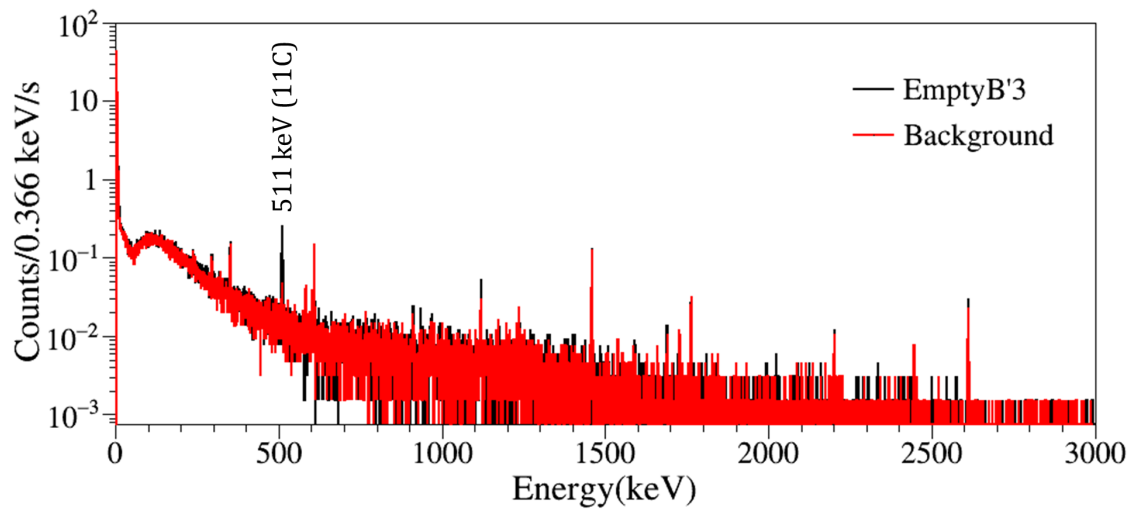


Figure 35: γ -ray spectrum from irradiated graphite capsule.

5 FESTA Experimental results

5.1 Introduction

In FESTA experiment, 20 run tests were conducted with different beam (proton & deuterons) intensities and irradiation times (Table 11).

Table 11: Beam intensity and irradiation time of each run

Run number	Beam type	Avg. Current (μ A)	Irradiation time (minutes)
1	Proton	0.05	48.4
2	Proton	3.88	60
3	Proton	3.15	51
4	Proton	4.06	15
05-Jan	Proton	4.03	10
05-Feb	Proton	4.04	10
6	Proton	4.07	10
7	Proton	4.12	60
8	Proton	4.13	60
9	Proton	4.11	60
10	Deuteron	0.84	20
11	Proton	4.07	15
12	Deuteron	1.03	60
13	Proton	1.01	60
14	Proton	1.01	60
15	Proton	1.01	60
16	Proton	1.02	60
17	Proton	1.01	60
18	Deuteron	0.83	60
19	Deuteron	0.72	60
20	Proton	1.03	60

To carry out FESTA we have performed the following studies:

Neutron flux measurements, which were carried out by irradiating with neutrons produced by the impact of protons (deuterons) in a beryllium target, different activation foils of high purity (> 99%) such as Al, Au, Cu, Fe, In, Mg, Ni, Sc and Zr. The gamma radiation of these samples was measured with a High Purity Germanium detector (HPGe).

Neutron captures rate measurements. Apart from the irradiation of activation foils, we have studied the neutron capture of a number of isotopes of particular interest in nuclear medicine such as ^{98}Mo , $^{\text{Nat}}\text{Lu}$, $^{\text{Nat}}\text{Re}$ and $^{\text{Nat}}\text{Ho}$. The measurement of the irradiated samples, i.e. the gamma peaks, was performed with a High Purity Germanium detector (HPGe).

Monte Carlo Simulation. To compare with the experimental results, Monte Carlo simulation works were performed. Originally, only FLUKA was considered for FESTA, however a problem was detected in the deuteron beam case conducting us to consider other MC codes like PHITS and GEANT4. SimpleGeo program was used for converting from FLUKA geometry into PHITS (or MCNP) geometry.

5.2 HPGe detector for spectrum analysis

The gamma spectra were analyzed by using the Gamma Vision software (ORTEC) as described in Chapter 4. The Activity of the irradiated samples at the end of bombardment was calculated using the following equation:

$$A_{EOB} = \frac{\lambda \times C_{net}}{\varepsilon \times I_{\gamma} \times e^{-\lambda t_{cool}} (1 - e^{-\lambda t_{real}})} \times \frac{t_{real}}{t_{live}}$$

where A_{EOB} is the activity at the end of beam (EOB), λ is the decay constant of the radionuclide, C_{net} is the net counts in the peak, ε is the efficiency of the HPGe detector, I_{γ} is the γ -ray intensity, t_{cool} is the cooling time between the end of bombardment and start of the measurement, t_{real} is the real time and t_{live} is the live time.

The γ -ray spectrum of each sample was measured several times (from 1 to 6 times). Only the main peaks of the spectrum were used to calculate the activity of each sample. The final activity is obtained from the weighted average from different measurements and different peaks.

5.3 Normalization

The specific activity is normalized per unit of mass (1g), per unit of current (1 μ A) and per unit of irradiation time (1h) and it also considers the purity (in %) of the samples.

For time normalization, it has to be considered the decay of the radionuclides, because their activities do not increase constantly:

$$\frac{dN}{dt} = N_0 \cdot \sigma \cdot \phi - \lambda N$$

where N is the number of activated nuclei, N_0 is the initial quantity of target atoms, σ is the cross-section, ϕ is the neutron flux, and λ is the decay constant.

From above differential equation, we can get the activity at end of the bombardment:

$$A = N_0 \cdot \sigma \cdot \phi \cdot [1 - \exp(-\lambda \cdot t_{irr})]$$

where A is the induced activity, and t_{irr} is the irradiation time.

Using this equation the time normalization factor (N_{time}) can be expressed as follows:

$$N_{time} = \frac{[1 - \exp(-\lambda \times t_{irr})]}{[1 - \exp(-\lambda \times 1 \text{ hour})]}$$

We can normalize the activity by multiplying with the normalization factor (N_f):

$$N_f = \frac{p}{m \cdot I} \cdot \frac{[1 - \exp(-\lambda \times t_{irr})]}{[1 - \exp(-\lambda \times 1 \text{ hour})]}$$

where p is the purity of the sample, m is the mass of the sample, I is the beam intensity (μ A) during the irradiation and t_{irr} is the irradiation time.

5.4 Statistical & Systematic errors

The global uncertainty of the measured activity takes into account systematic and statistical errors.

5.4.1 Systematic errors

The systematics errors include the uncertainty associated to different evaluation of the counts in the peaks, the uncertainty on the detector efficiency, the uncertainty on the efficiency curve, the uncertainty on the beam intensity, the uncertainty on mass of the samples and the uncertainty on the standard source.

The mass of ^{nat}Al , ^{nat}Zr , and ^{nat}Cu activation foils were provided by the company Shieldwerx, which have a precision of 10^{-4}g . The mass of the other samples was measured once by using two electronic scales, which have a precision of 10^{-3}g and 10^{-5}g respectively. The uncertainty of the mass is equal to the smallest increment.

The systematic error in the efficiency fit was taken as 4.97 % (the biggest deviation of the original values from the ones given by the fit). Moreover, another 1.68% is coming from the systematic error in the activity of the standard source.

The uncertainty of beam intensity was different according to the specific run (Table 12). In some cases (runs) the beam intensity was very unstable and in other cases the beam intensity was very stable, with a global systematic error less than 2%. During the runs with deuteron beams the systematic error was of few percentage, which was less stable than the runs with a proton beams.

Table 12: The uncertainty on the beam intensity

Run	Beam type	Error (%)	Run	Beam type	Error (%)
2	p	0.5	11	p	1.15
3	p	22.5	12	d	6.88
4	p	0.79	13	p	0.49
5 (1)	p	0.38	14	p	0.57
5 (2)	p	0.53	15	p	0.71
6	p	1.07	16	p	0.71
7	p	0.43	17	p	0.68
8	p	0.78	18	d	2.48
9	p	0.47	19	d	3.25
10	d	1.04	20	p	1.34

5.4.2 Statistical errors

Statistical uncertainty of gamma-ray peak was less than 1%, however some of the few observed peaks have a high statistical uncertainty due to its low count rate.

The statistical uncertainty is the uncertainty in the gross area and the uncertainty in the background added in quadrature (ORTEC).

5.4.3 Decay data uncertainty

The decay data and uncertainty of the decay data were taken from NuDat2.0 database (National Nuclear Data Center). The decay data means half-life, γ -ray intensity and branching ratio.

5.4.4 Self-absorption correction

Because of the high density of some materials, γ -rays are attenuated; it means a self-absorption correction would be necessary to apply for ^{nat}Lu , ^{nat}Re and ^{nat}W activation foils. This kind of analysis could be performed using preferably MCNPx code.

5.4.5 Global uncertainty

All these components added quadratically result in a value shown in Table 13 for all the runs in the FESTA experiment.

Table 13: Systematic errors in the calculation of the activity

Source	Error
Beam intensity	0 – 22.5%
γ -ray peak	0 - 50 %
Standard source	1.68%
Efficiency curve	4.97%
Mass	< 2 %
Decay data	0 - 7.5 %
Total	5 – 50 %

5.5 MonteCarlo Simulation codes

In order to reproduce the experimental data, Monte Carlo simulations were performed. Originally, only FLUKA was considered for the simulation campaign runs with proton and deuteron beams, however because of the inconsistency of FLUKA with the deuteron beam we have considered the use of other MC codes like PHITS and GEANT4.

SimpleGeo program (C. Theis, et al.) was used for converting FLUKA geometry into PHITS (or MCNP) geometry.

FLUKA & PHITS were used for the runs with a 60 MeV proton beam and only PHITS for the runs with a 30 MeV deuteron beam.

For all of the calculations in this report, the intensity of beam was assumed as 1 μA . The calculated physical quantities using MC simulations are particle fluxes, residual dose equivalent rates (FLUKA only), production yields and activities of the irradiated samples.

5.5.1 FLUKA

FLUKA is a particle physics Monte Carlo simulation package (A. Ferrari, et al.) & (T. T. Böhlen, A. Ferrari, P.G. Ortega, et al.). It has many applications in high-energy experiments, engineering, shielding, detector and etc. For low energy neutron (< 20 MeV), FLUKA uses ENDF/B-VII.1 data library and it is 260 group data. Ion split option was used for deuteron beam.

FLUKA can simulate with high accuracy the interaction and propagation in matter of about 60 different particles, including photons and electrons from 1 KeV to thousands of TeV, neutrinos, muons of any energy, hadrons of energies up to 20 TeV (up to 10 PeV by linking FLUKA with the DPMJET code) and all the corresponding antiparticles, neutrons down to thermal energies and heavy ions. The program can also transport polarised photons (e.g., synchrotron radiation) and optical photons. Time evolution and tracking of emitted radiation from unstable residual nuclei can be performed online ([Fluka Manual](#)). In Chapter 1, Section 1.1 Physics, of this Manual we can find that FLUKA hadron-nucleon interaction models are based on resonance production and decay below a few GeV, and on the Dual Parton model above. (A. Ferrari et al.)

The number of particles, which can be transported by FLUKA, was 25 in 1990; after the muon (anti) neutrinos and several hyperons were added, the number increased to 36. In 1992, transport of light ions (*deuterons*, tritons, ^3He , alpha) was introduced, without nuclear interactions. In 1996 tau leptons and neutrinos transport (and in some cases interactions) were added. In 1999 the transport of optical photons was set up. The same year charmed hadrons brought the total number of transported particles to 63, plus all kinds of heavy ions. The transport threshold, originally fixed at 50 MeV has been lowered since 1991 to the energy of the Coulomb barrier. Below that value, charged particles were ranged out to rest.

5.5.2 PHITS

PHITS (The Particle and Heavy Ion Transport code System) is a Monte Carlo simulation code, developed by JAEA (T. Sato et al.). It has been applied to estimate particle fluxes or radiation dose in accelerator design, radiation therapy and cosmic ray science, etc.

The Liège intra-nuclear cascade model (INCL) (A. Boudard et al.) is used for the calculations. INCL with Distorted Wave Born Approximation (DWBA) option (S. Hashimoto et al.) is also used for the deuteron beam case.

For the neutron transport and interactions, JENDL-4.0 cross-section data is used, and ENDF/B-VII.1 was used for some materials like Holmium, Rhenium and Lutetium.

In FESTA, PHITS 2.76 latest version was used.

5.5.3 Modeling of the Be Target

The beryllium target in simulation on Figure 36 is fixed as 2 cm thick and 4 cm diameter for proton and deuteron beams. This thickness is suitable for 60 MeV proton beam, but it is very thick for a 30 MeV deuteron beam.

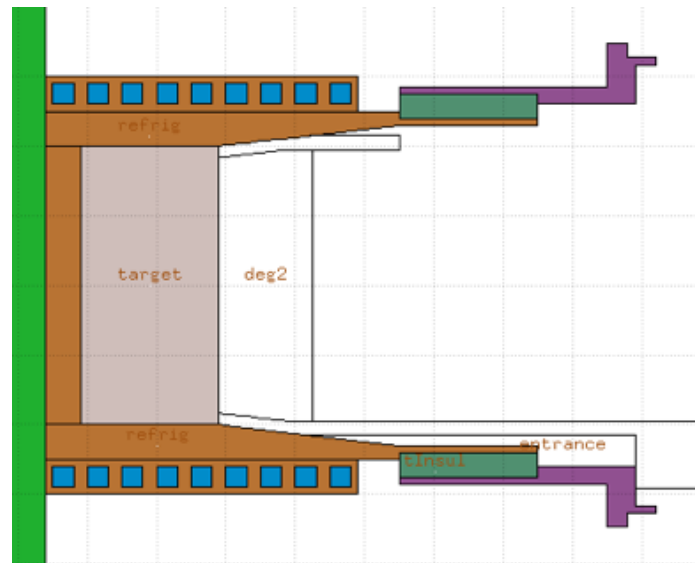


Figure 36: Geometrical model of the Be target (FLUKA).

Stopping power is one of the important parameters for the design of the target, because due to the charged particle beam loses its kinetic energy inside of the target and produce heat. In addition to this, it decides the projected range. The target needs enough thickness to stop the incident particles. A computer program Stopping and Range of Ions in Matter (SRIM) was used for the calculation of the stopping power and the projected range.

The density of beryllium is $1.8480 \text{ g/cm}^3 = 1.2349 \times 10^{23} \text{ atoms/cm}^3$ for the SRIM calculation.

The projected ranges in beryllium are 20.34 mm and 3.31 mm for 60 MeV proton and 30 MeV deuteron beams respectively. Figure 37 and Figure 38 show SRIM results for the stopping power of beryllium and the projected range in beryllium.

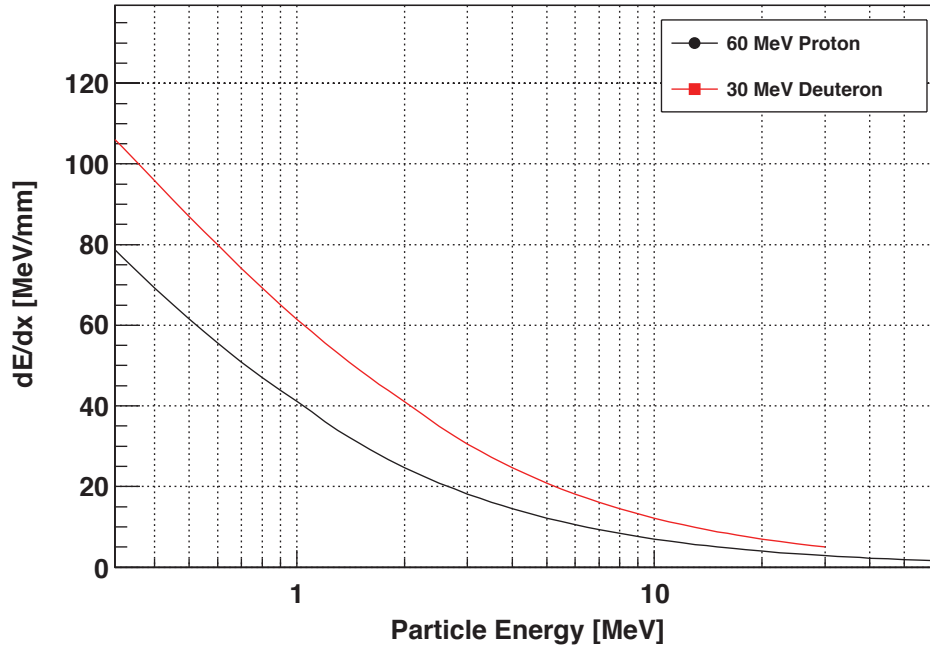


Figure 37: Stopping power of the Be target.

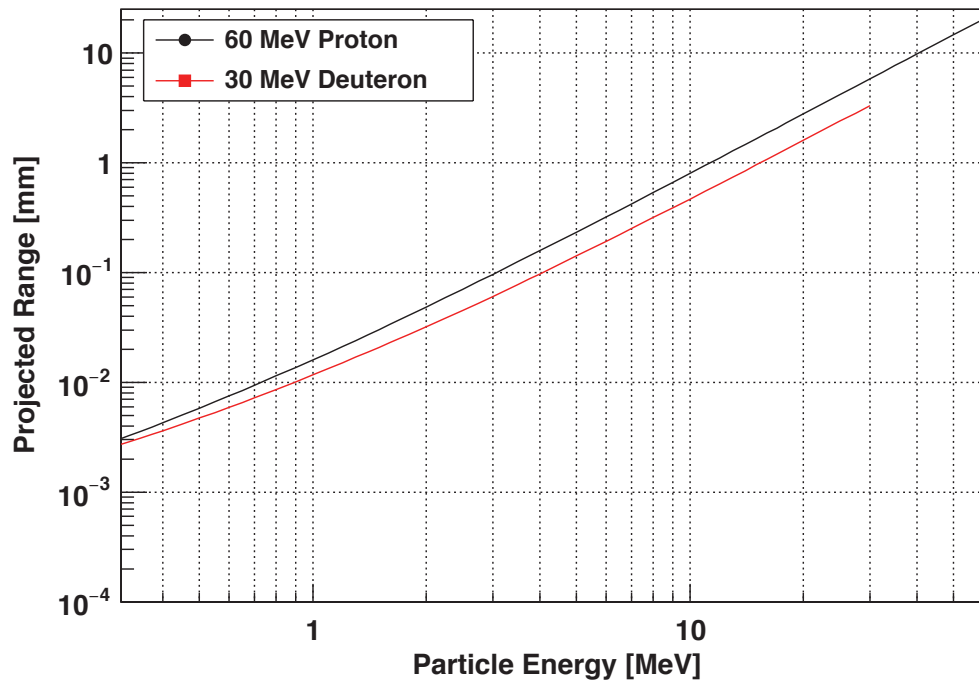


Figure 38: Projected ranges of the Be target.

5.5.4 Modeling of the Activator

The geometry model used in the simulation code is shown in Figure 39. The lead buffer size is $60 \times 60 \times 80 \text{ cm}^3$ and it is coupled with graphite reflector and polyethylene layers.

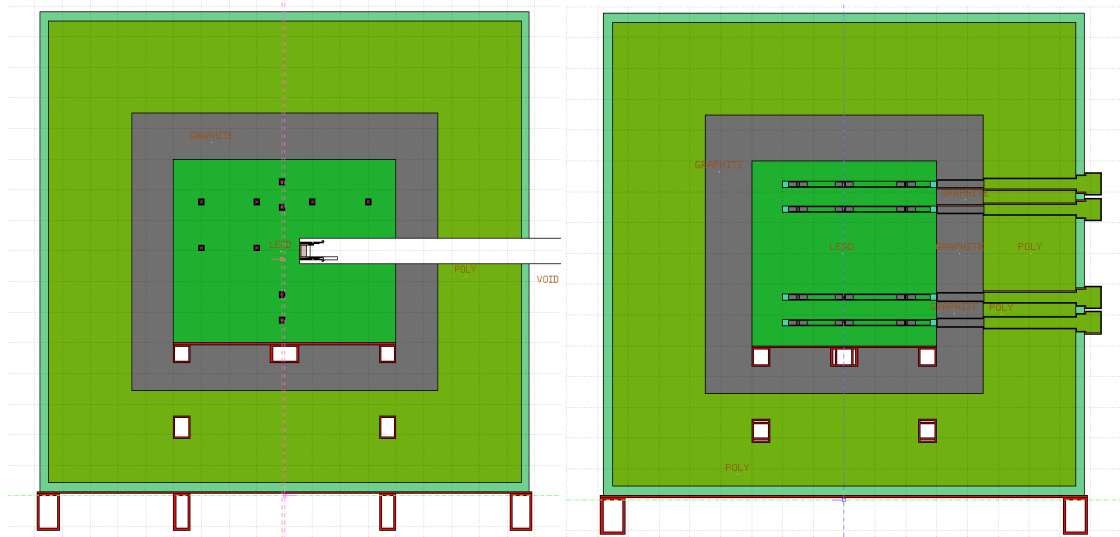


Figure 39 Geometrical model of the Activator as simulated with FLUKA.

5.5.5 Sample positions

There are 30 sample positions inside the Activator, all accessible via Al tubes (sample holders). Each tube carries three samples and is numbered from 1 to 10. The samples inside the tube are located in three different positions from the beam center ($x = 0$) as shown in Figure 40 & Figure 41.

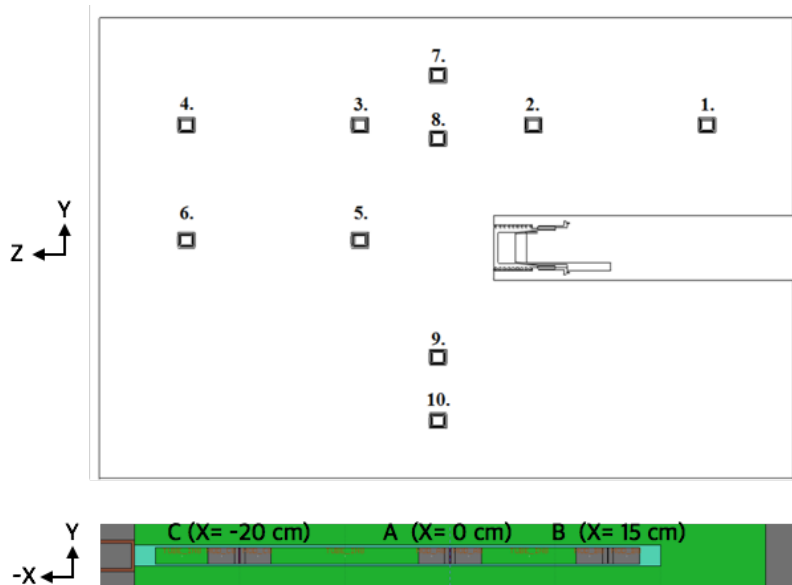


Figure 40: Side view of the Activator (10 holes) (FLUKA).

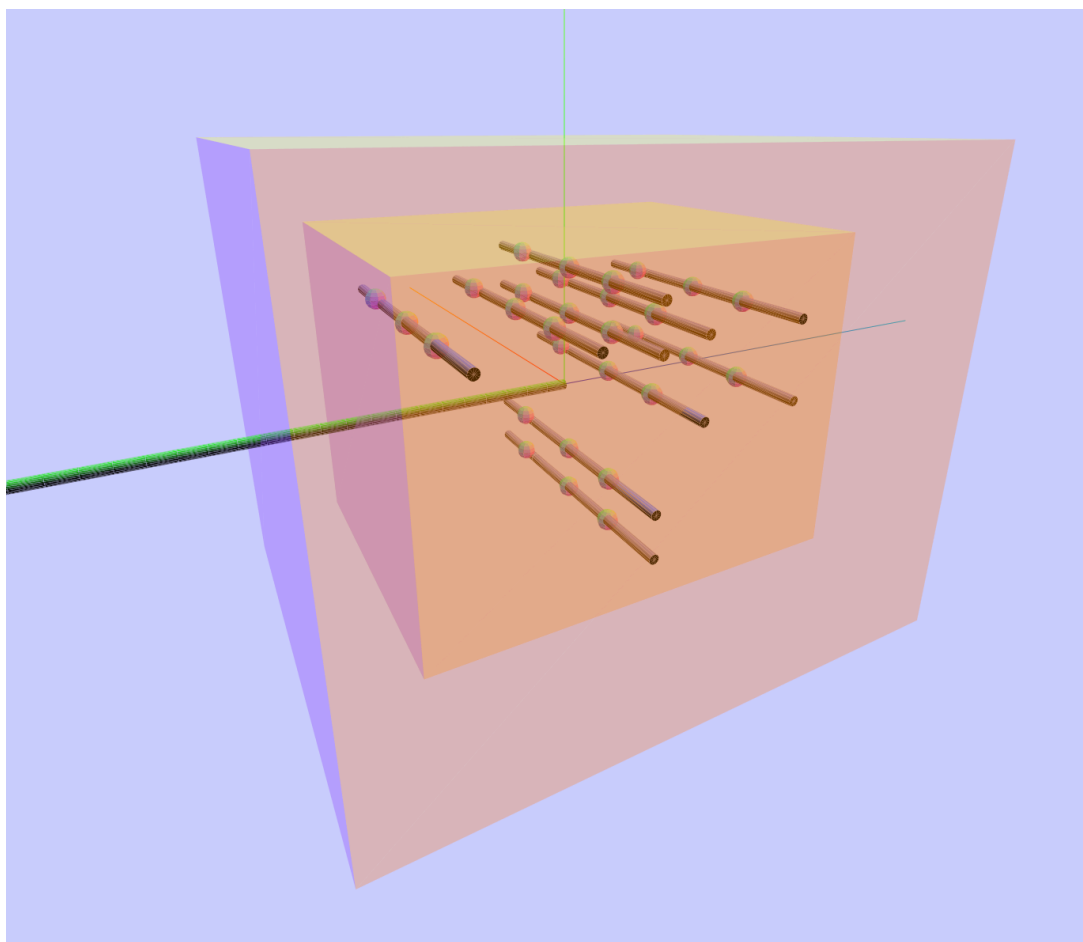


Figure 41: Side view of the Activator (10 holes & 30 positions) (3js).

5.5.6 Special study using FLUKA with deuterons

A full report (Luis, Raul) describing the neutron characteristics of the FESTA Activator using FLUKA with 15 MeV deuterons and 40 μA of beam intensity is available in Annex 1. This report evaluates the fluxes and power deposition in the target system and in the shielding assembly. It is important to remark that the neutron flux is estimated to be 10^{14} neutrons/ cm^2/s in the Be target and then decreases to approximately 10^6 neutrons/ cm^2/s after the shielding region. The activity of ^{99}Mo , one second after the EOB at 15.3 cm from the target, was also evaluated at 6.8×10^7 Bq/ cm^3 hour irradiation @ 40 μA .

Four configurations were tested in this report, using two different thicknesses for the ^{98}Mo targets (0.1 and 1 cm) and two different thicknesses of the local reflector/moderator (2.5 cm and 5 cm). The results show that there is no advantage in using 1 cm thick targets: increasing the thickness of the targets increases the total activity but decreases the specific activity.

Concerning the size of the local reflector/moderator, the choice between using 2.5 cm or 5 cm was not solved. For some positions, there is advantage in using 5 cm of graphite on each side of the targets. In other positions, the activities are higher with 2.5 cm of graphite.

5.5.7 Neutron fluence

Neutron fluence should be understood as the integral of the flux ($\phi = V \times n$) over a certain period of time. The flux ϕ is a quantity that multiplied by the macroscopic cross section $\Sigma = N \times \sigma$, for the process considered (N is the number of scattering centers per unit volume and σ is the cross section for the process) one obtain the neutron reaction rate per unit of volume.

We are interested in the neutron fluence produced by proton (deuteron) beams of a given energy. In addition, it is very important in FESTA to reproduce, in the same way as was done in TARC experiment (Arnould, 1999), the neutron fluence (Figure 42) as it is extracted from the resonance activity at energies 1.46 eV, 4.96 eV, 18.9 eV, from activation foils as a function of radial distance from the center of the lead volume. It is understood that for both experiments the conditions are different, in fact in TARC it was used 10^9 protons at 3.5 GeV/c.

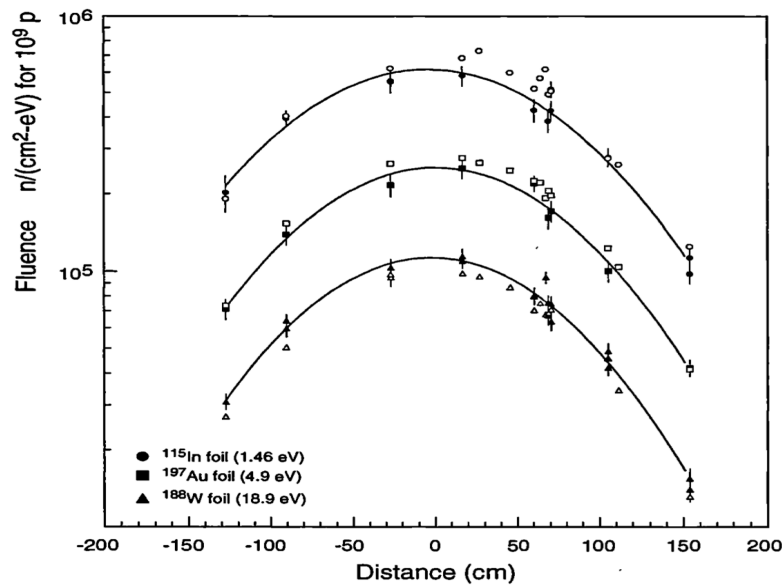


Figure 42: Neutron fluence in TARC Experiment (Arnould, 1999).

In FESTA we have recorded the number of interactions in the activation foils, therefore we can calculate the quantity $(1/\Sigma) \times (dN/dV)$, where dN is the number of interaction in volume element dV .

We used Monte Carlo simulation code FLUKA in the case of a 60 MeV proton beam, in order to make the comparison with the measurements. The complete geometry of the Activator has been totally included in the simulation. The Simulation also includes the Be Target system and neutrons created from a proton (deuteron) beams that collides on the Be target. These neutrons are traced until they reached the energy to be captured by the element or exit from the activator assembly.

The experimental data of the activation foils which are used to extract the neutron fluence at a given energy which is characteristic of the activated element, are described in Section 5.6.

The activation foils must be distributed over the whole volume of the activation region in order to make use of the whole flux and to avoid self-screening of the sample in the large cross section energy regions, which are the most efficient in the activation. This is why the sample capsules may need low-activation, neutron transparent materials like graphite.

Figure 43 shows the MC simulation of the neutron fluence in each part of the activator: Be Target, Lead diffuser, Graphite reflector, Polyethylene & Borated Polyethylene Layer, for no appreciable capturing sample.

The dependence of the neutron fluence on energy and space validates the neutron transport and tests the reliability of the neutron-induced cross-sections of lead.

We observe that the neutron flux is quasi iso-lethargic in the diffuser and graphite reflector regions, in the energy range between 0.1 eV and 1 KeV, which corresponds to the resonance region of interest for the ARC method as claimed in the Patent PCT/EP2014/069463.

In the case of lead diffuser the ratio between fast neutrons (> 0.1 MeV) and epithermal neutrons (1 eV to 100 KeV) is of the order of 10. One can also observe the sharp slowing down/moderation of the fast neutrons after exiting the Be Target (100 times less). This is due namely to the large (n, xn) cross section of lead, that contributes significantly to the multiplication on the neutrons produced in the target.

One can also verify that the polyethylene layer is quite efficiently reducing the quantity of neutron escaping the system (efficient neutron shielding).

If we compare Figure 43 & Figure 44, (n, xn) is less because with 30 MeV deuterons on Be the average neutron energy is less than in the case of 60 MeV on Be. Secondly we clearly observe the signature of deuteron break-up through the neutron peak at 15 MeV. The neutron flux has a mono-energetic peak at more than 10^{13} n/cm²/μC, which is very high, originating from the breakup of deuterons.

As for the remaining part of the Activator, the neutron flux distribution follows the same behavior as for 60 MeV protons on Be, but with the notable exception that the flux levels are 20 times higher in the case of 30 MeV deuteron as is predicted by FLUKA.

For PHITS (Figure 45 & Figure 46) the results are quite different and indicates:

- No more monoenergetic peak at 15 MeV, which possibly means a properly production of secondary particles induced by light ions nuclear reactions, allowing a drastic removal of the peak in the case of FLUKA simulation.
- However, 60 MeV protons produce 2-4 times more neutrons than in the case of 30 MeV deuterons, contrary to FLUKA simulation, which indicates that the codes do not treat data in the same way.

It is very interesting to note that the polyethylene layer (moderator) in both cases, has an iso-lethargy behavior, it is almost constant and energy independent in the region of epithermal energy of 1 eV to 100 eV.

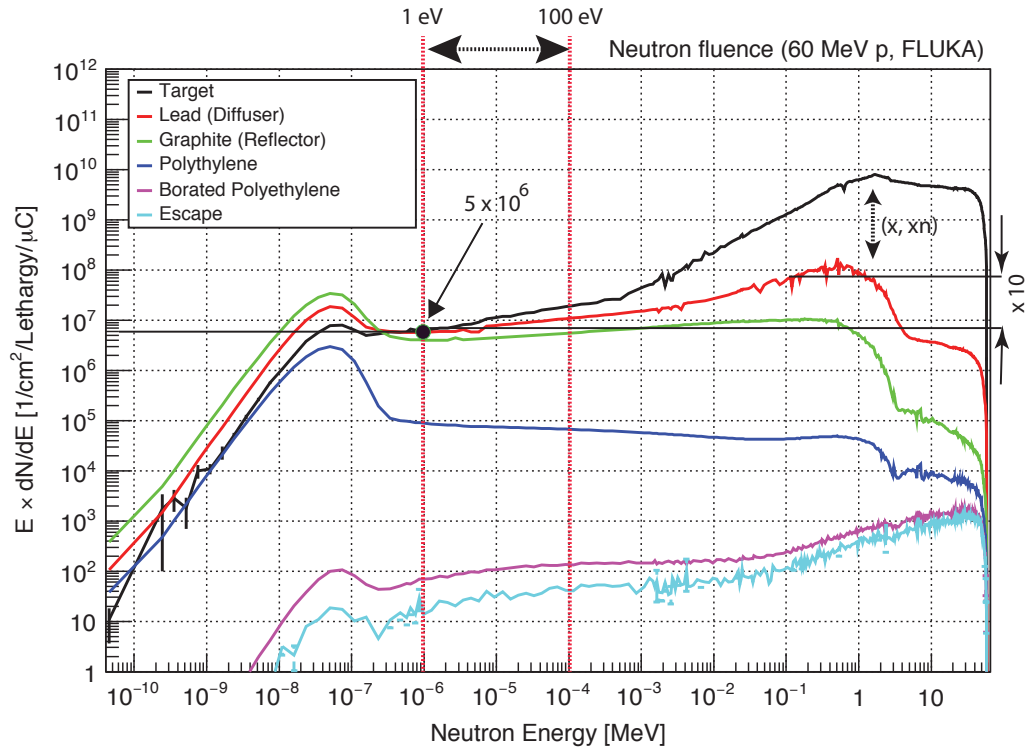


Figure 43: Neutron fluence (60 MeV protons with FLUKA).

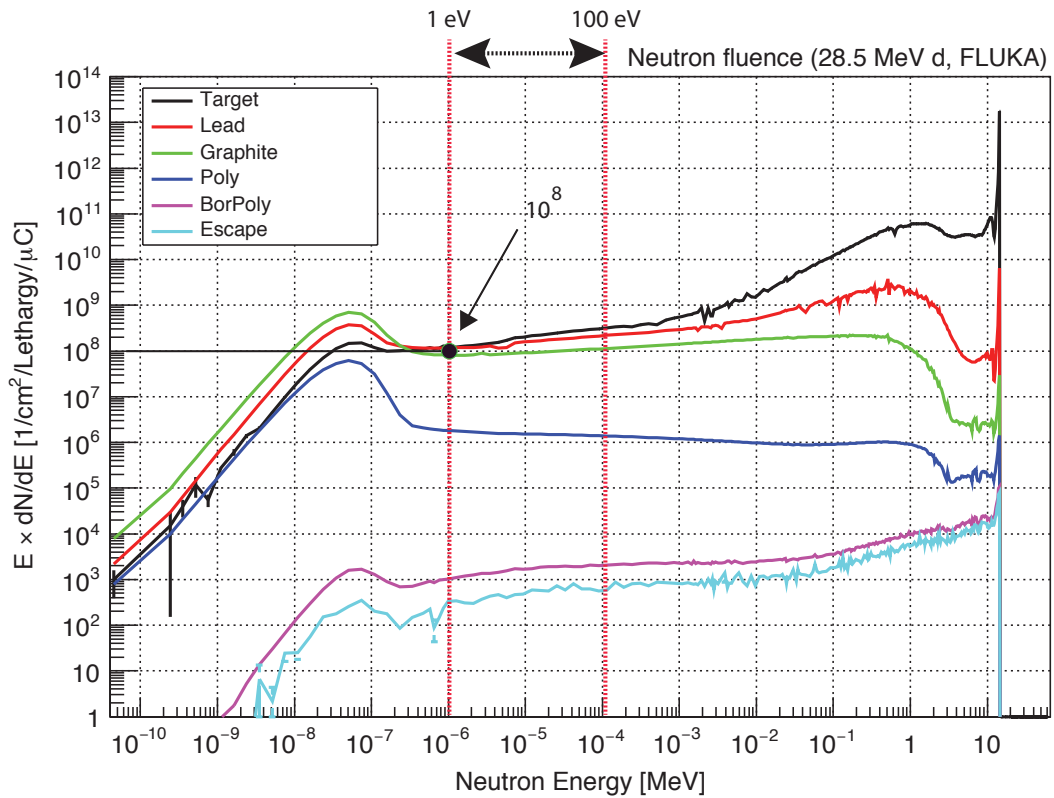


Figure 44: Neutron fluence (30 MeV deuterons with FLUKA).

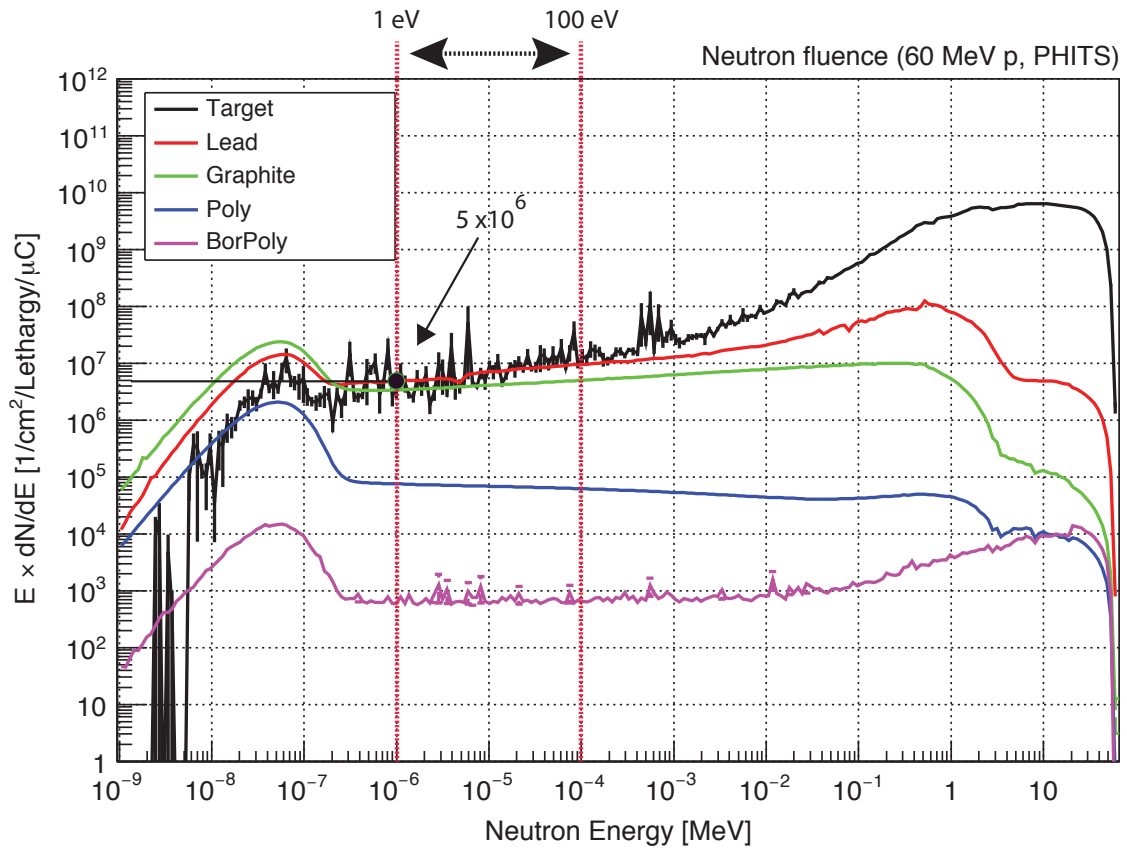


Figure 45: Neutron fluence (60 MeV protons with PHITS).

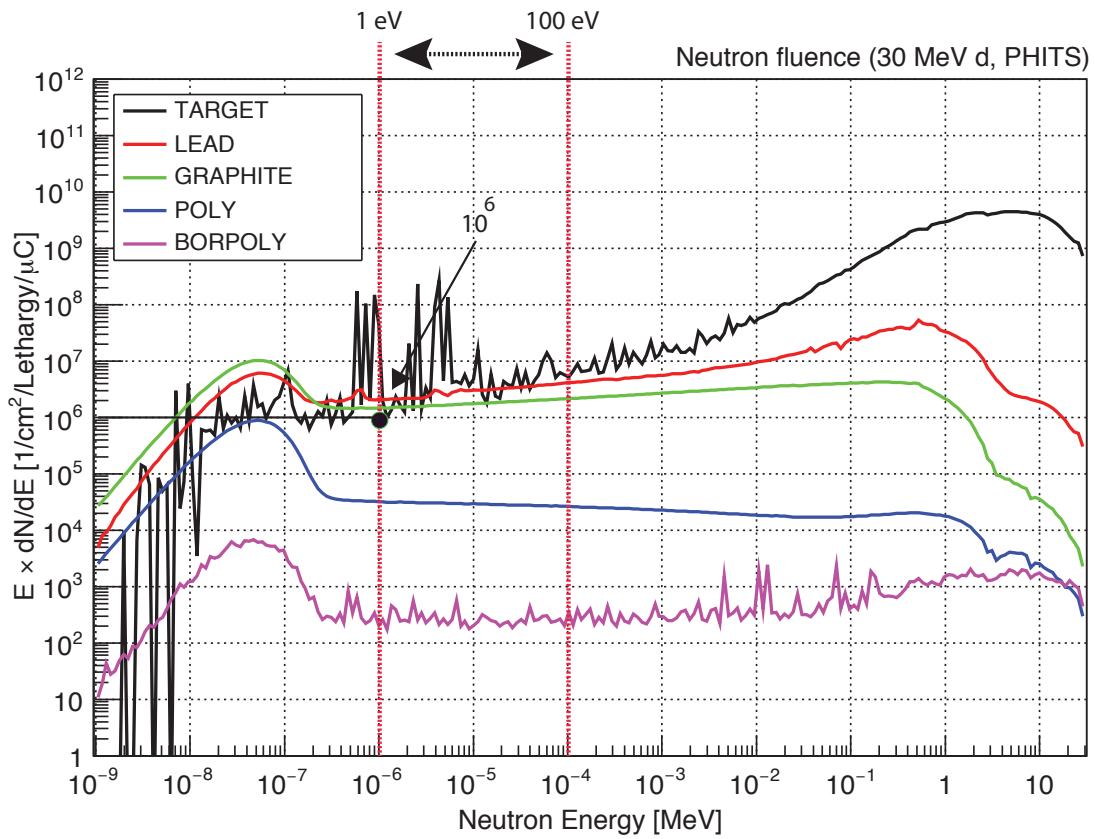


Figure 46: Neutron fluence (30 MeV deuterons with PHITS).

5.5.8 Energy time correlation

The energy time correlation shows the time required for a neutron to slow-down its energy to be thermalized. As we can see in the next figures it takes at least few μs for a neutron to be in the resonance region where the ARC method can take effect. A correlation exists between the slowing-down time and the neutron energy inside the lead block, $\bar{E}_n = K/(t + t_0)^2$, where $\bar{E}_n$ is the mean energy in KeV, K is the slowing-down constant in $\text{KeV} \times \mu\text{s}^2$, t is the measured time in μs and t_0 can be determined by simulations or experimentally, which in FESTA was not conducted. However, simulations were performed with FLUKA & PHITS, taking into account the lead impurities, the influence of the other layer materials in the Activator and finally the Be target unit. Experimentally, the values of K and t_0 depend, among other parameters, on the neutron mean free path in the propagating medium and can be measured by neutron capture on different samples. The gamma emission from the neutron capture allows us to measure the activity after each irradiation of the samples. Each sample used in FESTA has a large neutron capture resonances at characteristics neutron energies.

The neutron flux is an important parameter of the Activator that is necessary to determine a cross section. It can be simulated using the above-mentioned MC codes at different positions of the samples in the lead diffuser. At the end the results can be compared to measurements of the activities of the irradiated samples.

In Figure 47, one can also distinguish four neutron group families (1, 2 & 3). Neutrons produced in the Be target have an average energy close to 30 MeV (27 MeV according to Lone paper) (M. A. Lone et al., 1981).

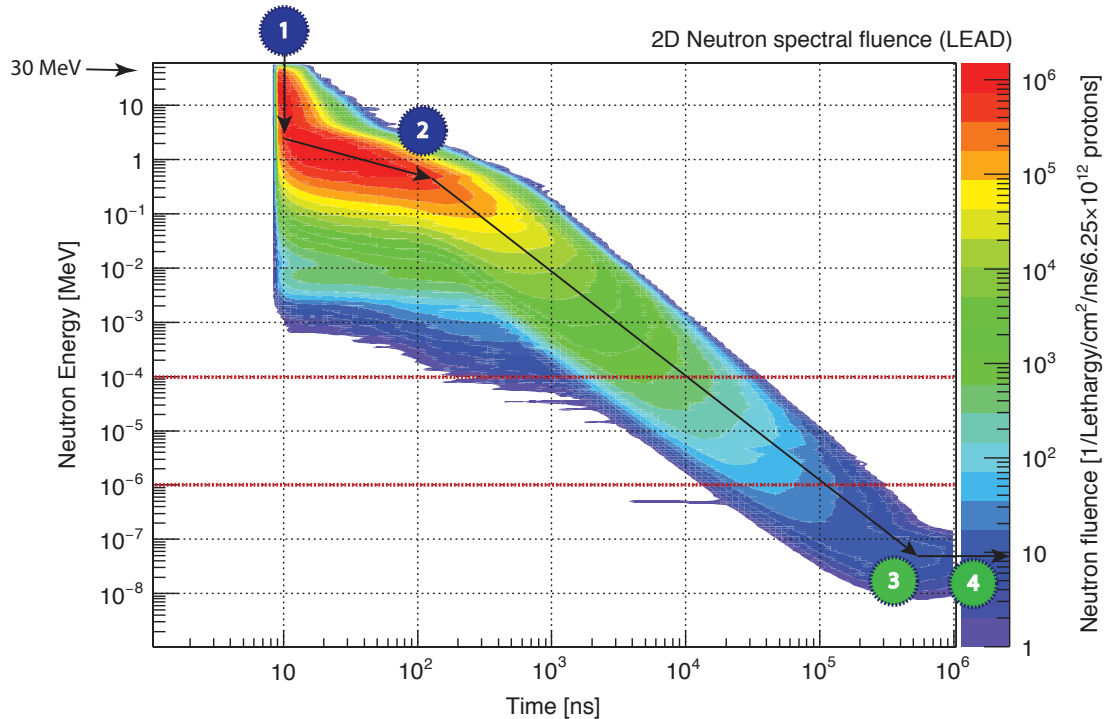


Figure 47: Energy-time correlation in Lead (60 MeV proton, PHITS).

Immediately after leaving the target, neutrons lose their energies by strong inelastic reactions (n, xn) in Lead from 30 MeV down to 1 MeV in few nanoseconds (between 10 & 20 ns) (step 1).

The same neutrons then diffuse adiabatically in the lead diffuser for as long as 100 ns and where their energies varies very slowly from 1 MeV to 0.1 MeV (step 2). Neutron multiplication occurs inside Lead at early stage (first 100 ns) due to the large ($n, 4n$), ($n, 3n$) and ($n, 2n$) reaction cross sections in Lead. One can deduce that after 100 ns neutron does not remember where it comes from, 60 MeV protons or 30 MeV deuterons.

After diffusion in Lead, the neutrons are then moderated in the graphite (step 3) layer, which corresponds to a rapid change of energy (4 decades in almost 10 μ s) compare to 1 decade in 100 ns in Lead.

Those neutrons are finally thermalized in polyethylene layer and only undergo elastic diffusion as where there energy remains constant until they are absorbed or escape the system (step 4) see Figure 48.

In case of deuteron beam (Figure 49 & Figure 50), The source of neutrons are less affected by the strong inelastic collisions in Lead which are only linked to ($n, 2n$) as opposed to ($n, 4n$) with 60 MeV protons on Be and because of the lower energy of source of neutrons (almost 12 MeV). As for the rest of the neutrons behave in a very similar way indicating that after 2-3 collisions neutrons in lead do not remember if they were produced from 60 MeV protons or 30 MeV deuterons.

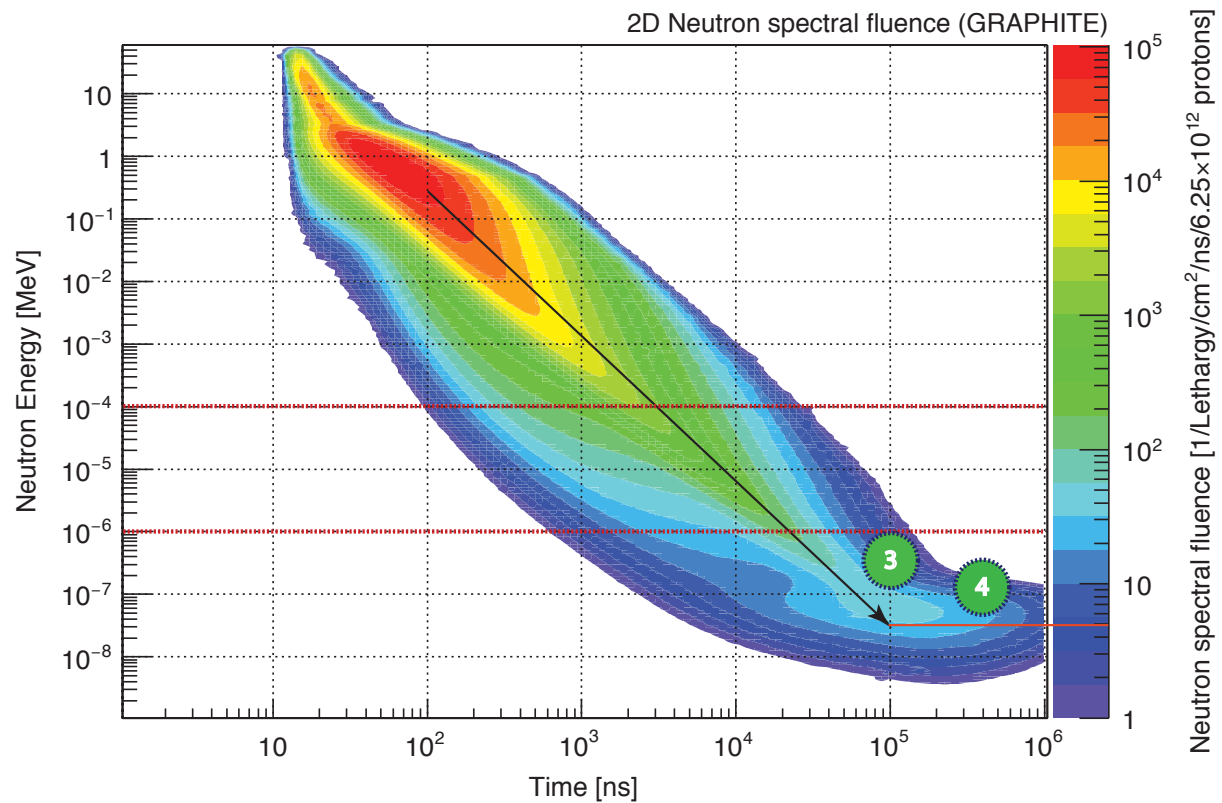


Figure 48: Energy-time correlation in Graphite (60 MeV proton, PHITS).

2D Neutron spectral fluence (LEAD)

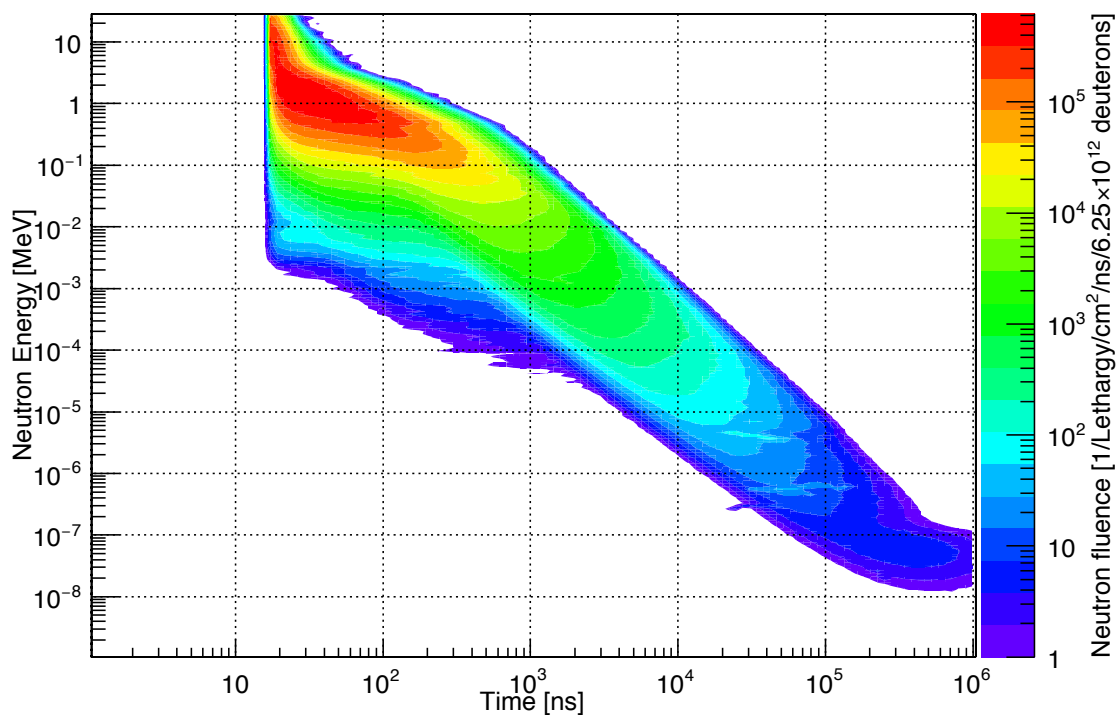


Figure 49: Energy-time correlation in Lead (30 MeV deuteron, PHITS).

2D Neutron spectral fluence (GRAPHITE)

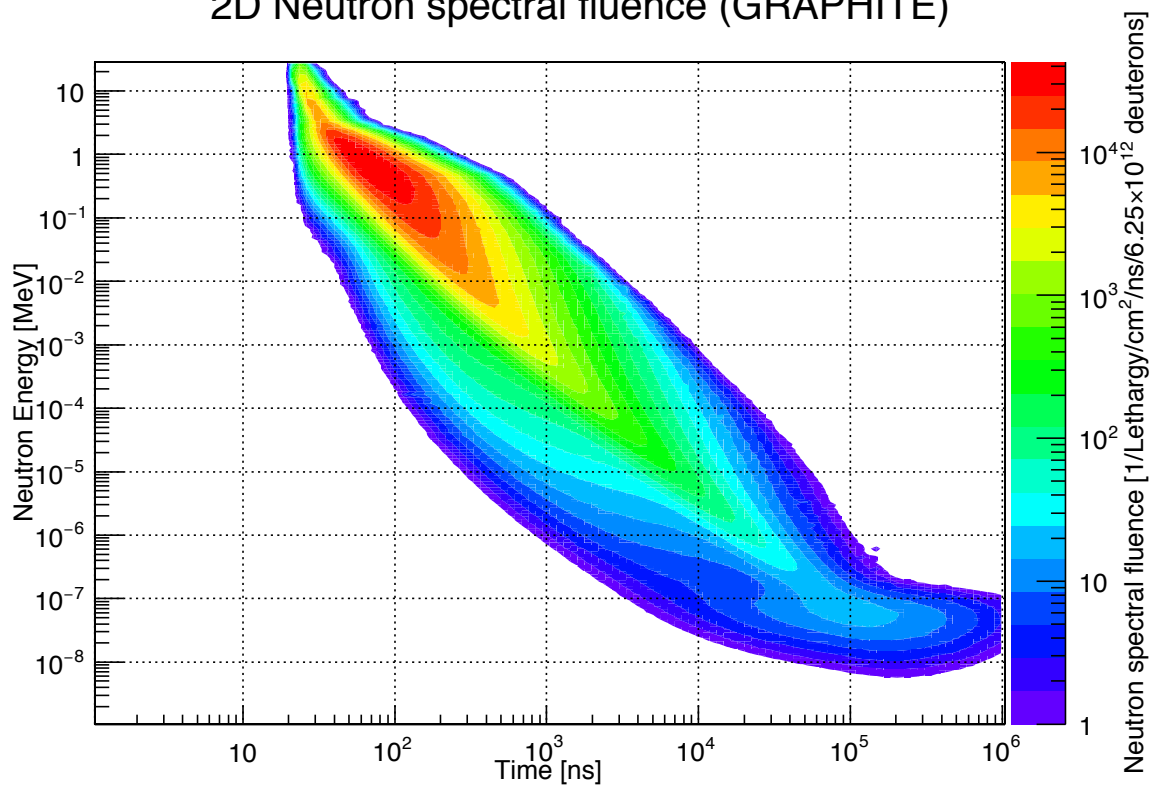


Figure 50: Energy-time correlation in Graphite (30 MeV deuteron, PHITS).

5.5.9 Ambient dose

Ambient dose calculation is important for planning the experiment and deciding a cooling time for accessing the activator after the beam irradiation. The structures like the concrete wall, the beam-line, the magnet, etc. are not included in this simulation. The beam current is fixed as $1\text{ }\mu\text{A}$ and the irradiation time is 1 hour.

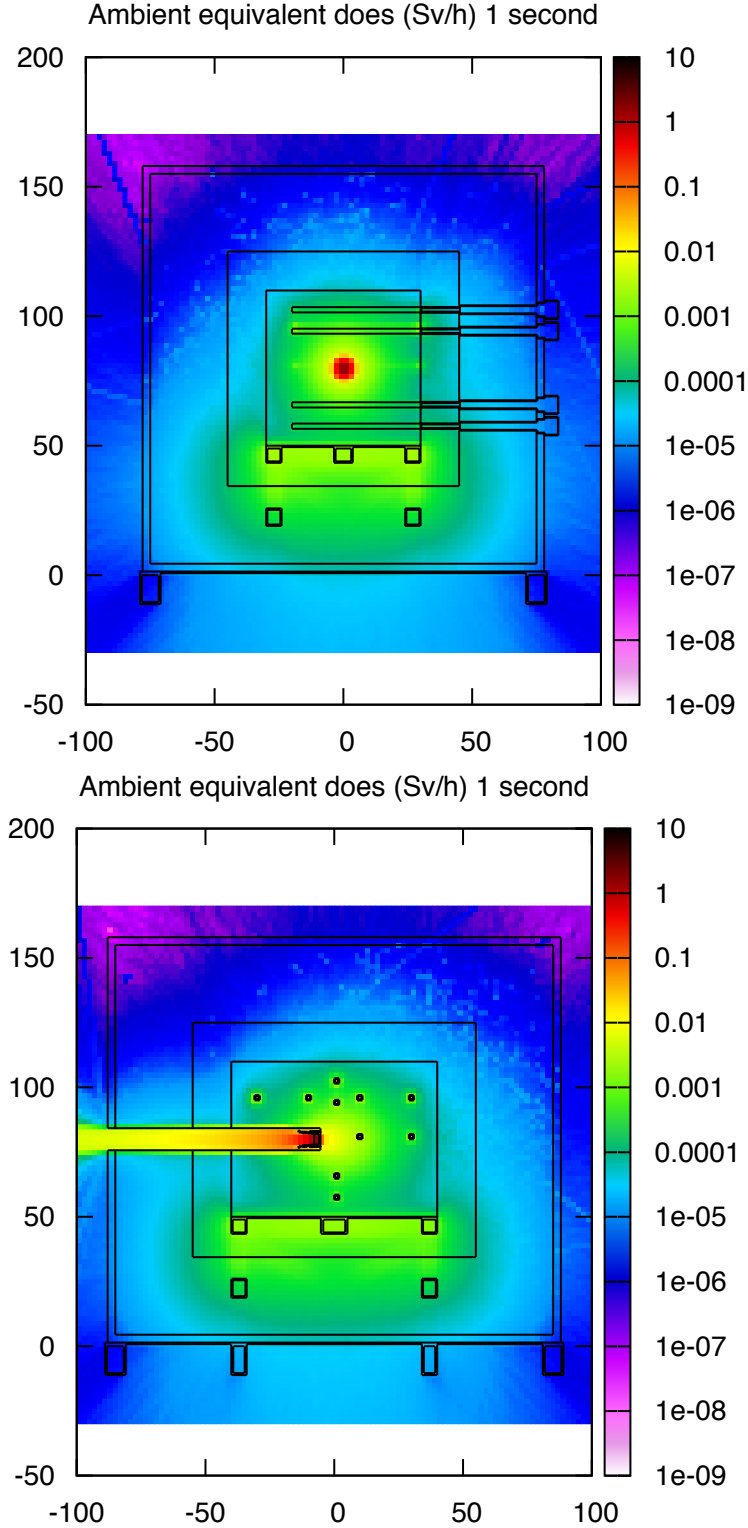


Figure 51: Ambient dose calculation dose at $T_{\text{cool}} = 1$ second after 60 MeV proton beam.

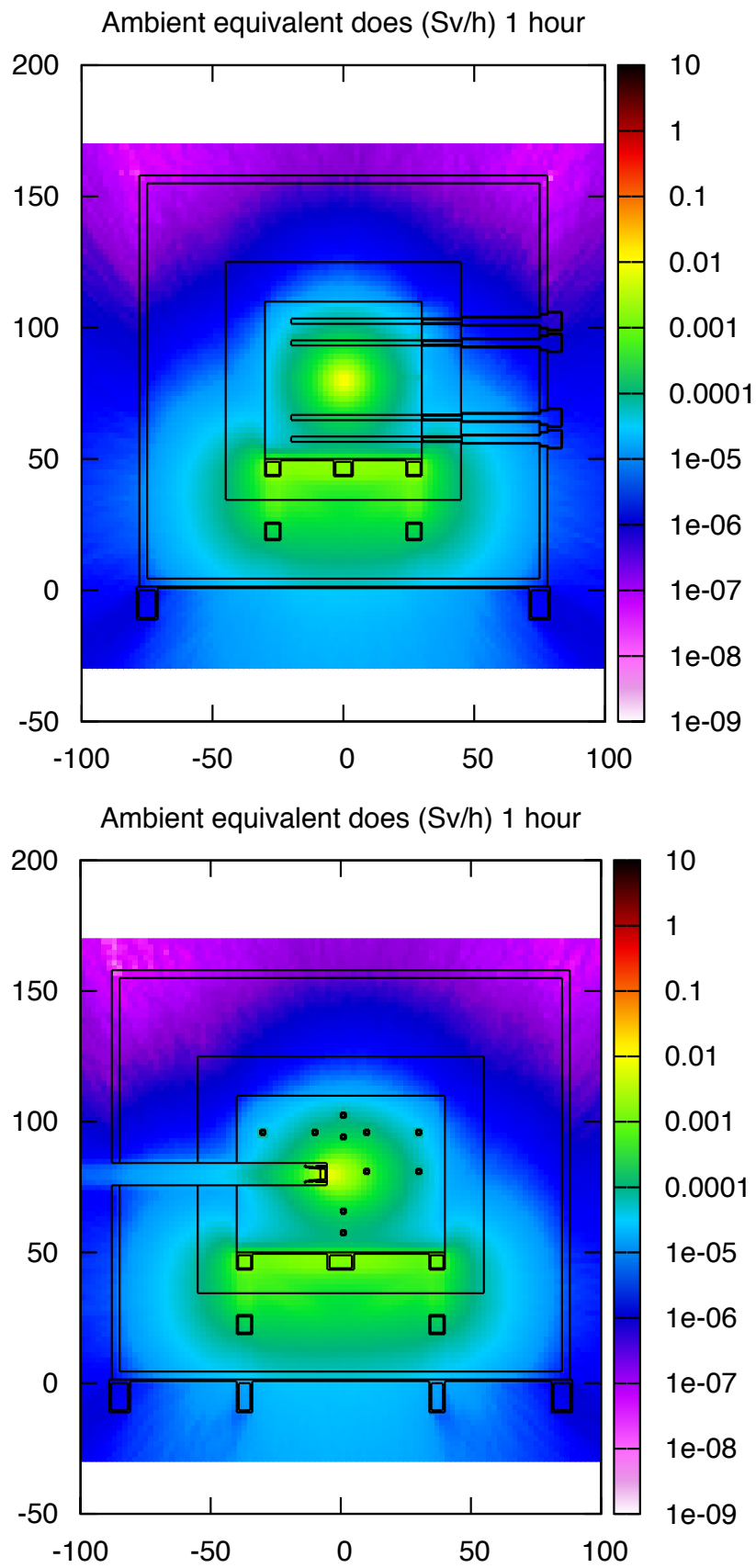


Figure 52: Ambient dose calculation dose at $T_{cool} = 1\text{ h}$ after 60 MeV proton beam.

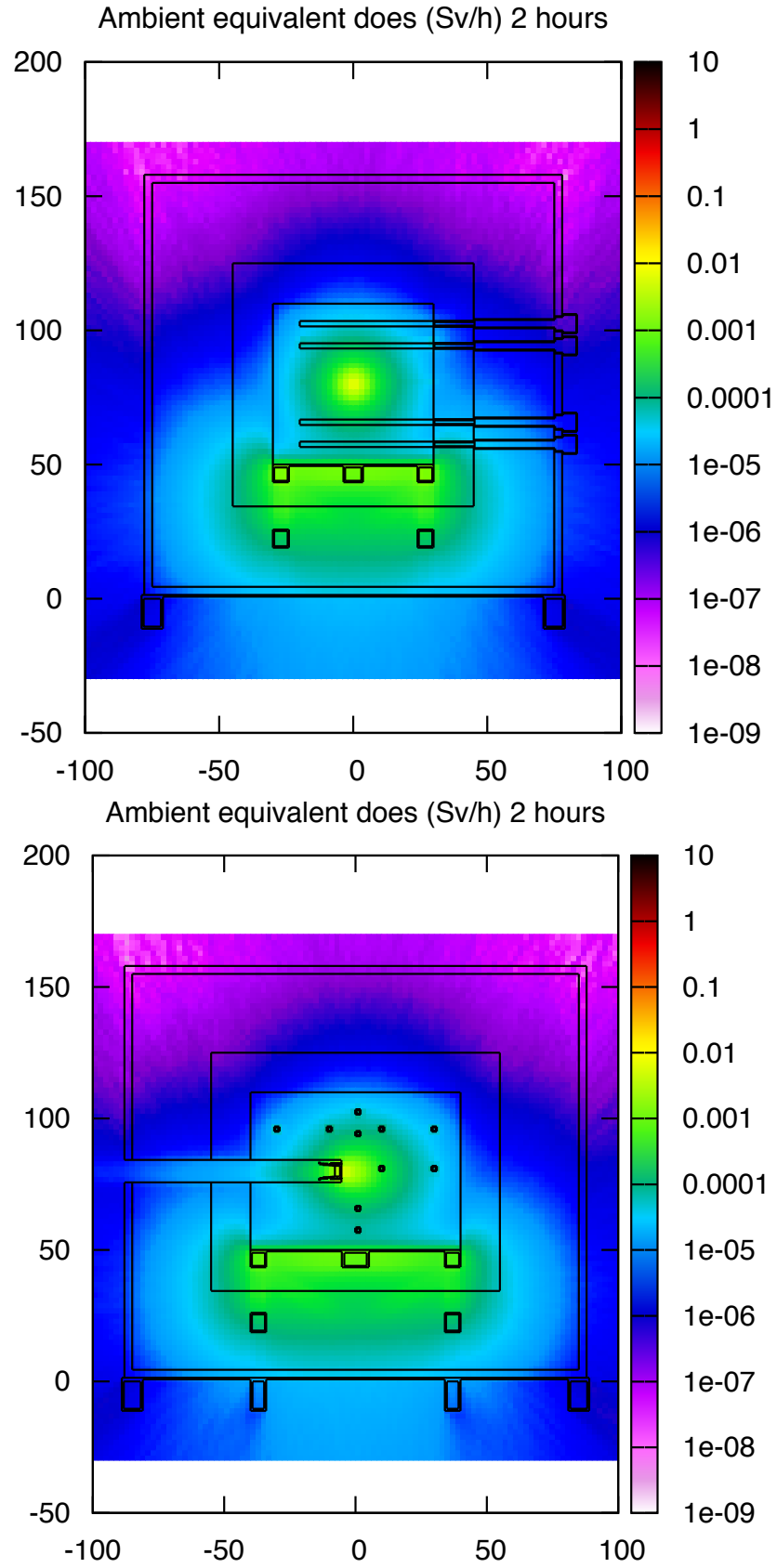


Figure 53: Ambient dose calculation dose at $T_{cool} = 2$ h after 60 MeV proton beam.

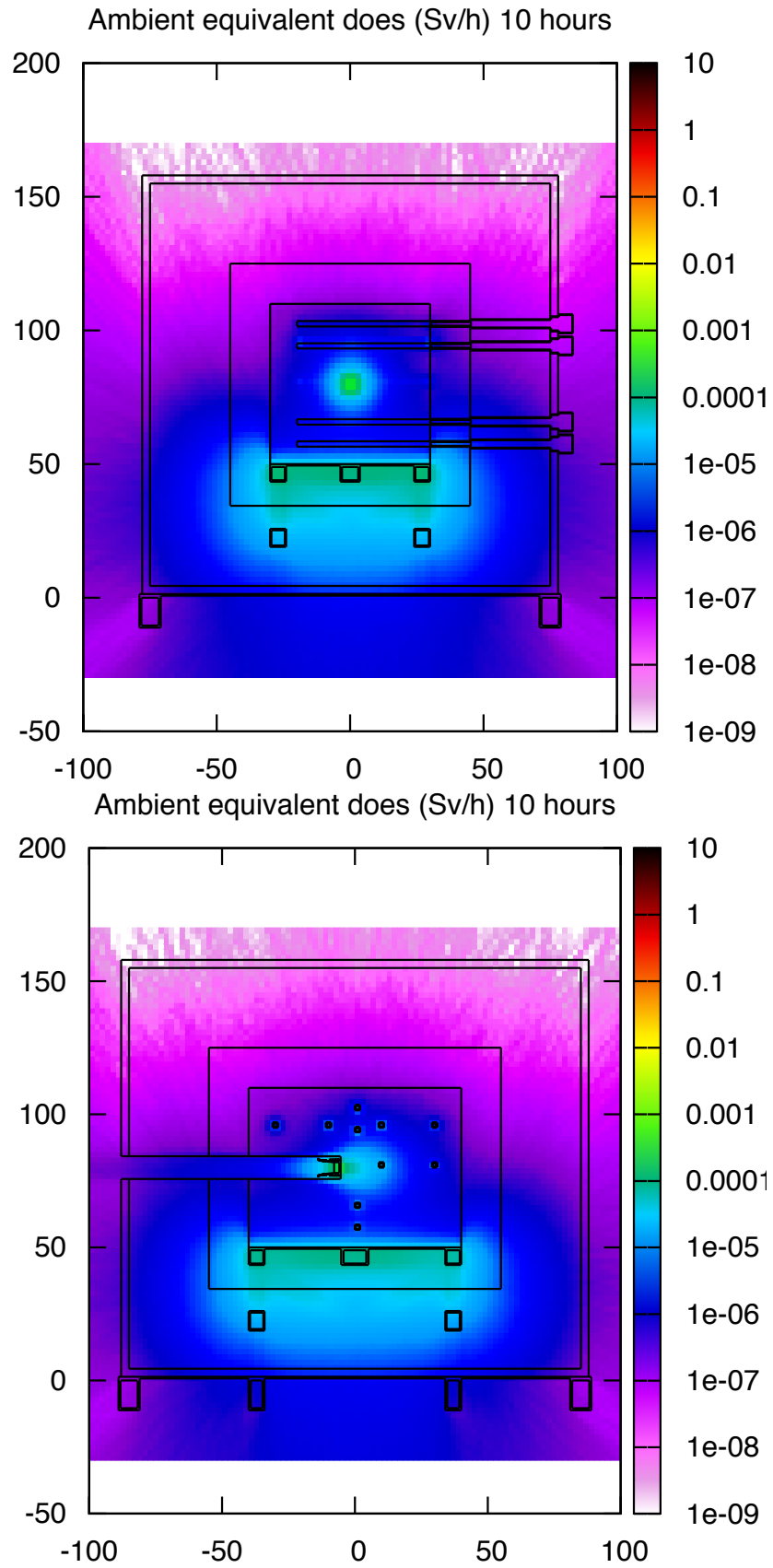


Figure 54: Ambient dose calculation dose at $T_{cool} = 10$ h after 60 MeV proton beam.

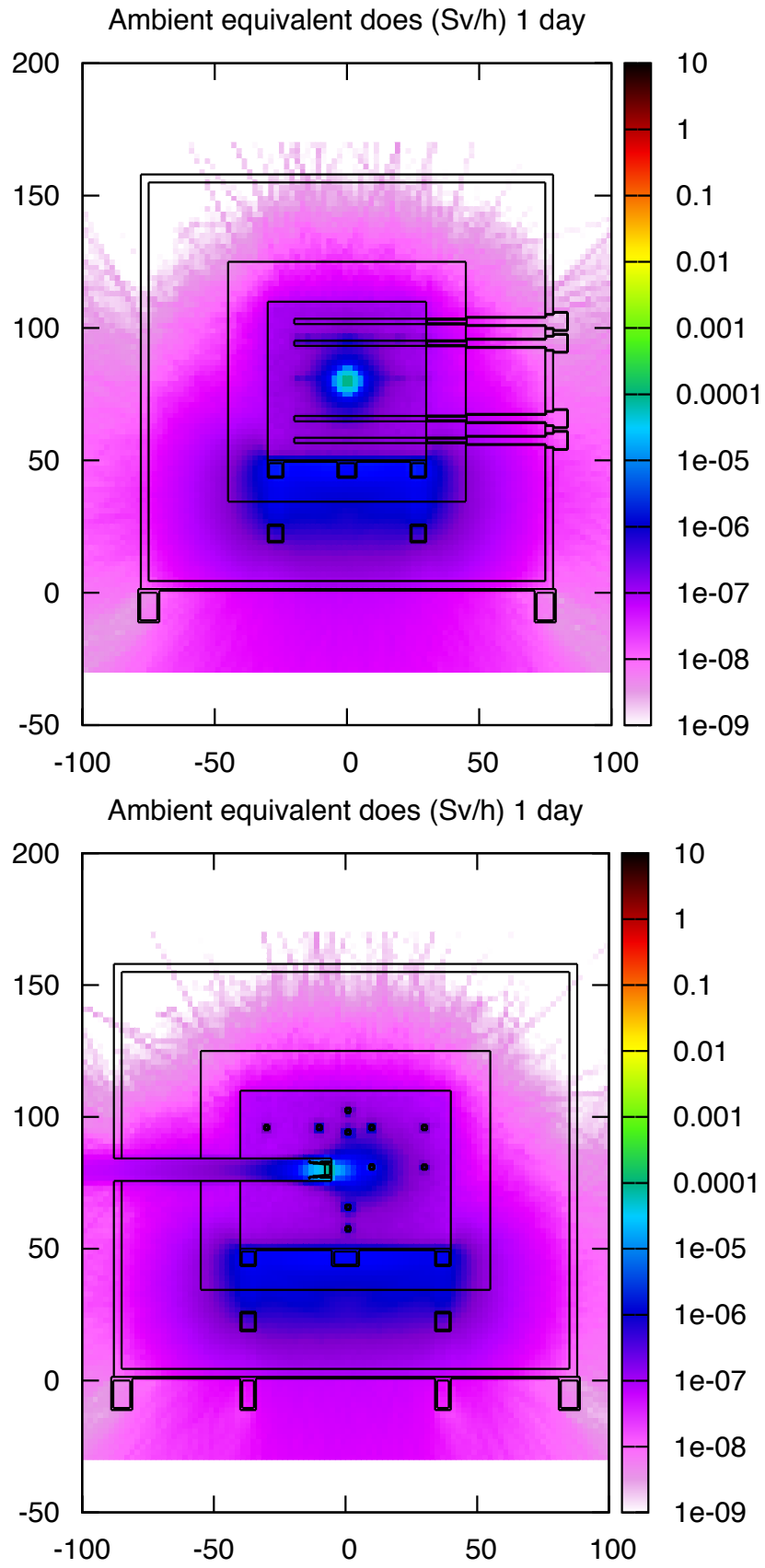


Figure 55: Ambient dose calculation dose at $T_{cool} = 1$ day after 60 MeV proton beam.

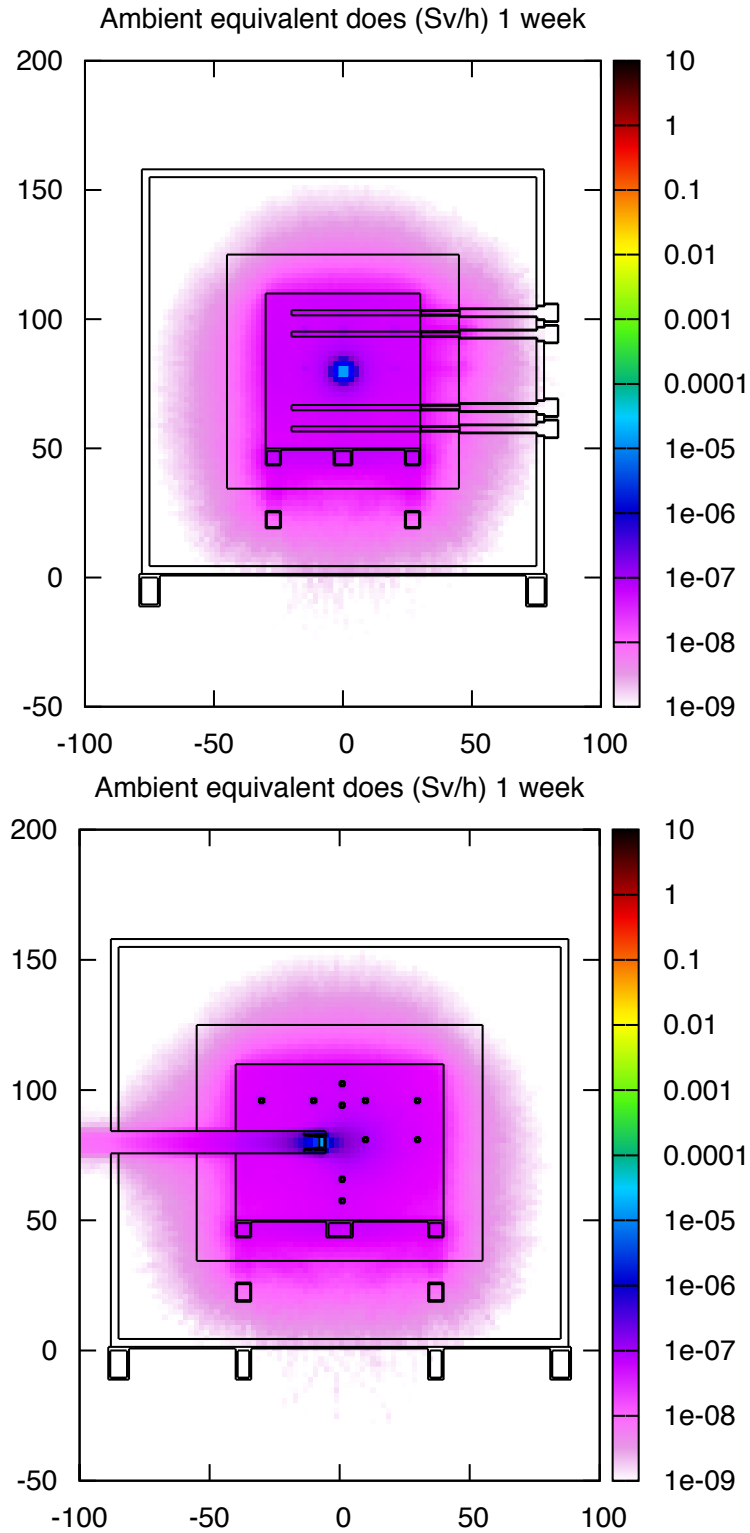


Figure 56: Ambient dose calculation dose at $T_{cool} = 1$ week after 60 MeV proton beam.

The MC Simulations indicate that one can have access to the irradiated samples 1 to 2 hours after irradiations where the dose levels have reduced to few tens of $\mu\text{Sv/h}$. This is validated by the independent measurements carried out before and after each irradiation. An example of the radioprotection control on each run is presented in Annex 2.

5.6 Neutron flux measurements

Activation foils are one of the neutron detectors types most widely used due to the following advantages: a) they take relatively little space; b) foils can be activated and measurements can be made at the convenience for the experiment; c) the presence of gamma rays or other types of radiation does not interfere with the neutron activation of the foils (Arnould, 1999).

In FESTA we have had the opportunity to work with MEDICYC (60 MeV proton beam and 30 MeV deuteron beams) a cyclotron dedicated to the proton therapy, within the context of a scientific collaboration, although the huge number of patients who come to CAL to be treated for ocular proton therapy.

The experimental results with a 60 MeV proton beam and 30 MeV deuteron beam are presented in this section for the different activation foils and medical isotopes.

The MC simulation results for 60 MeV proton and 30 MeV deuteron beams are also shown in this section.

5.6.1 Activation foil measurements

The Neutron Activation Analysis (NAA) method was used in this experiment for the study of different activation foils. The energies 1.46 eV, 4.96 eV, and 18.9 eV (see Figure 57, Figure 67 and Figure 75) have been chosen because at these energies ^{197}Au , ^{115}In , and ^{186}W exhibit strong resonances in their neutron absorption cross-sections. $^{\text{nat}}\text{Al}$ was used for testing the behavior of fast neutrons across the (n, α) reactions occurred when it is irradiated. These activation foils were placed in a graphite capsule and then inserted in the sample holder to definitely allocate them inside of the Activator lead diffuser at different positions from the center of the Beryllium target.

In the following sections, we will remark the agreement of the experimental data (obtained irradiating the samples with 30 MeV deuteron beam) with MC simulation results done with PHITS. Previous results obtained with FLUKA overestimate the experimental results, almost by a factor of 70 on average. Unfortunately this remarkable difference was observed at the time when we started to have the first experimental results using a deuteron beam.

In FESTA, the deuteron breakup effect was overestimated based on the FLUKA simulation results, and the scientific literature on previous experimental results, where the production of neutrons with deuterons appeared to be quite significant.

This forced us to carry out an exhaustive work of validation of Monte Carlo simulation codes by which we conclude that PHITS was best suited to accurately reproduce the experimental results of FESTA. We can also categorically confirm that FLUKA has not been able to reproduce the FESTA experimental results for a 30 MeV deuteron beam and we have reported it at CERN claiming that the deuteron transport model of FLUKA is not able to process the deuteron break-up effect. Due to

this inconsistency we had to reprocess again with PHITS, all simulations of the samples in the Activator.

Below, we present the experimental results obtained from the activities of irradiation of the activation foils with protons and deuteron beams.

5.6.1.1 Natural In Activation Foil

Natural Indium has two stable isotopes ^{113}In (4.29%) and ^{115}In (95.71%). The reaction $^{115}\text{In}(n, \gamma)^{116}\text{In}$ leads the ground state ^{116}In ($T_{1/2} = 14.1$ s), and two metastable states $^{116\text{m}}\text{In}$ and $^{116\text{m}2}\text{In}$ with $T_{1/2} = 54.29$ min and $T_{1/2} = 2.18$ s, respectively. $^{116\text{m}2}\text{In}$ with its short half-life decays into $^{116\text{m}}\text{In}$ immediately. While the β -decay $^{116\text{g}}\text{In}$ ground state to the ground state leads directly ^{116}Sn , the isomeric states decay of the excited states in ^{116}Sn (Shirley, 1978). $^{115}\text{In}(n, \gamma)$ cross-section is shown in Figure 57.

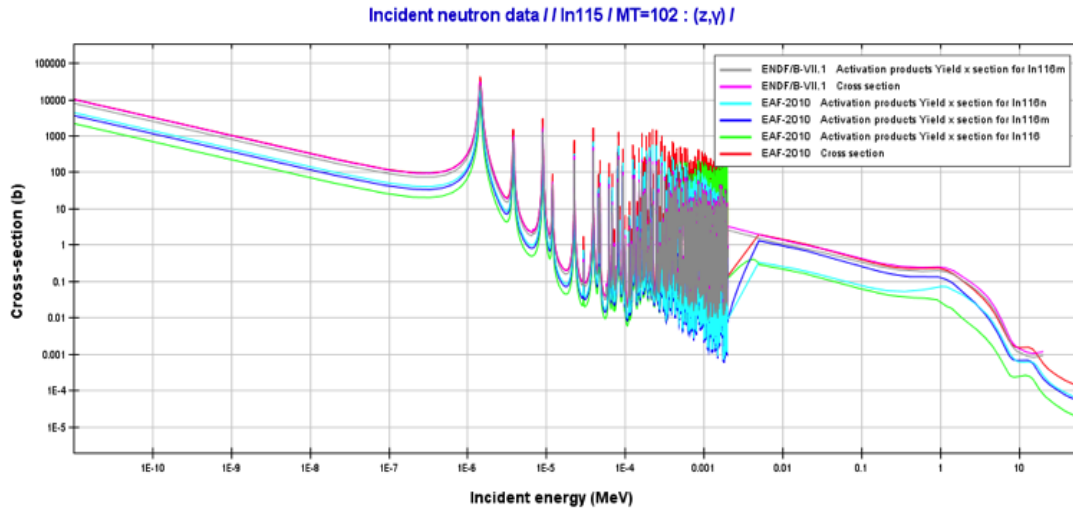


Figure 57: $^{115}\text{In}(n, \gamma)$ Capture neutron Cross Section.

The $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ reaction ($T_{1/2} = 4.486$ h) is used also for the measurement of the neutron flux of fast neutrons in the range of MeV region. The $^{115\text{m}}\text{In}(n, n')$ cross section is shown in Figure 58.

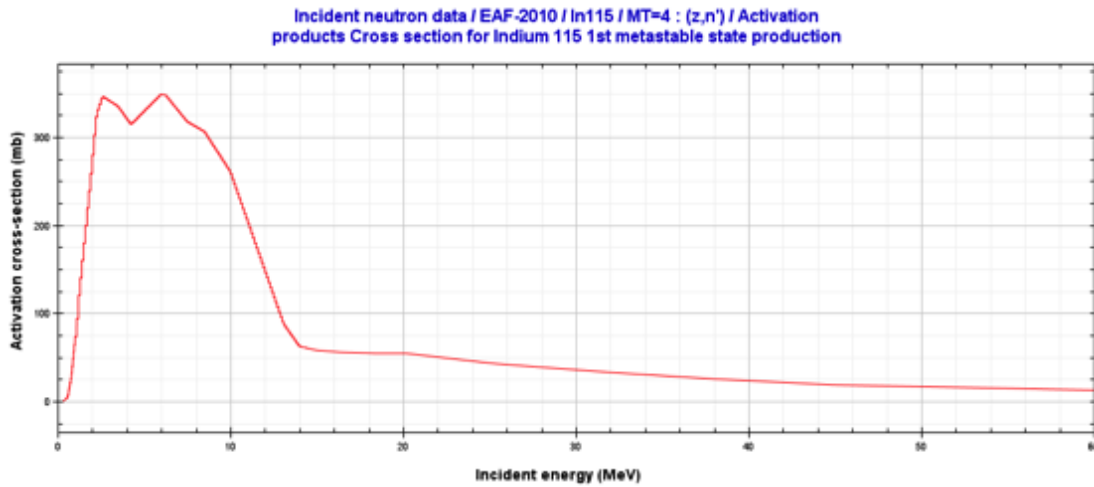


Figure 58: $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$ Cross Section.

^{Nat}In activation foils were placed in different positions inside of the Activator. Figure 59, Table 14, show the positions of the activation foils in case of proton beam and Figure 60 & Table 15 in case of deuteron beam.

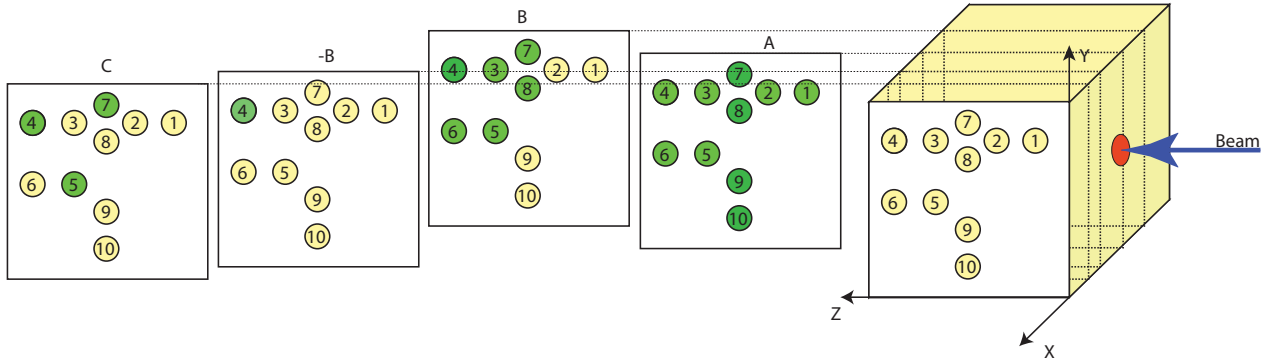


Figure 59: ^{Nat}In sample positions inside of the Activator (p_{beam} , Runs: 4, 5, 13, 14 & 20)

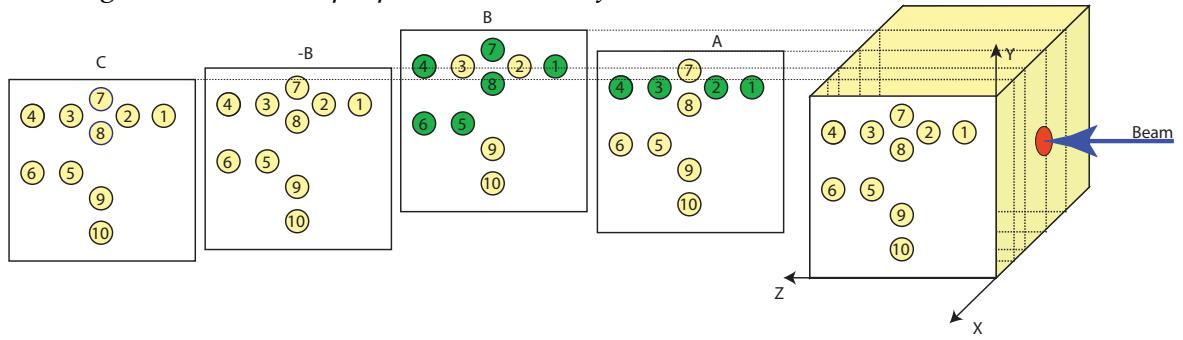


Figure 60: ^{Nat}In sample positions inside of the Activator (d_{beam} , Run12 & Run19)

Table 14: ^{Nat}In sample positions inside of the Activator (60 MeV p)

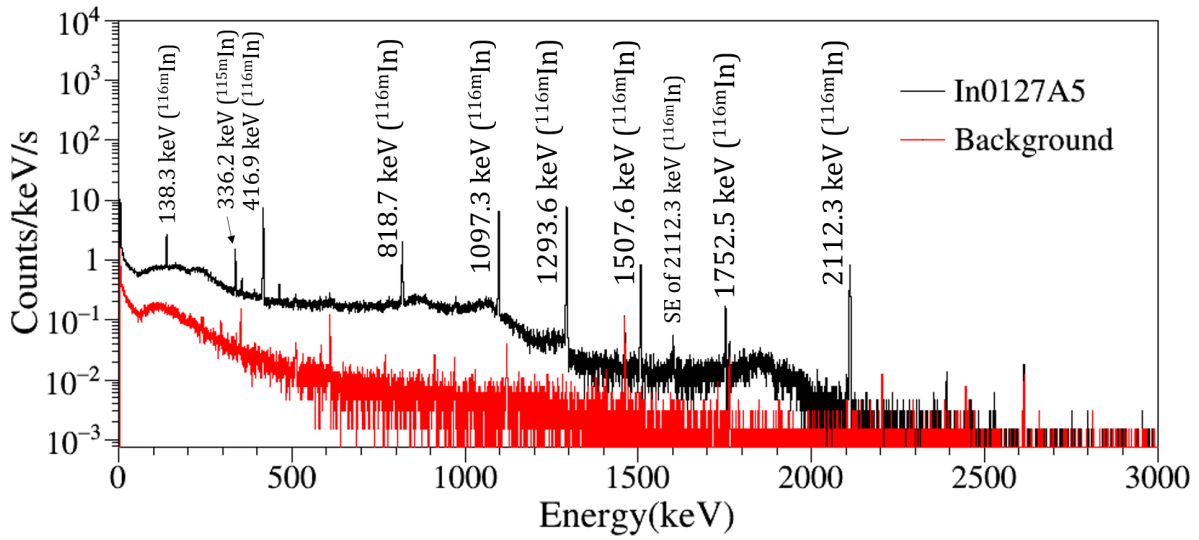
Run	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
4	0.127	A5	16.9	0.03474
4		A6	36.9	0.03437
4		B7	28.2	0.0279
4		B8	22.1	0.02838
5	0.127	A1	28.1	0.03474
5		B4	42.9	0.0279
5		C4	44.9	0.02838
5		C7	31.1	0.03437
5		A2	16.3	0.0291
5		B5	22.6	0.03296
5		B6	39.8	0.02646
5		C5	26.2	0.03557
14	0.127	A3	23.3	0.03474
14		A4	40.2	0.0279
14		A7	23.8	0.03296
14		B3	27.7	0.0291
20	0.127	A7	23.8	0.02646
20		A8	16.3	0.02838
20		A9	16.3	0.03557
20		A10	23.8	0.03437
20		B4	42.9	0.0324

Table 15: ^{Nat}In sample positions inside of the Activator (30 MeV d)

Run	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
12	0.127	B1	31.9	0.02894
12	0.127	B4	42.9	0.0324
12	0.127	B5	22.6	0.3294
12	0.127	B6	39.8	0.03044
12	0.127	B7	28.2	0.03137
12	0.127	B8	22.1	0.02871
19	0.127	A1	28.1	0.03474
19	0.127	A2	16.3	0.0291
19	0.127	A3	23.3	0.0279
19	0.127	A4	40.2	0.03296

These activation foils were also used to study the neutron leakage (Run20) by checking the symmetry between top part (above the target) and the bottom (below the target) of the Activator.

$^{115\text{m}}\text{In}$ ($T_{1/2} = 4.486$ h) and $^{116\text{m}}\text{In}$ ($T_{1/2} = 54.29$ m) were identified in the γ -ray spectrum (Figure 61 & Figure 62). $^{116\text{m}}\text{In}$ comes from $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$ reaction. In the case of $^{116\text{m}}\text{In}$, 3 main peaks were used for the activity calculation ($E_\gamma = 416.9$ KeV, $I_\gamma = 27.2$ KeV; $E_\gamma = 1097.3$ KeV, $I_\gamma = 58.5\%$; $E_\gamma = 1293.56$ KeV, $I_\gamma = 84.8\%$). In the $^{115\text{m}}\text{In}$ case, one γ -ray peak ($E_\gamma = 336.2$ KeV, $I_\gamma = 45.8\%$) was used for calculation. The spectrum in the case of protons is above the background, while for deuterons are at the same level.

Figure 61: γ -ray spectrum from the irradiated ^{Nat}In foil (run4, p_{beam} , $T_{\text{cool}} = 5.5\text{h}$)

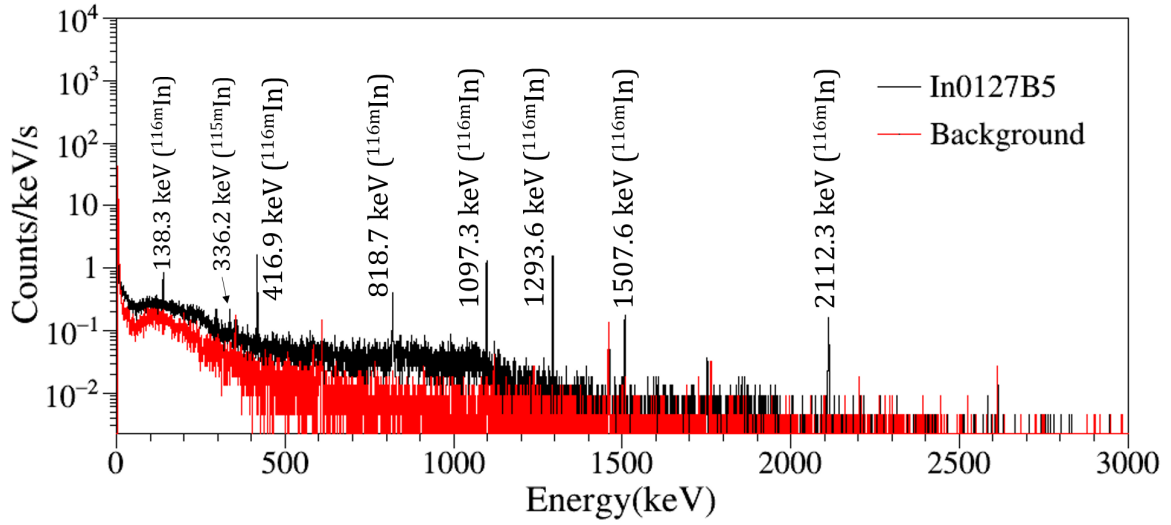


Figure 62 γ -ray spectrum from the irradiated ^{nat}In foil (run12, d_{beam} , $T_{\text{cool}} = 6.2\text{h}$)

Table 16 & Table 17 show the measured activity for $^{116\text{m}}\text{In}$ & $^{115\text{m}}\text{In}$ correspondingly from the irradiated samples with a 60 MeV proton beam.

The measured activity in function of the distance from the center of the Beryllium target is shown in Figure 63 & Figure 65 where we can remark that for $^{115}\text{In}(n, n')^{115\text{m}}\text{In}$, its maximum cross-section located at few MeV, which means that it has a similar behavior with ^{24}Na activity produced by $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ reaction. Furthermore, fast neutrons escape from the activator and the activity decreases exponentially when the distance from the Beryllium target increases. In the case of $^{115}\text{In}(n, \gamma)^{116\text{m}}\text{In}$, the cross-section has a maximum resonance value located at 1.46 eV, so the neutron flux in the activator in the eV region is almost the same along the distance from the Be target. We know that the activity A is proportional to the $\Phi \times \sigma$ and the $\Phi \approx 1/r^2$. So A should reduce as the distance from the target increases. However this is not the case, which means that s , is changing (increasing) as we go away from the target (more epithermal neutrons close to reflector), where the neutron flux at epithermal energy is increasing.

Is very interesting to notice the activities in positions A5 at 16.9 cm, A2 & A8 at 16.3 cm have totally different values (Table 17), that could explained because these positions have different angles with respect to the center of the Be target.

Table 16: Experimental and MC Simulation Activities for ^{116m}In

Run #	Thickness (cm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
5	0.0127	A1	28.1	3.01E+07	5.65	1.98E+07	1.94	1.57E+07	8.58
20	0.0127	A10	23.8	1.82E+07	5.25	1.29E+07	3.53	1.28E+07	8.16
5	0.0127	A2	16.3	2.83E+07	5.7	1.87E+07	3.5	1.48E+07	7.92
14	0.0127	A3	23.3	2.78E+07	4.62	1.79E+07	3.31	1.41E+07	7.85
14	0.0127	A4	40.2	2.81E+07	4.63	1.72E+07	1.58	1.38E+07	8.45
4	0.0127	A5	16.9	2.31E+07	4.76	1.38E+07	1.33	1.19E+07	7.68
4	0.0127	A6	36.9	2.51E+07	4.77	1.53E+07	0.97	1.28E+07	8.53
14	0.0127	A7	23.8	3.49E+07	4.63	2.33E+07	2.19	1.81E+07	8.12
20	0.0127	A7	23.8	3.45E+07	5.23	2.33E+07	2.19	1.81E+07	8.12
20	0.0127	A8	16.3	2.80E+07	5.23	1.85E+07	3.1	1.41E+07	7.7
20	0.0127	A9	16.3	1.91E+07	5.24	1.35E+07	2.9	1.11E+07	7.92
14	0.0127	B3	27.7	3.13E+07	4.63	2.01E+07	1.98	1.60E+07	7.85
5	0.0127	B4	42.9	2.83E+07	5.64	1.82E+07	1.52	1.42E+07	8.3
20	0.0127	B4	42.9	2.91E+07	5.22	1.82E+07	1.52	1.42E+07	8.3
13	0.0127	-B4	42.9	2.75E+07	4.6	1.82E+07	1.52	1.42E+07	8.3
5	0.0127	B5	22.6	2.53E+07	5.69	1.63E+07	2.68	1.40E+07	7.91
5	0.0127	B6	39.8	2.54E+07	5.7	1.70E+07	1.91	1.33E+07	8.28
4	0.0127	B7	28.2	3.72E+07	4.76	2.41E+07	1.54	1.87E+07	7.96
4	0.0127	B8	22.1	3.20E+07	4.78	2.04E+07	2.18	1.56E+07	7.9
5	0.0127	C4	44.9	2.76E+07	5.65	1.85E+07	2.63	1.49E+07	8.01
5	0.0127	C5	26.2	2.62E+07	5.69	1.78E+07	1.57	1.42E+07	7.77
5	0.0127	C7	31.1	3.31E+07	5.65	2.48E+07	1.64	1.87E+07	8.59

Table 17: MC and measured Activity for ^{115m}In

Run #	Thickness (cm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
4	0.0127	A5	16.9	7.15E+04	8.5	1.18E+05	1.34	1.55E+05	14.8
4	0.0127	A6	36.9	8.97E+03	11.3	1.18E+04	1.09	1.64E+04	20.9
4	0.0127	B7	28.2	6.63E+03	14.9	1.42E+04	5.76	1.86E+04	16.9
4	0.0127	B8	22.1	1.95E+04	10.08	3.37E+04	2.28	4.16E+04	13.5
5	0.0127	A2	16.3	3.23E+04	10.53	5.49E+04	3.03	5.98E+04	21.1
5	0.0127	B4	42.9	3.11E+03	20.76	5.77E+03	7.18	7.25E+03	21.6
5	0.0127	B5	22.6	2.68E+04	11.81	4.36E+04	3.17	5.95E+04	15.4
5	0.0127	C5	26.2	1.10E+04	15.33	2.44E+04	0.86	3.27E+04	12.7
20	0.0127	A8	16.3	4.85E+04	12.69	7.74E+04	1	9.64E+04	16.5
20	0.0127	A9	16.3	4.24E+04	11.63	7.72E+04	1.14	9.79E+04	17.6
20	0.0127	A10	23.8	1.52E+04	19.35	2.50E+04	1.5	3.42E+04	23.9

The measured activity for ^{116m}In & ^{115m}In correspondingly from the irradiated samples with a 30 MeV deuteron beam are shown in Table 18 & Table 19.

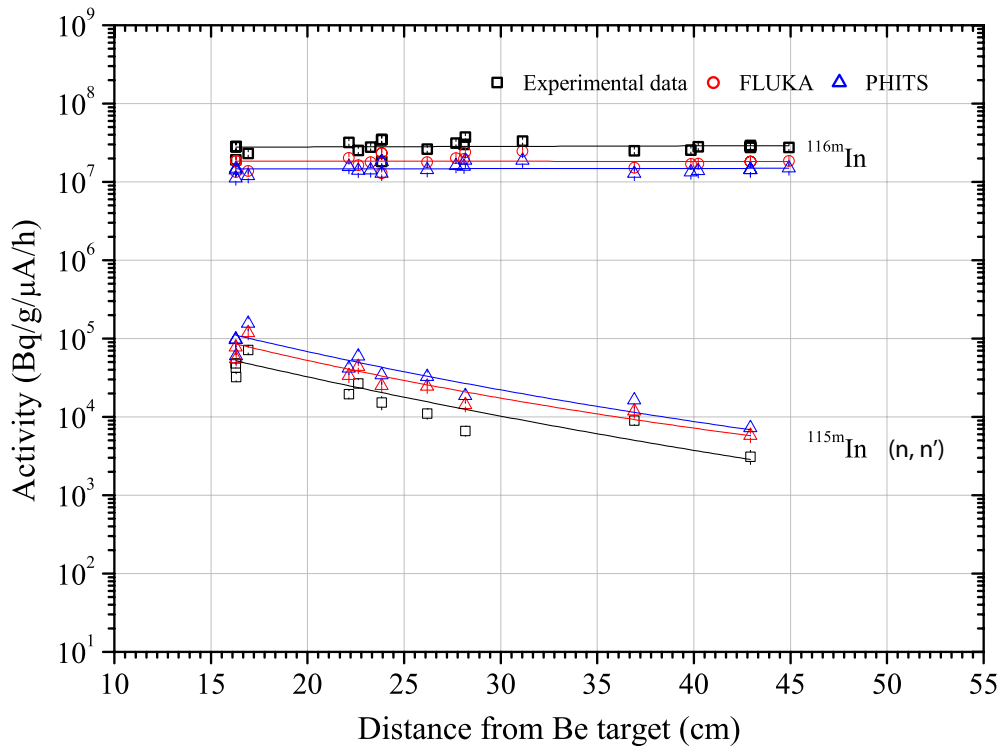
Table 18: Experimental and MC Simulation Activities for ^{116m}In

Run #	Thickness (cm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
12	0.0127	B1	31.9	1.11E+07	14.48	3.82E+08	0.26	7.31E+06	8.81
12	0.0127	B4	42.9	8.57E+06	14.51	4.35E+08	0.24	5.76E+06	9.37
12	0.0127	B5	22.6	8.07E+05	14.52	3.43E+08	0.17	5.84E+06	8.25
12	0.0127	B6	39.8	8.01E+06	14.54	3.63E+08	0.13	5.25E+06	9
12	0.0127	B7	28.2	1.13E+07	14.52	4.69E+08	0.15	8.11E+06	8.76
12	0.0127	B8	22.1	9.65E+06	14.56	4.40E+08	0.17	6.40E+06	7.97
19	0.0127	A1	28.1	1.44E+07	10.46	3.83E+08	0.15	7.41E+06	8.94
19	0.0127	A2	16.3	1.32E+07	10.47	3.68E+08	0.19	6.58E+06	8.27
19	0.0127	A3	23.3	1.21E+07	10.47	3.71E+08	0.15	6.55E+06	8.46
19	0.0127	A4	40.2	1.16E+07	7.9	3.94E+08	0.33	5.58E+06	8.64

Table 19: Experimental and MC Activity for ^{115m}In

Run #	Thickness (cm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
19	0.0127	A1	28.1	4.32E+03	23.06	1.52E+05	1.48	5.94E+03	28
19	0.0127	A2	16.3	1.34E+04	10.18	9.12E+05	0.72	2.87E+04	23
19	0.0127	A3	23.3	7.19E+03	12.32	1.04E+06	0.55	1.89E+04	26.5

The activity calculation of the irradiated $^{\text{Nat}}\text{In}$ is shown in Figure 63 & Figure 65.

Figure 63: Measured activity for ^{115m}In & ^{116m}In (60 MeV p).

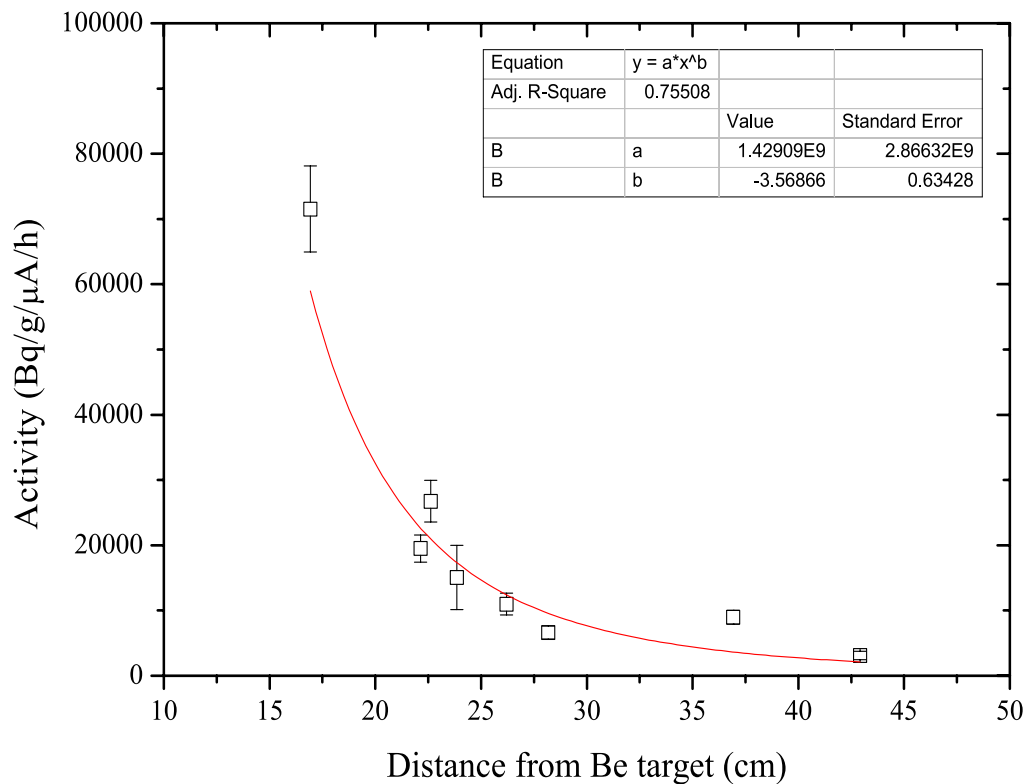


Figure 64: Measured activity (fitting curve) for ^{115m}In (60 MeV p).

Following the law $1/r^n$, one can observe that $n = 3.6$, which means that the fast neutrons disappear very quickly as we move away from the target.

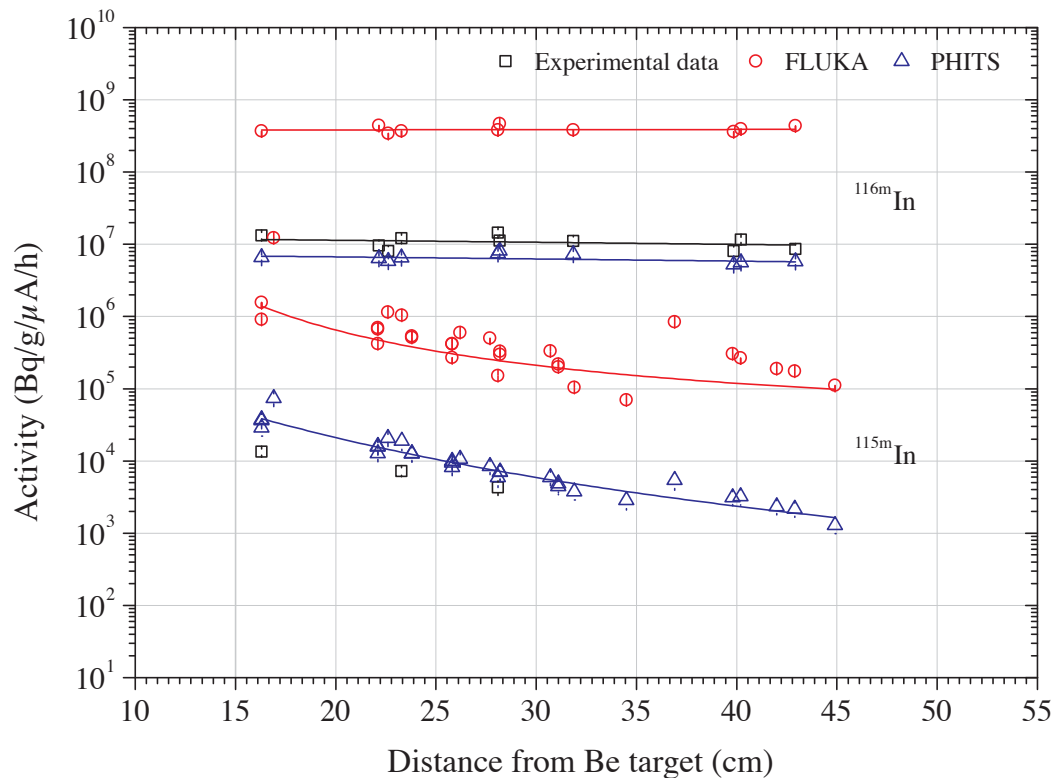


Figure 65: Measured activity for ^{115m}In & ^{116m}In (30 MeV d).

Figure 66 shows the experimental activities for proton and deuteron beams.

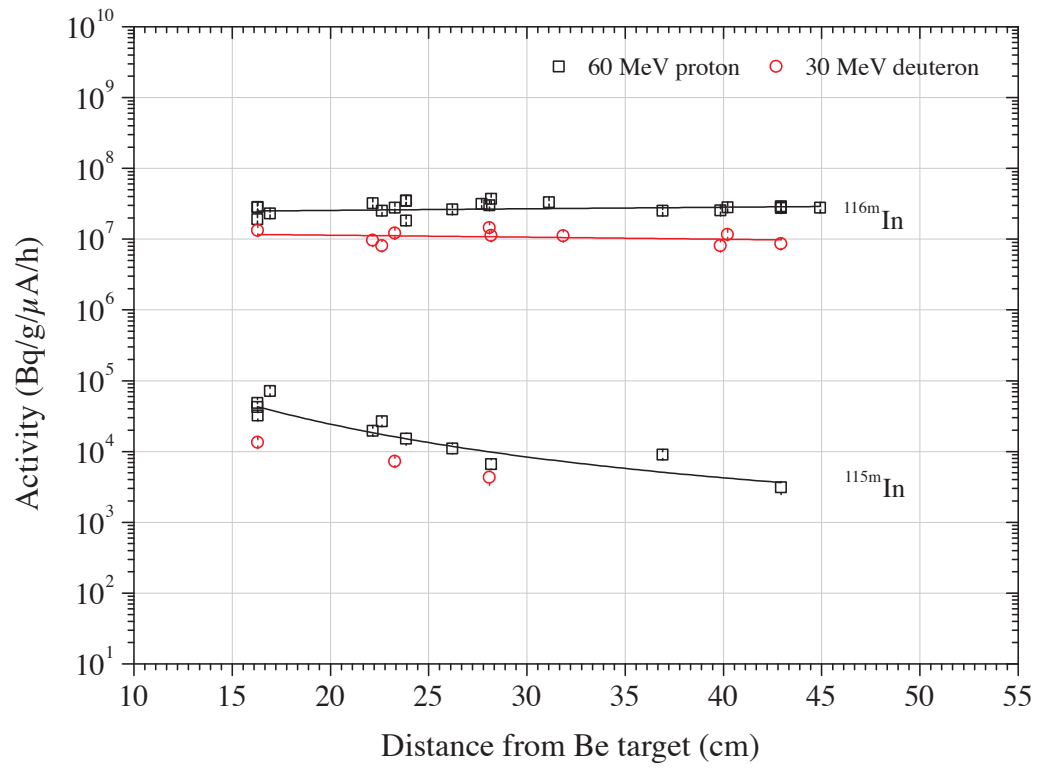


Figure 66: Measured activity for ^{115m}In & ^{116m}In with p & d.

5.6.1.2 Natural Au Activation Foil

Gold has only one stable isotope ^{197}Au . The capture reaction in gold produces the ^{198}Au ground state ($T_{1/2} = 2.6947$ d) with a probability of 99.95% while only 0.05% goes to the metastable state. $^{197}\text{Au}(n, \gamma)^{198}\text{Au}$ cross section is shown in Figure 67.

Natural Au activation foils were placed and irradiated in different positions inside of the Activator (Figure 68 & Figure 69 and Table 20 & Table 21).

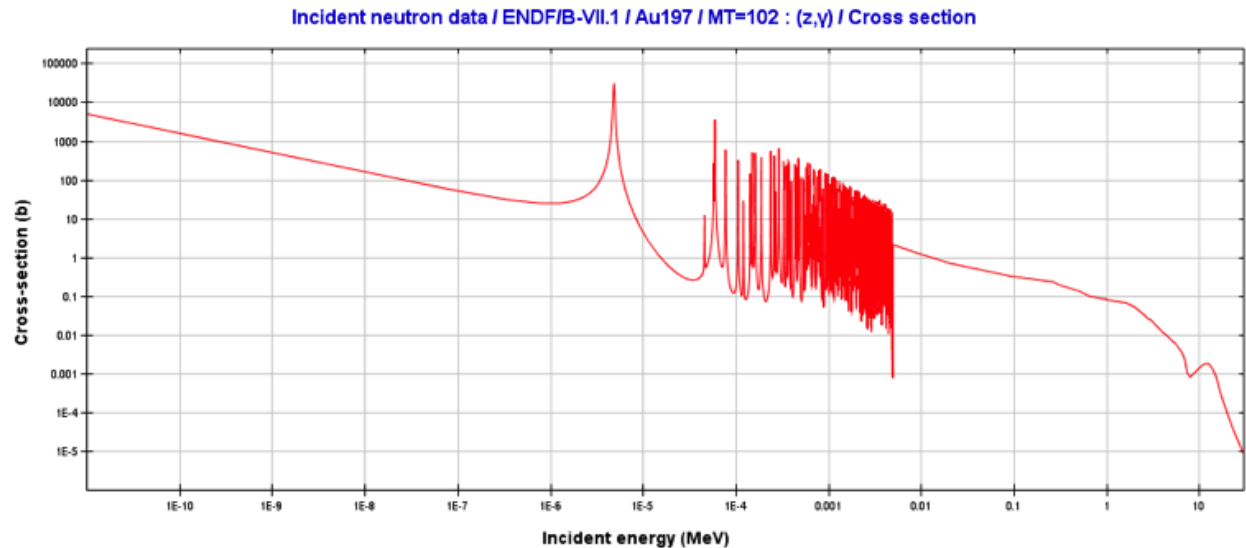


Figure 67: ^{197}Au (n, γ) Capture neutron Cross Section

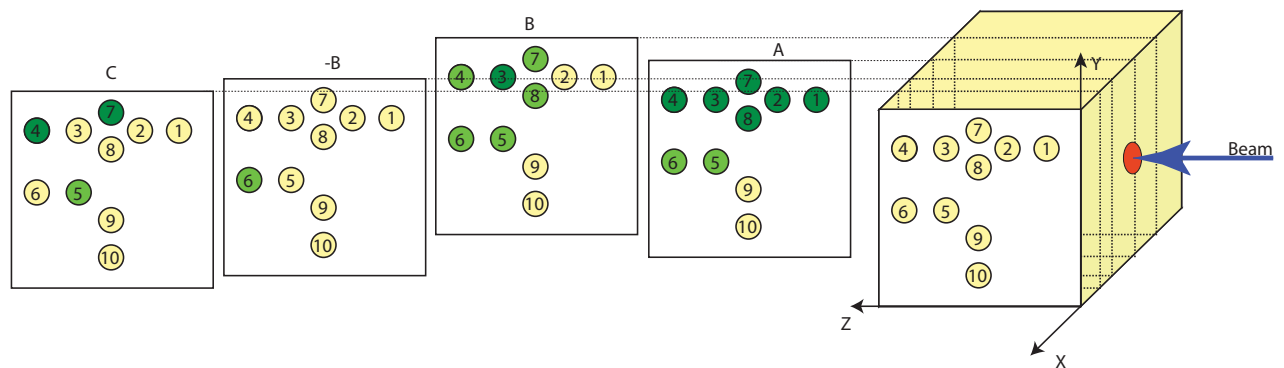


Figure 68: ^{197}Au sample positions inside of the Activator (p_{beam} , Run06 & Run15)

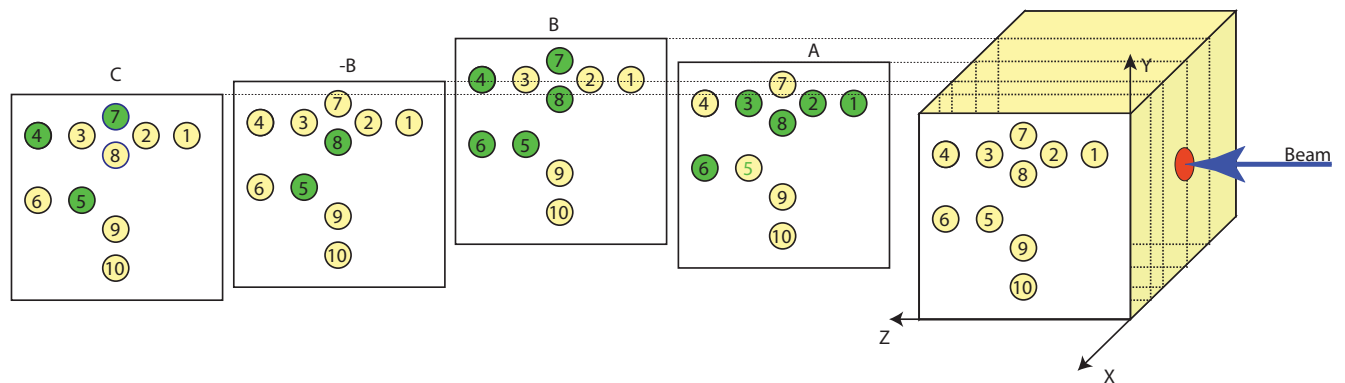


Figure 69: ^{197}Au sample positions inside of the Activator (d_{beam} , Run10, 12, 18 & 19)

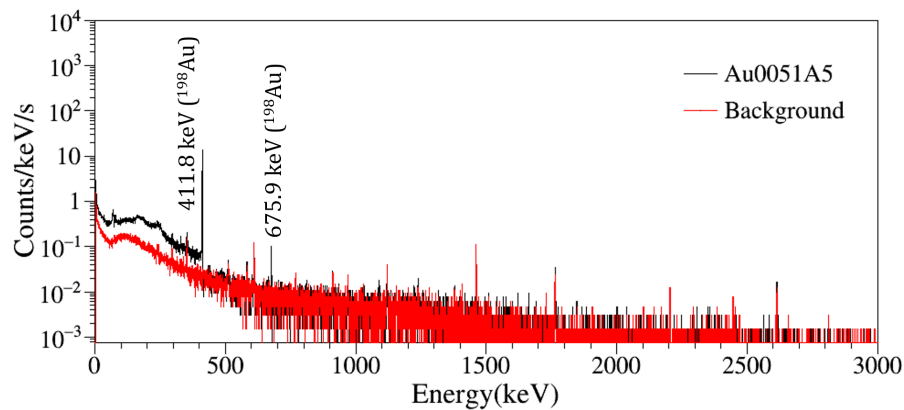
Table 20: ^{Nat}Au sample positions inside of the Activator (60 MeV p)

Run	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
6	0.051	A5	15.3	0.03609
6		A6	35.3	0.0302
6		B4	41.5	0.03175
6		B5	21.4	0.03048
6		B6	38.3	0.0273
6		B7	27.8	0.03616
6		B8	21.6	0.03098
6		C5	25.2	0.0253
15	0.051	A1	28.1	0.03048
15		A2	16.3	0.03098
15		A3	23.3	0.0253
15		A4	40.2	0.0302
15		A7	23.8	0.03364
15		A8	16.3	0.02844
15		B3	27.7	0.03175
15		C4	44.9	0.03335
15		C7	31.1	0.03002

Table 21: ^{Nat}Au sample positions inside of the Activator (30 MeV d)

Run	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
10	0.051	A1	28.1	0.03609
10	0.00508	A2	16.3	0.03048
10	0.00508	A3	23.3	0.03098
10	0.00508	A5	16.9	0.03364
10	0.00508	A6	36.9	0.03345
10	0.00508	A8	16.3	0.0302
10	0.00508	B4	42.9	0.03335
10	0.00508	B5	22.6	0.03002
10	0.00508	B6	39.8	0.03022
10	0.00508	B7	28.2	0.03361
10	0.00508	B8	22.1	0.02765
10	0.00508	C4	44.9	0.0253
10	0.00508	C5	26.2	0.02844
10	0.00508	C7	31.1	0.03175
12	0.00508	-B5	22.6	0.0273
12	0.00508	-B8	22.1	0.03616
18	0.00508	-B5	22.6	0.03002
19	0.00508	-B5	22.6	0.03048

Figure 70 shows the gamma ray spectrum of irradiated ^{Nat}Au foil at 60 MeV proton beam and Figure 71 shows gamma ray spectrum for irradiated ^{Na}Au foil at 30 MeV deuteron beam. ^{198}Au comes from $^{197}\text{Au}(n, \gamma)^{198}\text{Au}$ reaction, which emits strong gamma line at 411.804 KeV ($I_\gamma = 95.5\%$) and proceeds directly through β -decay to ^{198}Hg .

Figure 70: γ -ray spectrum from the irradiated Au foil (run6, p_{beam} , $T_{cool} = 7.2h$)

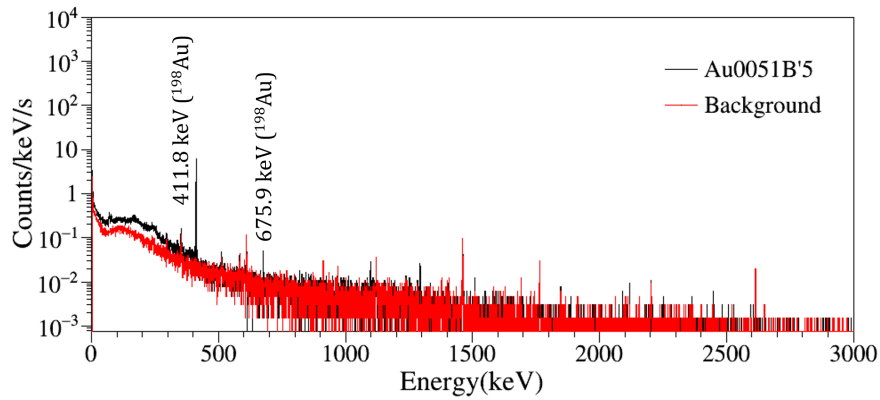


Figure 71: γ -ray spectrum from the irradiated ^{198}Au foil (run12, -B8 pos, d_{beam} , $T_{\text{cool}} = 2.6h$)

Table 22 shows the measured activity for ^{198}Au at different position inside of the Activator, which has a constant behavior along the distance from the center of the Be target (Figure 72 & Figure 73).

Table 22: Experimental and MC Simulation Activities for ^{198}Au .

	Run #	Thickness (cm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
60 MeV Proton Beam	15	0.0051	A1	28.1	2.86E+05	3.89	1.24E+05	2.68	8.86E+04	6.67
	15	0.0051	A2	16.3	2.73E+05	3.89	1.27E+05	1.27	9.17E+04	7.1
	15	0.0051	A3	23.3	2.49E+05	3.9	1.19E+05	2.51	8.70E+04	6.97
	15	0.0051	A4	40.2	2.53E+05	3.89	1.07E+05	6.03	8.34E+04	6.81
	6	0.0051	A5	16.9	2.16E+05	4.2	1.01E+05	2.88	7.51E+04	7.69
	6	0.0051	A6	36.9	2.26E+05	4.2	9.59E+04	5.37	8.03E+04	6.27
	15	0.0051	A7	23.8	3.10E+05	3.88	1.44E+05	2.28	1.17E+05	6.7
	15	0.0051	A8	16.3	2.57E+05	3.9	1.21E+05	3.11	8.60E+04	7.36
	15	0.0051	B3	27.7	2.74E+05	3.89	1.13E+05	3.39	8.55E+04	6.37
	6	0.0051	B4	42.9	2.55E+05	4.2	1.06E+05	4.93	8.60E+04	6.32
	6	0.0051	B5	22.6	2.49E+05	4.19	1.03E+05	2.7	8.19E+04	7.07
	6	0.0051	B6	39.8	2.39E+05	4.2	1.03E+05	2.71	7.72E+04	6.06
	13	0.0051	B'6	39.8	2.27E+05	3.75	1.03E+05	2.71	7.72E+04	6.06
	6	0.0051	B7	28.2	3.16E+05	4.2	1.49E+05	2.08	1.13E+05	6.69
	6	0.0051	B8	22.1	2.81E+05	4.2	1.31E+05	1.06	9.17E+04	7.09
	15	0.0051	C4	44.9	2.50E+05	3.89	1.07E+05	3.42	8.76E+04	5.86
	6	0.0051	C5	26.2	2.53E+05	4.2	1.22E+05	3.01	8.86E+04	6.61
	15	0.0051	C7	31.1	3.07E+05	3.89	1.43E+05	2.57	1.16E+05	6.35
30 MeV Deuteron Beam	10	0.0051	A1	28.1	1.21E+05	4.29	2.53E+06	0.15	4.13E+04	7.12
	10	0.0051	A2	16.3	1.15E+05	4.3	2.31E+06	0.17	4.23E+04	7.13
	10	0.0051	A3	23.3	9.70E+04	4.32	2.36E+06	0.24	3.84E+04	9.02
	10	0.0051	A5	16.9	8.50E+04	4.32	1.69E+06	0.31	3.18E+04	6.49
	10	0.0051	A6	36.9	8.66E+04	4.33	2.15E+06	0.23	3.16E+04	8.4
	10	0.0051	A8	16.3	1.03E+05	4.32	2.30E+06	0.18	3.64E+04	7.19
	10	0.0051	B4	42.9	9.75E+04	4.32	2.74E+06	0.25	3.11E+04	7.66
	10	0.0051	B5	22.6	9.80E+04	4.32	2.43E+06	0.27	3.51E+04	7.5
	10	0.0051	B6	39.8	8.94E+04	4.33	2.43E+06	0.18	2.94E+04	8.03
	10	0.0051	B7	28.2	1.28E+05	4.3	3.05E+06	0.19	4.51E+04	4.96
	10	0.0051	B8	22.1	1.15E+05	4.32	2.69E+06	0.17	4.40E+04	5.11
	10	0.0051	C4	44.9	9.25E+04	4.35	2.51E+06	0.25	3.35E+04	6.74
	10	0.0051	C5	26.2	1.00E+05	4.33	2.76E+06	0.15	3.45E+04	7.23
	10	0.0051	C7	31.1	1.23E+05	4.3	2.95E+06	0.27	4.57E+04	6.54
	12	0.0051	-B5	22.6	7.84E+04	14.23	2.43E+06	0.27	3.51E+04	7.5
	12	0.0051	-B8	22.1	8.83E+04	14.23	2.69E+06	0.17	4.40E+04	5.11
	18	0.0051	-B5	22.6	1.01E+05	6.15	2.43E+06	0.27	3.51E+04	7.5
	19	0.0051	-B5	22.6	1.05E+05	7.46	2.43E+06	0.27	3.51E+04	7.5

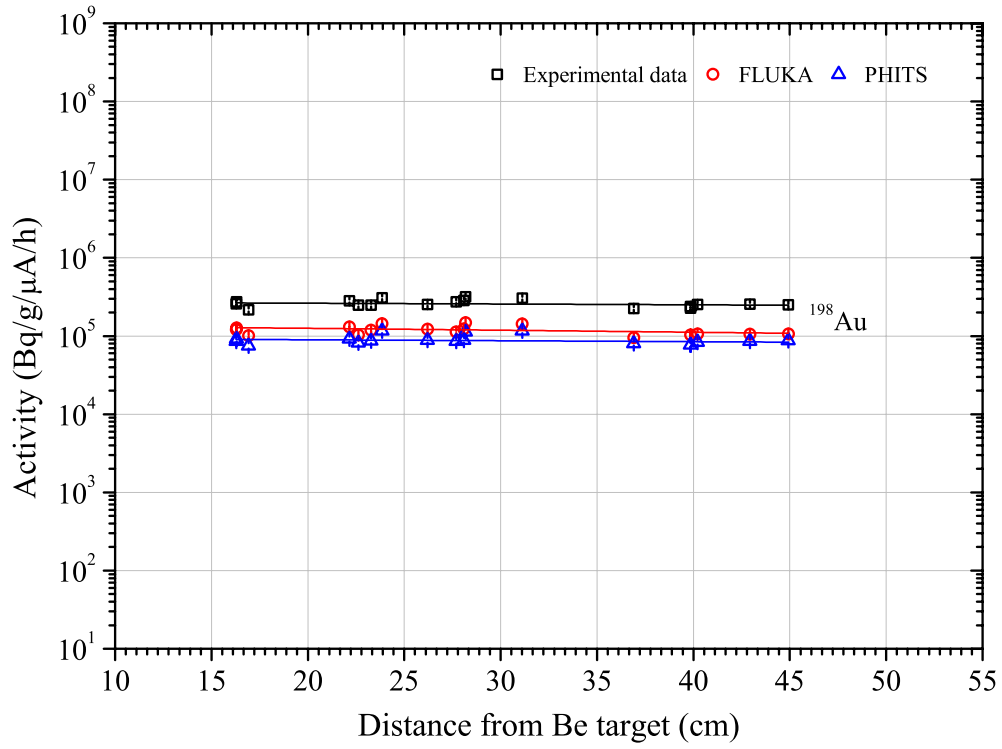


Figure 72: Measured activity for ^{198}Au (60 MeV p).

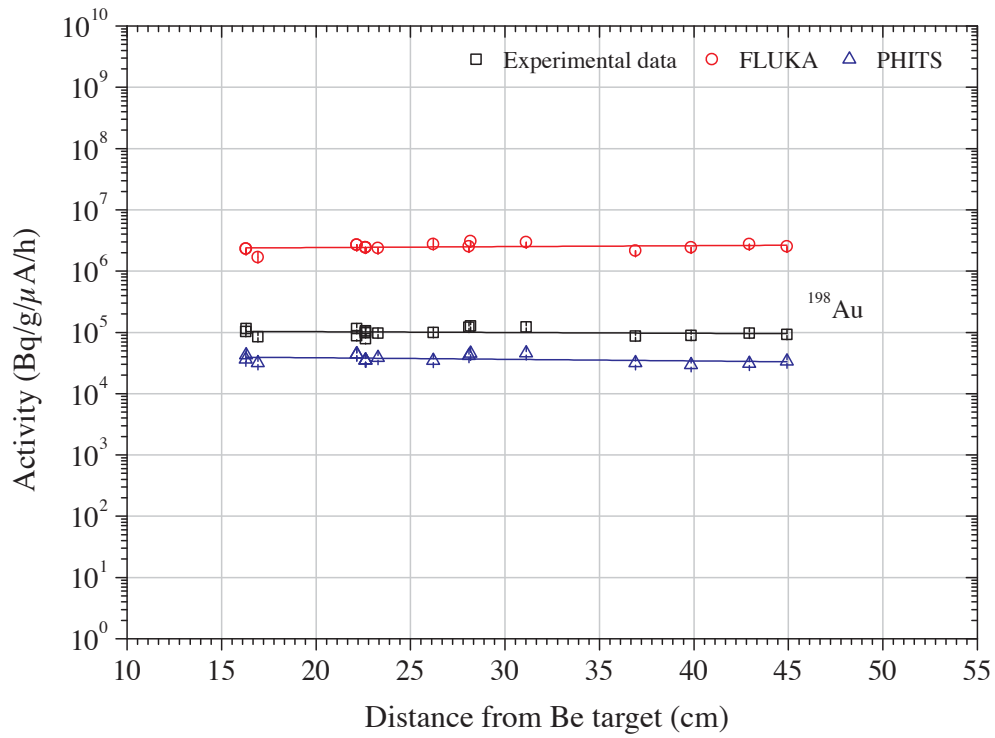


Figure 73: Measured activity for ^{198}Au (30 MeV d).

It seems the position of the Au activation foil sample in the Activator is irrelevant and possibly neutrons are well reflected from the graphite layers in order to maintain (confine) the neutron flux in the eV region to allow the resonance capture reactions.

However the observed results are not at all as we expected. If we are using activation foils, which are standards materials with a very known cross section, the difference of the MC simulation calculation with the experimental data must come from other variables in the equation of the activity ($A \approx N \times \sigma \times \Phi$). As we can notice for a specific sample with a very well known N & σ , the only variable to analyze is the Flux.

Figure 74 shows the experimental activities with a proton and deuteron beams.

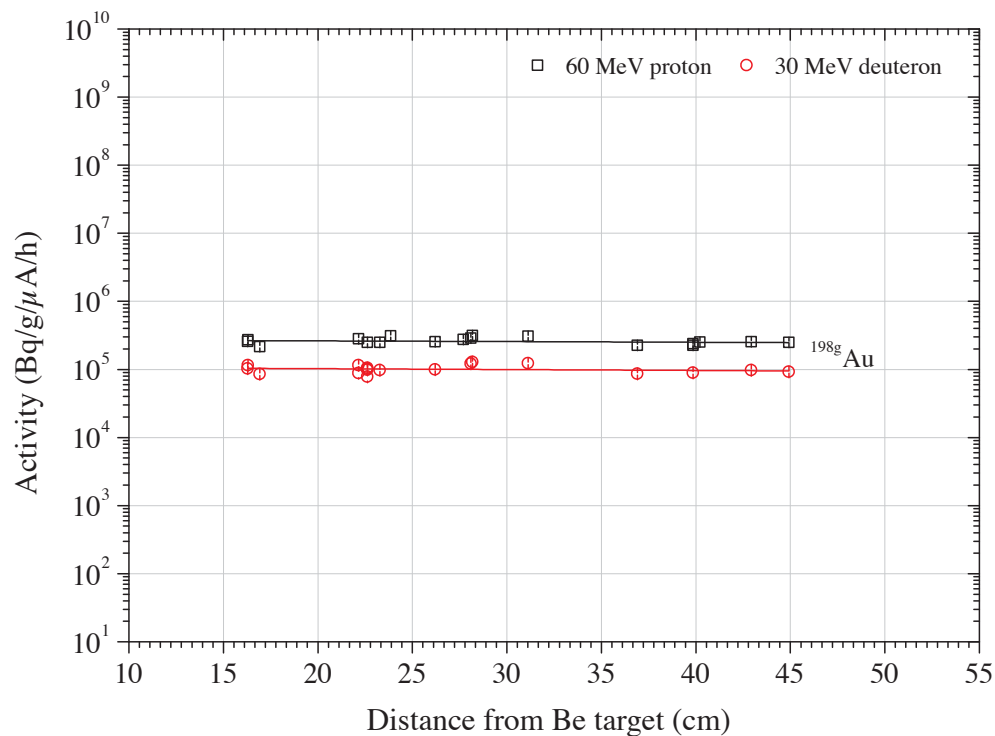


Figure 74: Measured activity for ^{198}Au wit p & d .

5.6.1.3 Natural W Activation Foil

Tungsten has five stable isotopes ^{180}W (0.12%), ^{182}W (26.5%), ^{183}W (14.3%), ^{184}W (30.6%), and ^{186}W (28.4%). We are interested in the (n, γ) reaction on the ^{186}W isotope. $^{186}\text{W}(n, \gamma)$ cross section is shown in Figure 75, where we can remark that $^{186}\text{W}(n, \gamma)$ has very strong resonance peak at 18.9 eV.

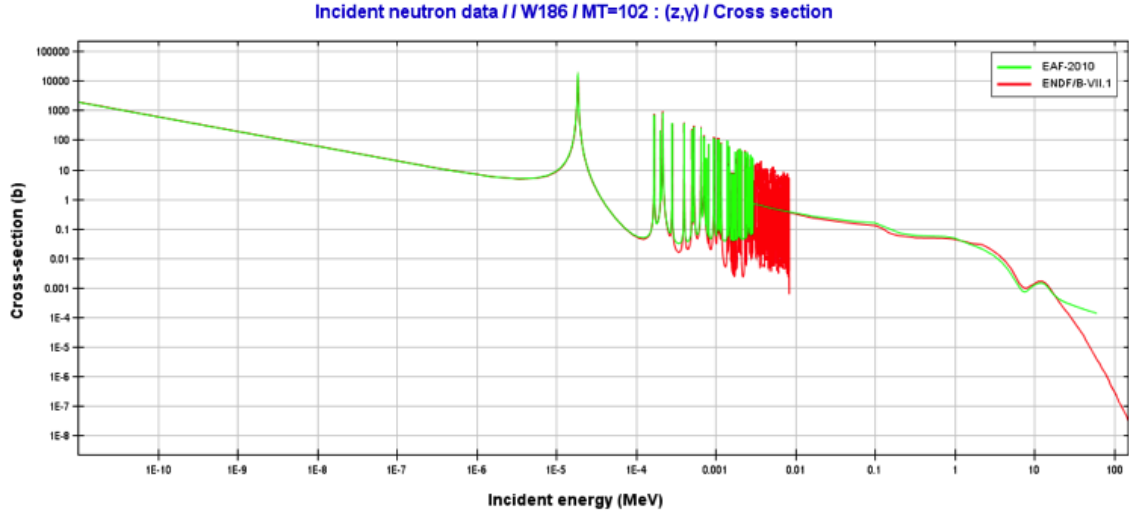


Figure 75: $^{186}\text{W}(n, \gamma)$ Capture neutron Cross Section.

^{186}W activation foils were placed and irradiated with a proton (Figure 76) and deuteron (Figure 77) beams at different positions inside of the Activator.

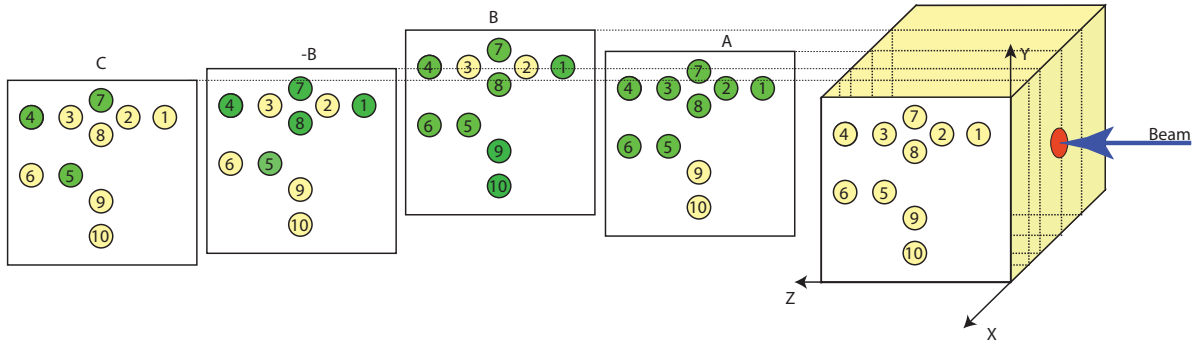


Figure 76: ^{186}W sample positions inside of the Activator (p_{beam} , Runs: 07, 08, 11, 13 & 20).

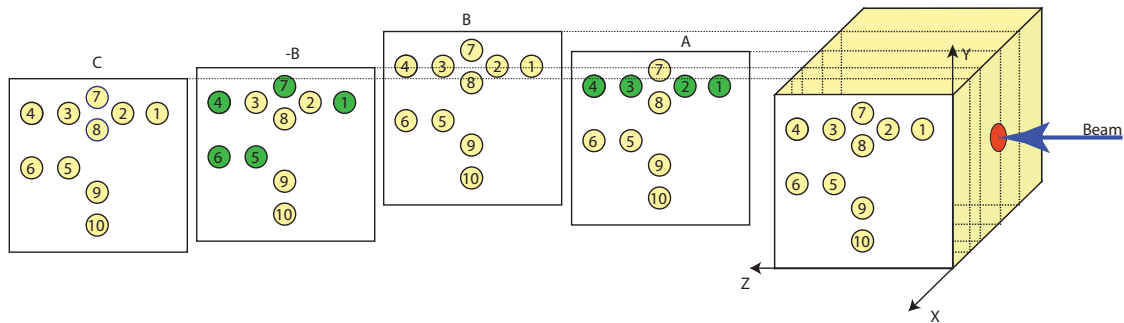


Figure 77: ^{186}W sample positions inside of the Activator (d_{beam} , Run12 & Run19).

^{Nat}W activation foils used in the experiment are shown in Table 23. These activation foils were also used to study the neutron leakage (Run20) by checking the symmetry between top part (above the target) and the bottom (below the target) of the Activator.

Table 23: ^{Nat}W sample positions inside of the Activator for all runs

	Run	Thickness (mm)	Position	d (cm)	Mass (g)		Run	Thickness (mm)	Position	d (cm)	Mass (g)
60 MeV Proton Beam	7	0.25	A1	28.1	0.359	30 MeV Deuteron Beam	12	0.25	A1	28.1	0.317
	7	0.25	A2	16.3	0.369		12	0.25	A2	16.3	0.36
	7	0.25	A3	23.3	0.355		12	0.25	A3	23.3	0.348
	7	0.25	A4	40.2	0.381		12	0.25	A4	40.2	0.35
	7	0.25	A5	16.9	0.378		12	0.25	-B1	31.9	0.382
	7	0.25	A6	36.9	0.372		12	0.25	-B4	42.9	0.34
	7	0.25	A7	23.8	0.381		12	0.25	-B6	39.8	0.332
	7	0.25	A8	16.3	0.382		12	0.25	-B7	28.2	0.408
	7	0.25	B4	42.9	0.38		19	0.25	B5	22.6	0.414
	7	0.25	B5	22.6	0.402						
	7	0.25	B6	39.8	0.361						
	7	0.25	B7	28.2	0.369						
	7	0.25	B8	22.1	0.406						
	7	0.25	C4	44.9	0.372						
	7	0.25	C5	26.2	0.371						
	7	0.25	C7	31.1	0.406						
	8	1.75	-B4	42.9	2.599						
	8	1.75	-B7	28.2	2.644						
	8	1.75	-B8	22.1	2.648						
	8	1	B4	42.9	1.359						
	8	1	B7	28.2	1.364						
	8	1	B8	22.1	1.355						
	8	1	A5	16.9	1.509						
	8	1	A6	36.9	1.418						
	8	1	B1	31.9	1.282						
	8	0.5	A1	28.1	0.705						
	8	0.25	-B1	31.9	0.376						
	11	1	A1	28.1	1.359						
	11	0.75	-B4	42.9	1.083						
	11	0.75	-B7	28.2	1.133						
	11	0.75	-B8	22.1	1.151						
	11	0.5	A5	16.9	0.733						
	11	0.5	A6	36.9	0.75						
	11	0.5	B1	31.9	0.704						
	11	0.5	-B1	31.9	0.668						
	11	0.5	B4	42.9	0.788						
	11	0.5	B7	28.2	0.82						
	11	0.5	B8	22.1	0.727						
	13	0.25	B'5	22.6	0.359						
	20	0.25	B7	28.2	0.341						
	20		B8	22.1	0.341						
	20		B9	22.1	0.325						
	20		B10	28.2	0.33						

By capturing one neutron ^{186}W becomes ^{187}W , which emits several gamma rays, with a half-life of 24 hours (see Figure 78 & Figure 79). Two main peaks were used for the activity calculation ($E_{\gamma} = 479.5$ KeV, $I_{\gamma} = 26.6$ %; $E_{\gamma} = 685.8$ KeV, $I_{\gamma} = 33.2$ %).

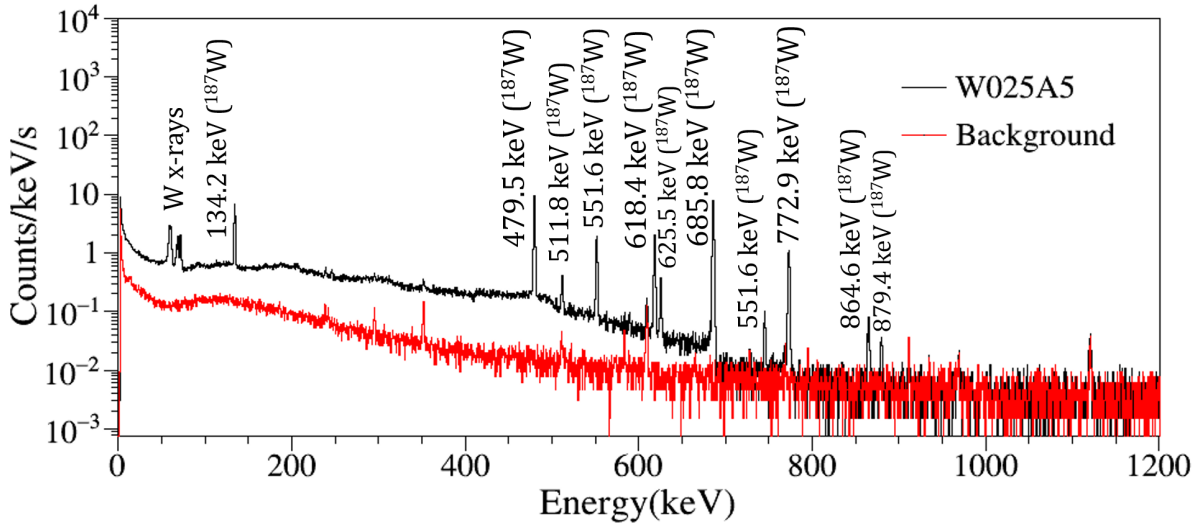


Figure 78: γ -ray spectrum from the irradiated W foil (run7, p_{beam} , $T_{\text{cool}} = 64.9\text{h}$).

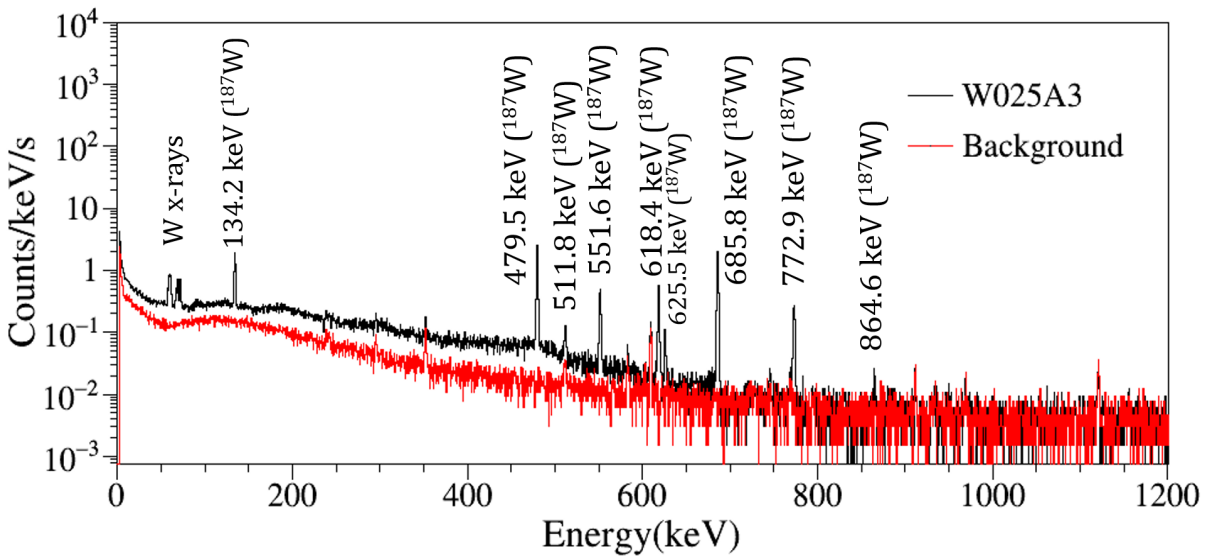


Figure 79: γ -ray spectrum from the irradiated ^{187}W foil (run12, d_{beam} , $T_{\text{cool}} = 27.6\text{h}$).

Table 24 & Table 25 show the activities for ^{187}W from the irradiated samples at different position inside of the Activator.

Currently we are working to obtain the MC simulation (FLUKA and PHITS) results for W Activation foils as shown in Table 26. These results are delayed because of the complex runs performed with different thicknesses of the activation foils.

Experimental results from the ^{187}W (Figure 80 & Figure 81) show a constant behavior of the measured activity along the distance from the center of the Be target. Neutrons are reflected from the graphite layer box keeping the neutron flux constants in the eV region, where neutron capture reactions are performed following the ARC method.

Table 24: Experimental and MC Simulation Activities for ^{187}W (60 MeV p)

Run #	Thickness (cm)	Position	d (cm)	Exp Activity (Bq/g/h/ μ A)	Err (%)	FLUKA Activity (Bq/g/h/ μ A)	Err (%)	PHITS Activity (Bq/g/h/ μ A)	Err (%)
7	0.025	A1	28.1	7.45E+04	4.56	8.05E+04	2.08	3.82E+04	7.14
7	0.025	A2	16.3	7.32E+04	4.56	7.69E+04	4.07	3.66E+04	8.64
7	0.025	A3	23.3	6.66E+04	4.56	7.64E+04	3.96	3.62E+04	7.99
7	0.025	A4	40.2	6.62E+04	4.56	7.12E+04	2.7	3.49E+04	7.28
7	0.025	A5	16.9	5.79E+04	4.56	6.39E+04	4.17	3.01E+04	9.28
7	0.025	A6	36.9	5.92E+04	4.56	7.01E+04	3.18	3.05E+04	7.85
7	0.025	A7	23.8	8.04E+04	4.56	8.73E+04	2.95	4.75E+04	8.38
7	0.025	A8	16.3	6.76E+04	4.56	7.69E+04	3.79	3.69E+04	8.97
7	0.025	B4	42.9	6.78E+04	4.56	7.22E+04	4.46	3.74E+04	6.66
7	0.025	B5	22.6	6.60E+04	4.56	7.06E+04	3.12	3.62E+04	8.39
7	0.025	B6	39.8	6.29E+04	4.56	6.86E+04	3.99	3.45E+04	8.05
7	0.025	B7	28.2	8.27E+04	4.56	9.14E+04	5.03	4.70E+04	7.08
7	0.025	B8	22.1	7.52E+04	4.56	8.47E+04	4.26	3.88E+04	7.94
7	0.025	C4	44.9	6.55E+04	4.56	6.70E+04	2.77	3.74E+04	6.71
7	0.025	C5	26.2	6.64E+04	4.56	7.84E+04	3.37	3.71E+04	7.94
7	0.025	C7	31.1	8.05E+04	4.56	9.30E+04	2.6	4.82E+04	7.25
13	0.025	B'5	22.6	6.62E+04	4.59	7.06E+04	3.12	3.62E+04	8.39
20	0.025	B7	28.2	8.92E+04	5.27	9.14E+04	5.03	4.70E+04	7.08
20	0.025	B8	22.1	7.88E+04	5.29	8.47E+04	4.26	3.88E+04	7.94
20	0.025	B9	22.1	6.43E+04	5.31	6.81E+04	2.77	3.24E+04	8.44
20	0.025	B10	28.2	5.77E+04	5.32	6.23E+04	5.01	3.48E+04	7.78
8	0.025	-B1	31.9	7.71E+04	4.21	7.79E+04	2.41	4.24E+04	7.9

Table 25: Experimental and MC Simulation Activities for ^{187}W (30 MeV d)

Run #	Thickness (cm)	Position	d (cm)	Exp Activity (Bq/g/h/ μ A)	Err (%)	FLUKA Activity (Bq/g/h/ μ A)	Err (%)	PHITS Activity (Bq/g/h/ μ A)	Err (%)
12	0.025	A1	28.1	2.90E+04	14.48	1.49E+06	0.16	1.80E+04	9.32
12	0.025	A2	16.3	2.68E+04	14.48	1.66E+06	0.25	1.77E+04	9.5
12	0.025	A3	23.3	2.27E+04	14.48	1.51E+06	0.22	1.61E+04	8.78
12	0.025	A4	40.2	2.19E+04	14.48	1.43E+06	0.33	1.54E+04	9.07
12	0.025	-B1	31.9	2.92E+04	14.48	1.33E+06	0.36	1.92E+04	7.27
12	0.025	-B4	42.9	2.19E+04	14.48	1.69E+06	0.27	1.43E+04	7.07
12	0.025	-B6	39.8	2.09E+04	14.49	1.36E+06	0.22	1.37E+04	10.9
12	0.025	-B7	28.2	2.81E+04	14.48	1.79E+06	0.26	2.03E+04	7.48
19	0.025	B5	22.6	3.08E+04	7.92	1.35E+06	0.22	1.46E+04	8.69

Table 26: Experimental and MC Simulation Activities for ^{187}W (60 MeV p)

Run #	Thickness (cm)	Position	d (cm)	Exp Activity (Bq/g/h/ μ A)	Err (%)	FLUKA Activity (Bq/g/h/ μ A)	Err (%)	PHITS Activity (Bq/g/h/ μ A)	Err (%)
8	0.05	A1	28.1	6.80E+04	4.21				
8	0.175	-B4	42.9	4.52E+04	4.22				
8	0.175	-B7	28.2	5.52E+04	4.21				
8	0.175	-B8	22.1	4.76E+04	4.22				
8	0.1	A5	16.9	4.42E+04	4.22				
8	0.1	A6	36.9	4.67E+04	4.21				
8	0.1	B1	31.9	6.17E+04	4.21				
8	0.1	B4	42.9	5.48E+04	4.21				
8	0.1	B7	28.2	6.75E+04	4.21				
8	0.1	B8	22.1	5.89E+04	4.21				
11	0.05	A5	16.9	5.13E+04	4.67				
11	0.05	A6	36.9	5.37E+04	4.67				
11	0.05	B1	31.9	7.05E+04	4.67				
11	0.05	-B1	31.9	6.96E+04	4.67				
11	0.05	B4	42.9	6.23E+04	4.67				
11	0.05	B7	28.2	7.71E+04	4.66				
11	0.05	B8	22.1	6.83E+04	4.67				
11	0.075	-B4	42.9	5.61E+04	4.66				
11	0.075	-B7	28.2	6.74E+04	4.66				
11	0.075	-B8	22.1	5.71E+04	4.66				
11	0.1	A1	28.1	5.91E+04	4.66				

A self-attenuation correction is necessary to perform for the experimental results of the W foils. We expect an increase of the activity of the order of 1-3% for W, 0.25 mm thickness. The self-attenuation for the thicker W is expected to be more than 3% for W activation foils with thicknesses bigger than 0.25 mm.

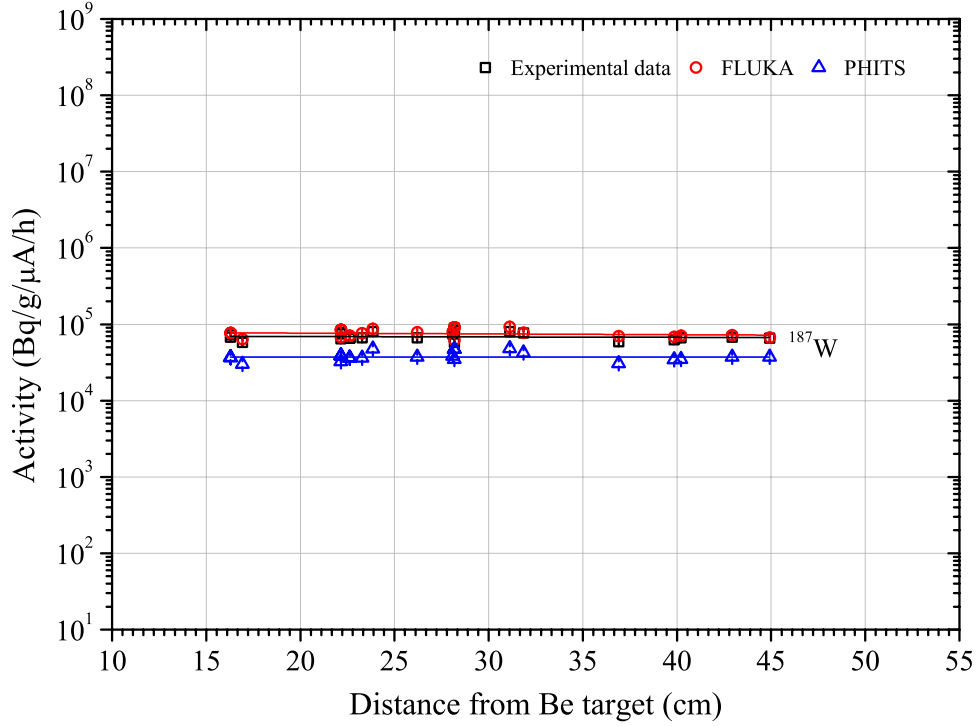


Figure 80: Measured activity for ^{187}W (60 MeV p).

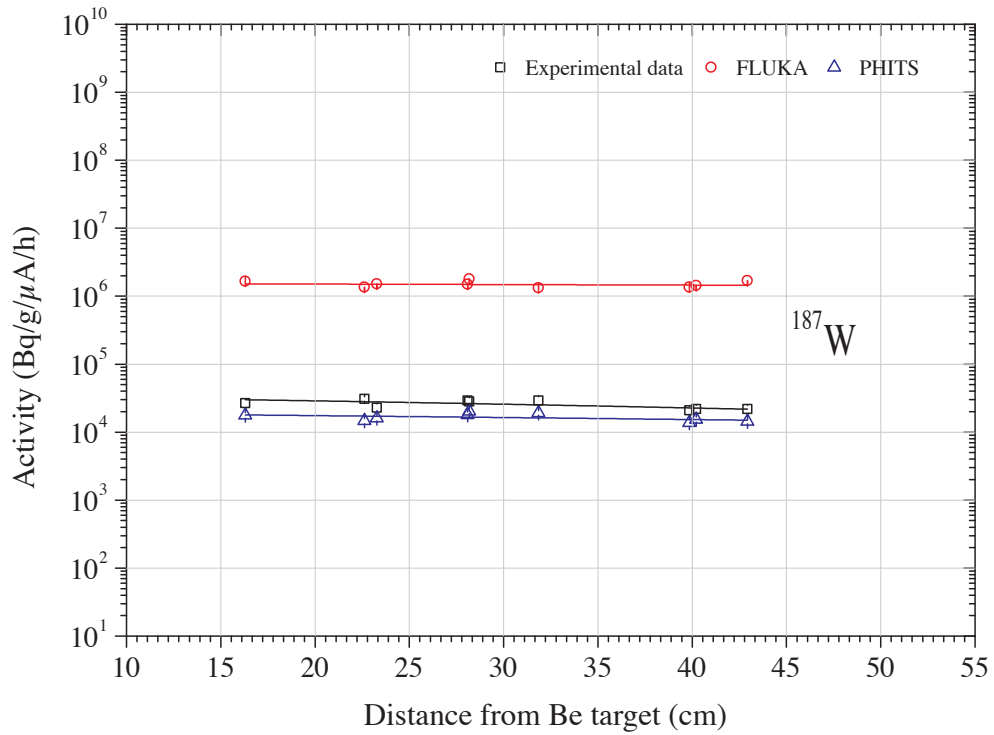


Figure 81: Measured activity for ^{187}W (30 MeV d).

Figure 82 shows the results for the measured activities for proton and deuteron beams.

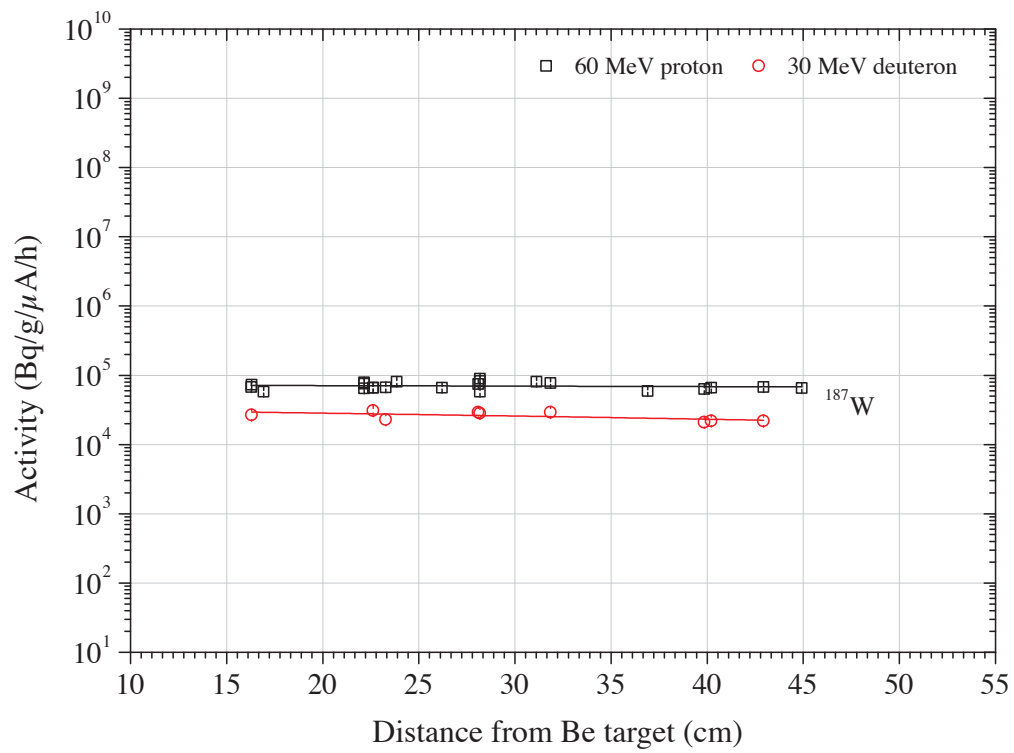


Figure 82: Measured activity for ^{187}W with p & d .

5.6.1.4 Natural Al Activation Foil

^{27}Al has only one stable isotope ^{27}Al . ^{24}Na ($T_{1/2} = 14.997$ h) comes from $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ reaction. The metastable state ^{24m}Na is also produced by (n, α) reaction. ^{24m}Na decays immediately due to very short half-life ($T_{1/2} = 20.18$ ms). $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ reaction cross section is shown in Figure 83.

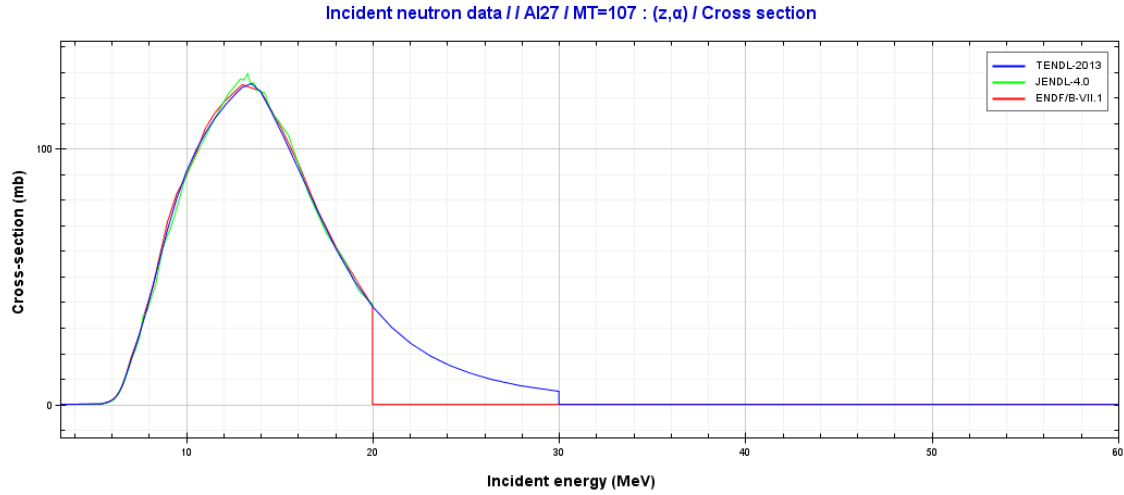


Figure 83: $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ neutron Cross Section.

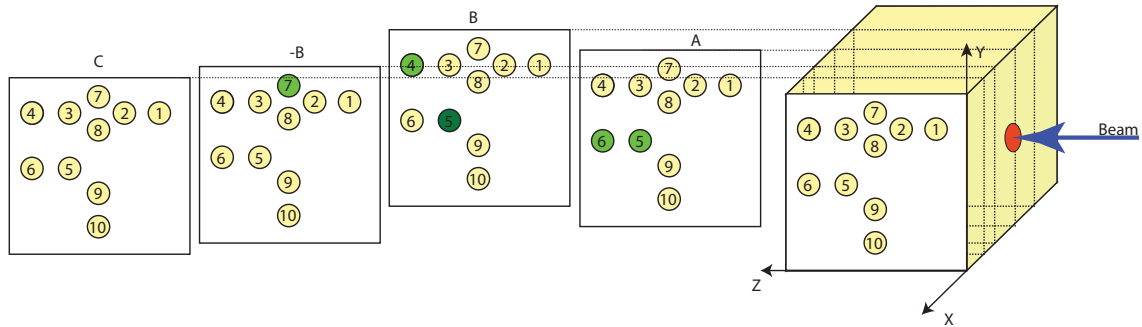


Figure 84: ^{27}Al sample positions inside of the Activator (p_{beam} , Runs 2, 3 & 14).

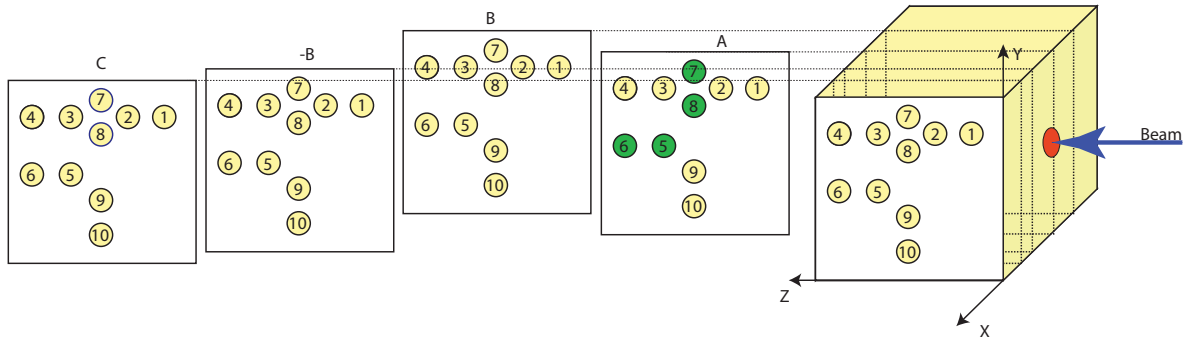


Figure 85: ^{27}Al sample positions inside of the Activator (d_{beam} , Run12).

Natural Al activation foils were placed and irradiated proton (Figure 84) and deuteron (Figure 85) beams in different positions inside of the Activator. Table 27 & Table 28 are the corresponding tables for these positions.

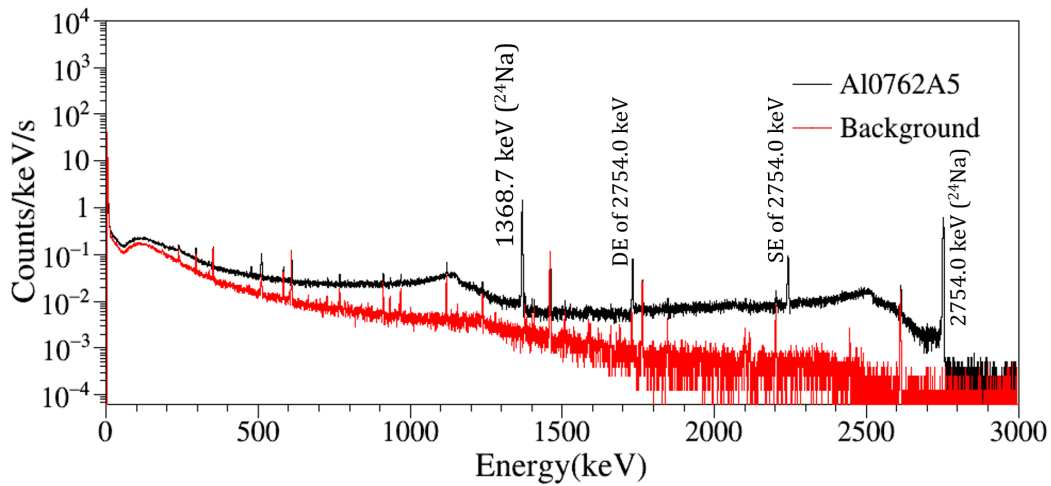
Table 27: ^{24}Al sample positions inside of the Activator (60 MeV p)

Run #	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
2	0.762	A5	16.9	0.2599
2	0.762	A6	36.9	0.2606
3 ¹	0.762	B4	42.9	0.2587
3	0.762	-B7	28.2	0.2601
14	0.762	B5	22.6	0.2599

Table 28: ^{24}Al sample positions inside of the Activator (30 MeV d)

Run	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
12	0.762	A5	16.9	0.2599
12	0.762	A6	36.9	0.2606
12	0.762	A7	23.8	0.2587
12	0.762	A8	16.3	0.2601

The γ -rays from ^{24}Na identified in the γ -ray spectrum are shown in Figure 86 & Figure 87. Escape peaks of 2754 KeV were also measured. DE and SE mean double escape peak and single escape peak. The γ -ray peak at 1368.7 KeV ($I_\gamma = 99.99\%$) was used for the calculation of the activity.

Figure 86: γ -ray spectrum from the irradiated ^{24}Al foil (run 02, p_{beam} , $T_{\text{cool}} = 14.5\text{h}$).

¹ The average beam intensity was of the order of $3.15\ \mu\text{A}$ and its standard deviation is of the order of $0.71\ \mu\text{A}$

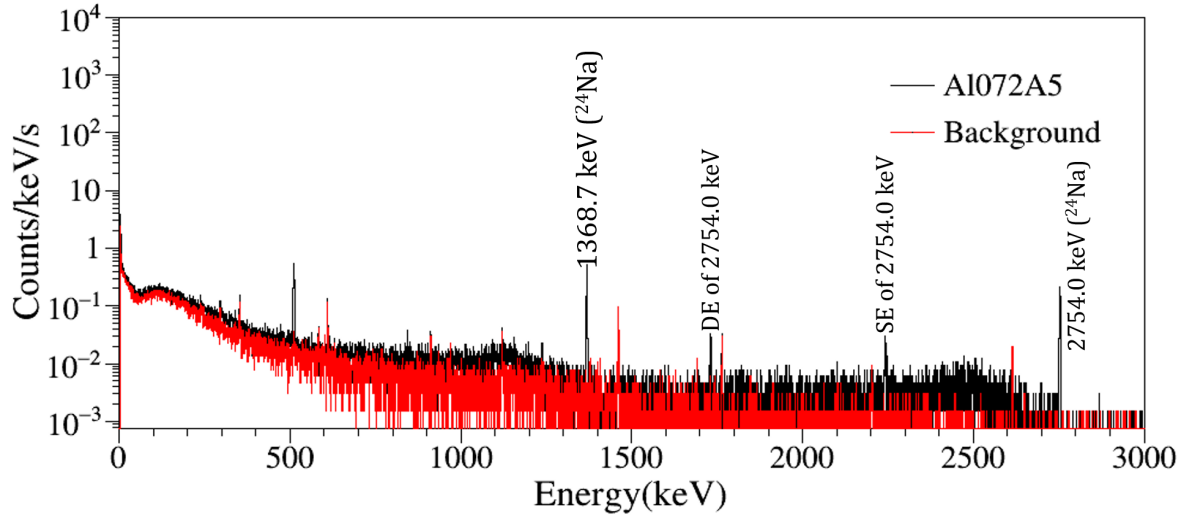


Figure 87: γ -ray spectrum from the irradiated ^{24}Na Al foil (run 12, d_{beam} , $T_{\text{cool}} = 1.2\text{h}$).

For the activity of ^{24}Na produced by $^{27}\text{Al}(n, \alpha)^{24}\text{Na}$ (Table 29 & Table 30) reaction shown in Figure 88 & Figure 89, we can remark that the activity decreases exponentially when the distance from Beryllium target increases.

Table 29: Experimental and MC Simulation Activities for ^{24}Na (60 MeV p)

Run #	Thickness (cm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
2	0.0762	A6	36.9	2.30E+02	9.23	9.81E+01	26.50	1.96E+02	26.30
3	0.0762	B4	42.9	5.44E+01	45.56	3.08E+01	18.30	3.64E+01	28.50
3	0.0762	B'7	28.2	1.59E+02	45.32	1.23E+02	19.50	1.07E+02	22.80
14	0.0762	B5	22.6	8.06E+02	4.24	6.15E+02	6.41	8.37E+02	16.70

Table 30: Experimental and MC Simulation Activities for ^{24}Na (30 MeV d)

Run #	Thickness (cm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
12	0.0762	A5	16.9	2.32E+03	14.34	5.81E+06	0.54	2.94E+03	16
12	0.0762	A6	36.9	1.07E+02	16.7	1.88E+05	0.9	6.63E+01	28
12	0.0762	A7	23.8	6.89E+01	26.08	4.89E+03	6.42	6.63E+01	36.4
12	0.0762	A8	16.3	2.76E+02	14.48	2.76E+04	1.92	2.08E+02	29.3

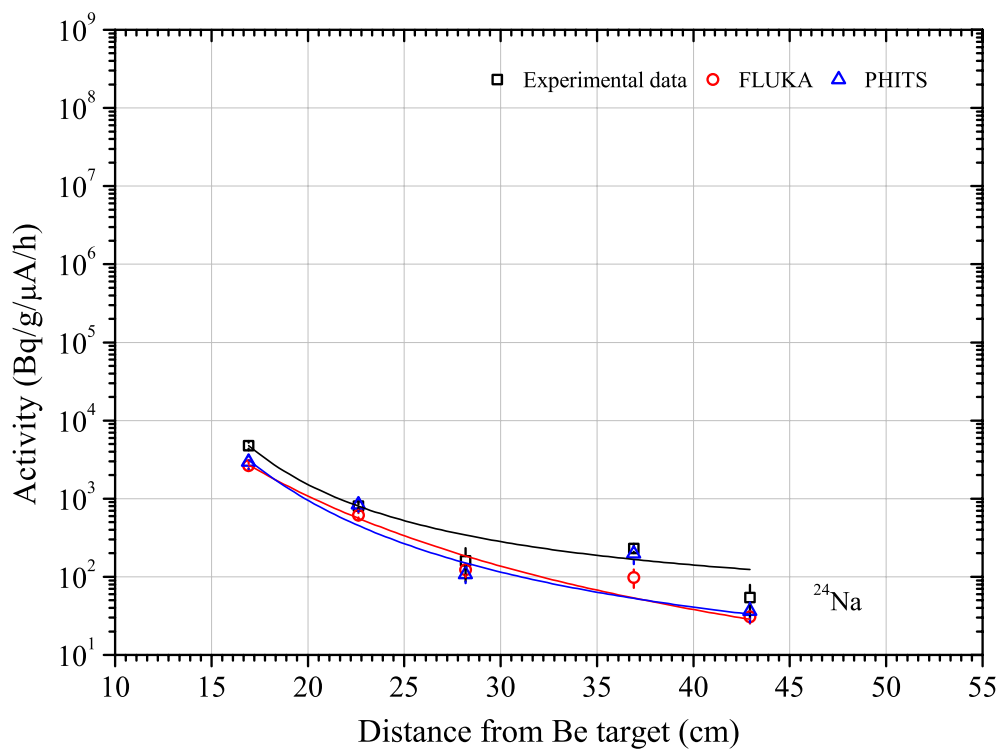


Figure 88: Measured activity for ^{24}Na (60 MeV p).

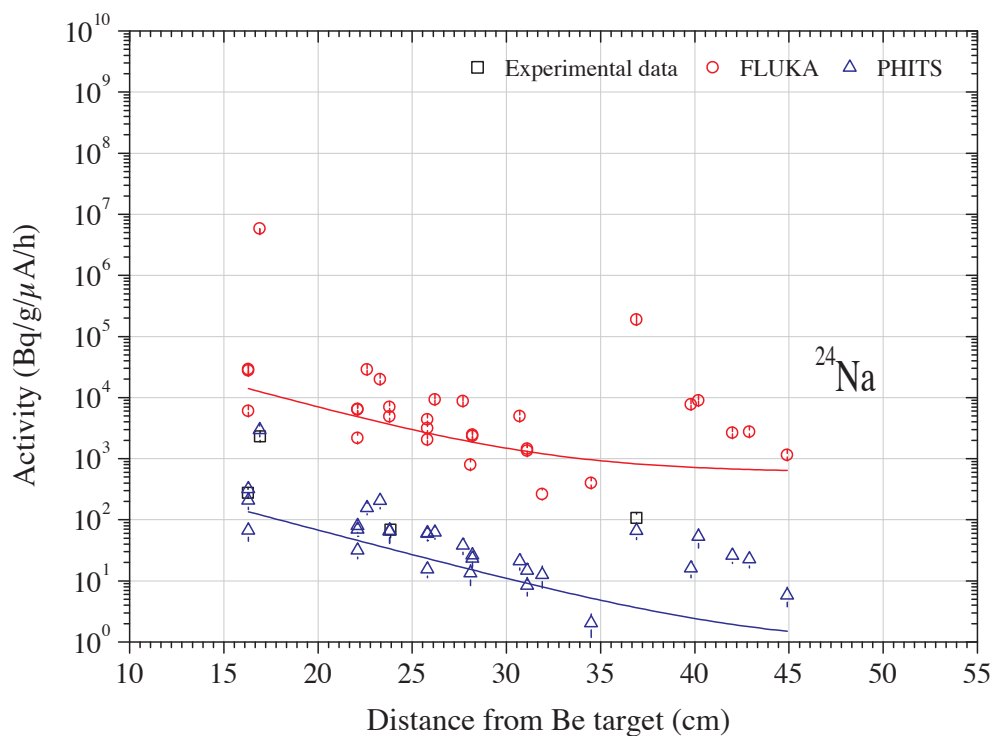


Figure 89: Measured activity for ^{24}Na (30 MeV d).

Figure 90 shows the results for the measured activities for proton and deuteron beams.

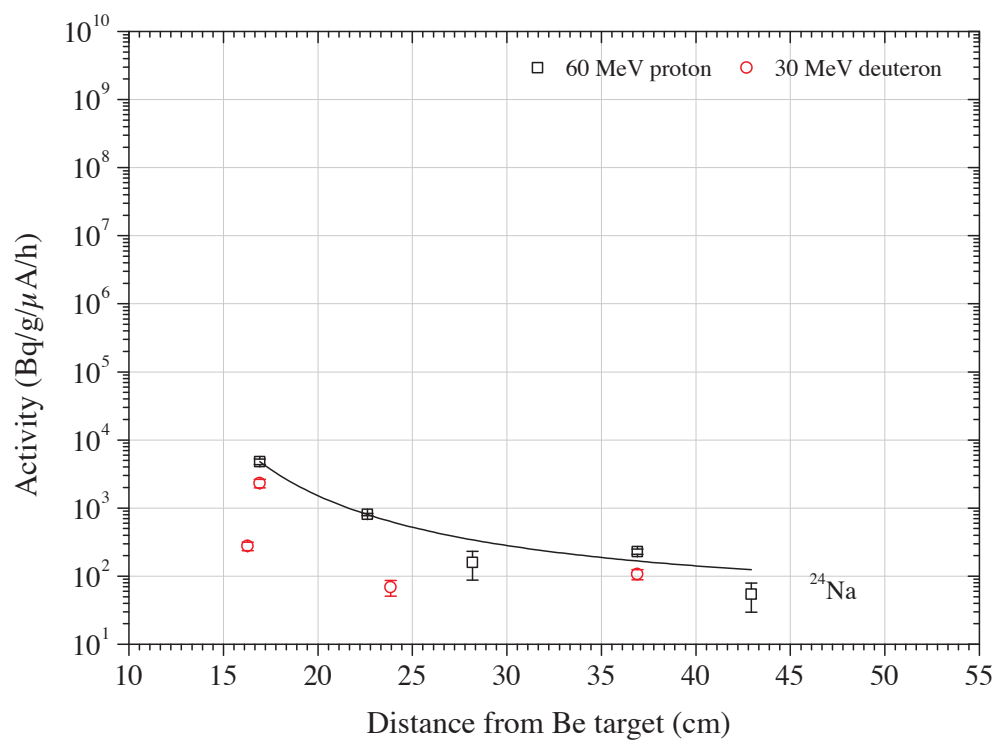


Figure 90: Measured activity for ^{24}Na with p & d .

5.6.1.5 Natural Zr Activation Foil

Natural Zr has five stable isotopes ^{90}Zr (51.45%), ^{91}Zr (11.22%), ^{92}Zr (17.15%), ^{94}Zr (17.38%), and ^{96}Zr (2.8%). We are interested in ^{97}Zr ($T_{1/2} = 16.749$ h) produced by $^{96}\text{Zr}(n, \gamma)^{97}\text{Zr}$ reaction (Figure 91) and ^{89}Zr ($T_{1/2} = 78.41$ h), which comes from $^{90}\text{Zr}(n, 2n)^{89}\text{Zr}$ reaction (Figure 92).

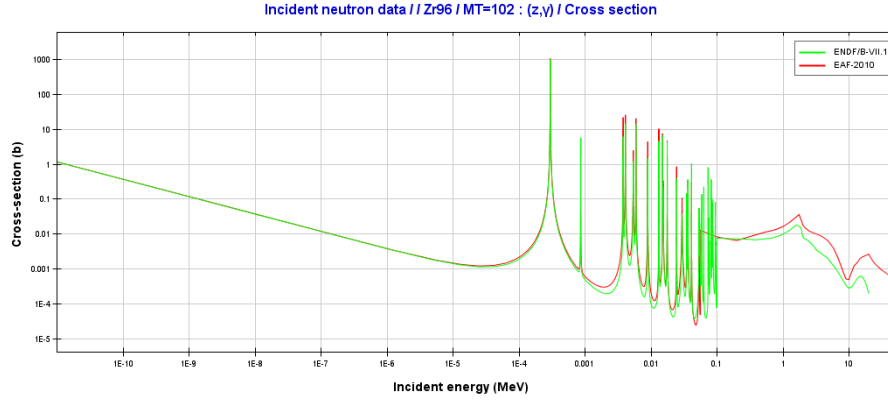


Figure 91: $^{96}\text{Zr}(n, \gamma)$ neutron Cross Section.

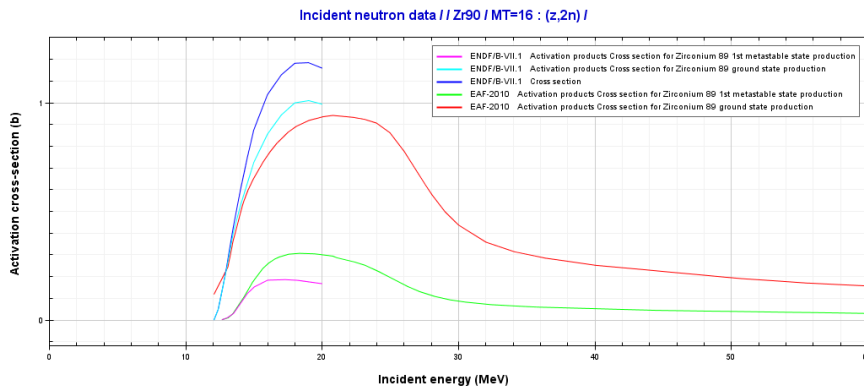


Figure 92: $^{90}\text{Zr}(n, 2n)$ neutron Cross Section.

60 MeV protons in different runs and positions inside of the Activator irradiated natural Zr. These activation foils were also used to study the neutron leakage (Run02) by checking the symmetry between top part (above the target) and the bottom (below the target) of the Activator.

^{90}Zr activation foils were placed and irradiated with a 60 MeV proton beam in different positions of the Activator device (Figure 93). ^{90}Zr activation foils used in the experiment are shown in Table 31.

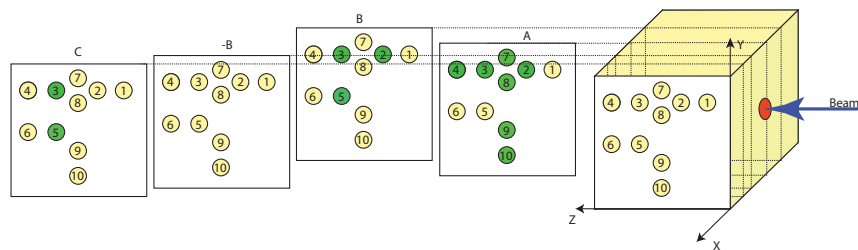
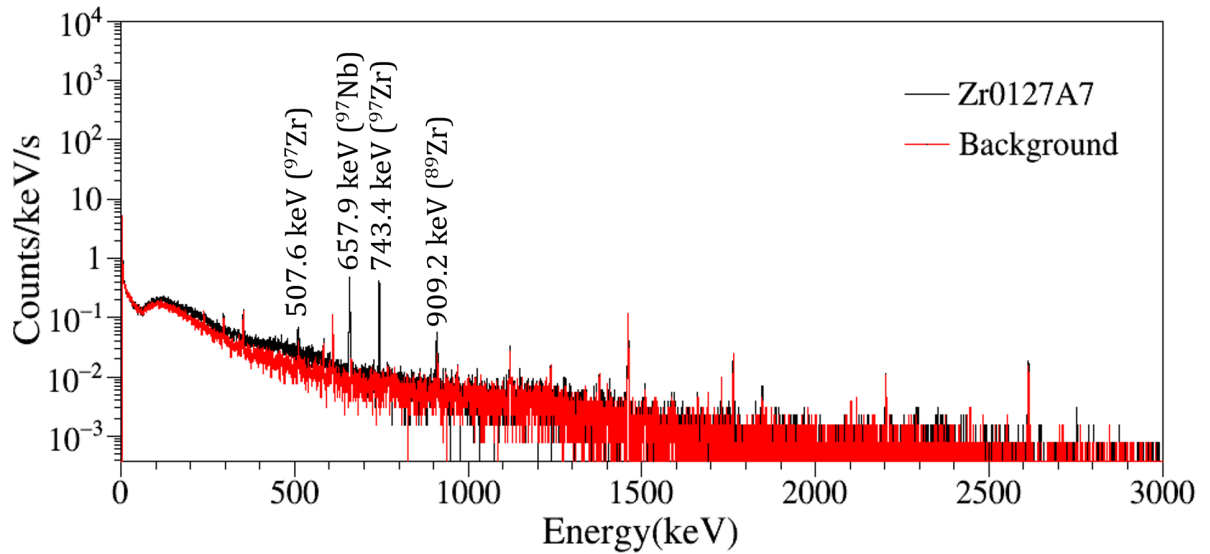


Figure 93: ^{90}Zr sample positions inside of the Activator (Runs 2, 8 & 11).

Table 31: ^{Nat}Zr sample positions inside of the Activator for all runs

Run #	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
2	0.127	A7	23.8	0.1121
2		A8	16.3	0.1116
2		A9	16.3	0.1117
2		A10	23.8	0.112
8	0.127	A2	16.3	0.1121
8		A4	40.2	0.1116
8		B5	22.6	0.1117
8		C5	26.2	0.112
11	0.127	A3	23.3	0.1121
11		B2	22.1	0.1116
11		B3	27.7	0.1117
11		C3	30.7	0.112

Three isotopes ^{89}Zr ($T_{1/2} = 78.41$ h), ^{97}Nb ($T_{1/2} = 72.1$ m), and ^{97}Zr ($T_{1/2} = 16.749$ h) were identified in the γ -ray spectrum (Figure 94). ^{97}Nb is the decay product ^{97}Zr .

Figure 94: γ -ray spectrum from the irradiated Zr foil (run02, p_{beam} , $T_{\text{cool}} = 4.8$ h)

The activity of ^{97}Zr from $^{96}\text{Zr} (n, \gamma)^{97}\text{Zr}$ reaction is shown in Table 32 and Figure 96 shows the activity in function of the distance from the center of the target and we can remark that it decreases slowly with the distance. It is important to notice that the most important resonance peak is located near to 300 eV, which in principle is similar to what we see in the activity behavior of ^{99}Mo (Figure 95).

Incident neutron data / ENDF/B-VII.1 / MT=102
: (z, γ) / Cross section + Std. deviation

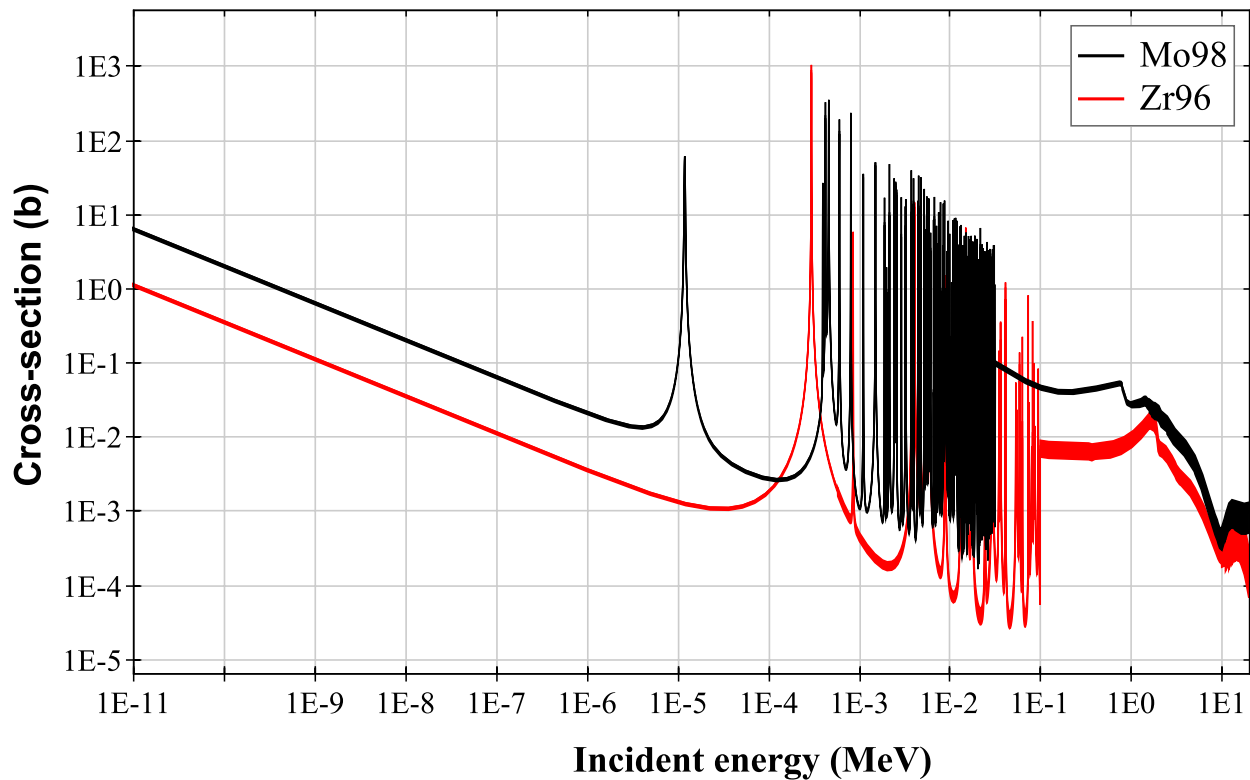


Figure 95: $^{98}\text{Mo}(n,\gamma)^{99}\text{Mo}$ & $^{96}\text{Zr}(n,\gamma)^{97}\text{Zr}$ reaction cross sections.

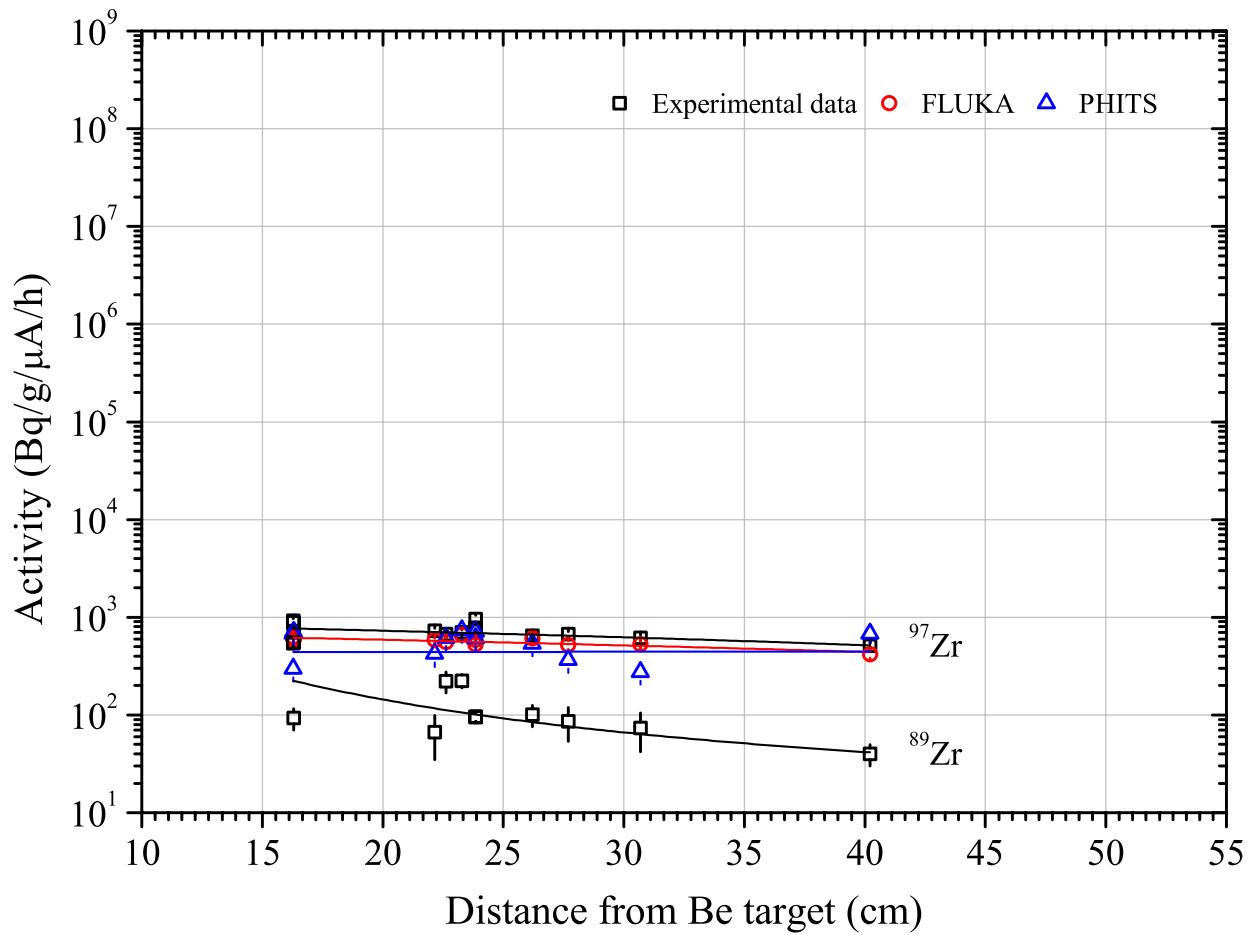
Table 32: Experimental and MC Simulation Activities for ^{97}Zr

Run #	Thickness (cm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
2	0.0127	A10	23.8	7.80E+02	9.19	5.28E+02	5.03	6.12E+02	24.4
2	0.0127	A7	23.8	9.59E+02	9.2	5.84E+02	8.46	7.13E+02	26.5
2	0.0127	A8	16.3	9.22E+02	9.37	6.04E+02	6.97	6.83E+02	27.6
2	0.0127	A9	16.3	8.88E+02	9.24	6.26E+02	5.87	2.99E+02	26.4
8	0.0127	A2	16.3	7.18E+02	4.27	5.86E+02	12.2	7.06E+02	22.7
8	0.0127	A4	40.2	5.12E+02	4.4	4.20E+02	9.61	6.81E+02	19.2
8	0.0127	B5	22.6	6.74E+02	4.28	5.57E+02	9.87	6.15E+02	25.1
8	0.0127	C5	26.2	6.45E+02	4.27	6.10E+02	11.1	5.35E+02	25.6
11	0.0127	A3	23.3	7.01E+02	6.25	6.67E+02	14	7.35E+02	25.5
11	0.0127	B2	22.1	7.27E+02	6.19	5.84E+02	4.93	4.22E+02	26.7
11	0.0127	B3	27.7	6.72E+02	6.2	5.21E+02	10.3	3.65E+02	26.1
11	0.0127	C3	30.7	6.16E+02	6.99	5.34E+02	5.96	2.76E+02	25.5

Table 33 and Figure 96 show the experimental measured activity for ^{89}Zr . For $^{90}\text{Zr}(n, 2n)^{89}\text{Zr}$ reaction we can observe the energy threshold is about 12 MeV from Figure 92, so we can deduce neutrons above this threshold can perform this kind of nuclear reaction.

Table 33: Experimental and MC Simulation Activities for ^{89}Zr

Run #	Thickness (cm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
2	0.0127	A10	23.8	9.48E+01	10.23				
2	0.0127	A7	23.8	9.63E+01	10.35				
2	0.0127	A8	16.3	5.55E+02	9.32				
2	0.0127	A9	16.3	5.40E+02	9.22				
8	0.0127	A2	16.3	9.33E+01	24.54				
8	0.0127	A4	40.2	4.01E+01	24.54				
8	0.0127	B5	22.6	2.22E+02	24.54				
8	0.0127	C5	26.2	1.01E+02	24.54				
11	0.0127	A3	23.3	2.24E+02	15.75				
11	0.0127	B2	22.1	6.69E+01	47.78				
11	0.0127	B3	27.7	8.64E+01	37.58				
11	0.0127	C3	30.7	7.35E+01	43				

Figure 96: Measured activity for ^{97}Zr & ^{89}Zr .

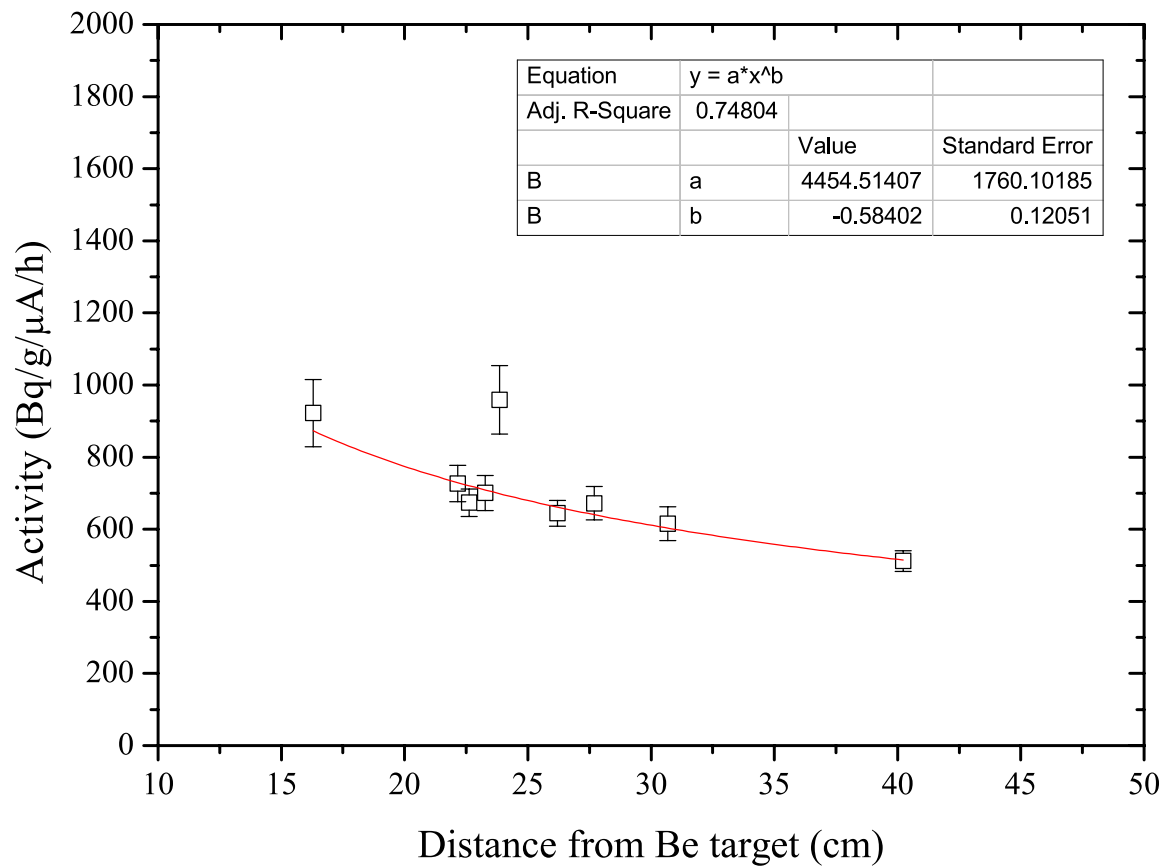


Figure 97: Measured activity (fitting curve) for ^{97}Zr .

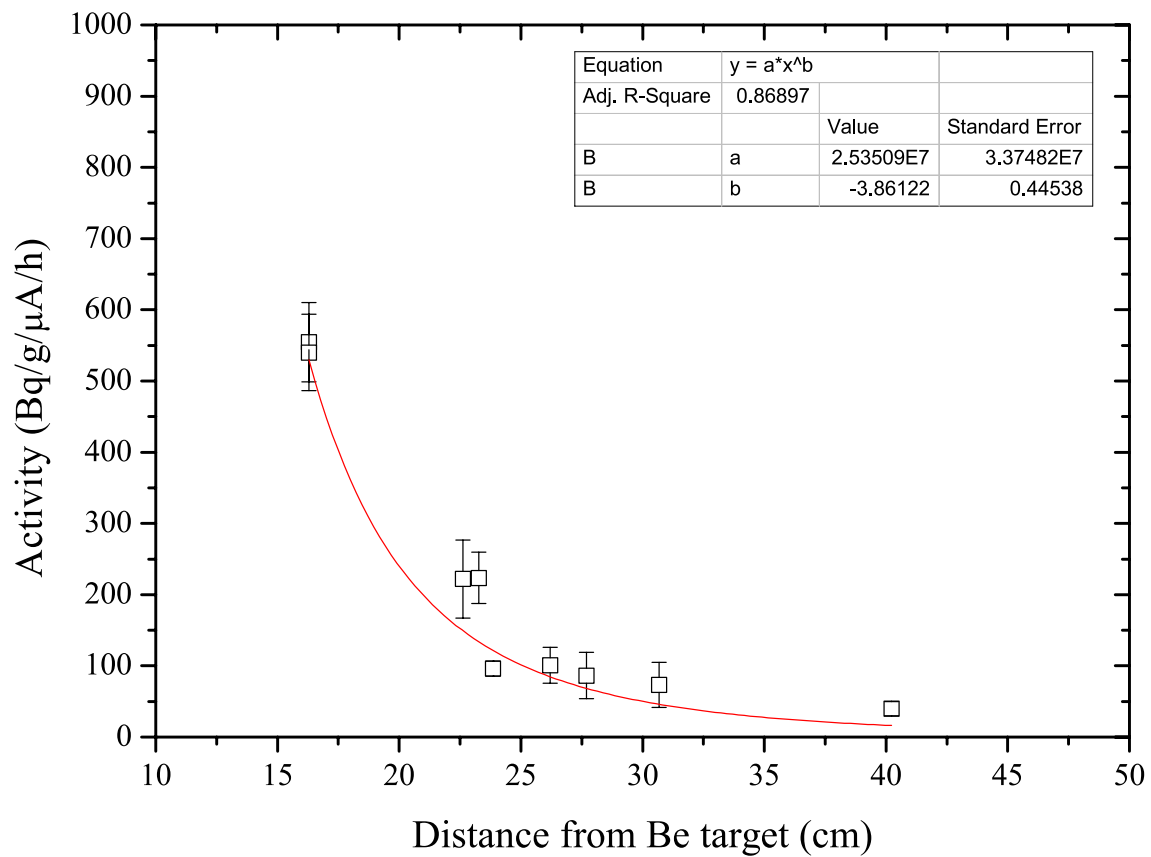


Figure 98: Measured activity (fitting curve) for ^{89}Zr .

5.6.1.6 Natural Cu Activation Foil

Natural Cu has two stable isotopes ^{63}Cu (69.15%), ^{65}Cu (30.85%). We are interested in ^{64}Cu ($T_{1/2} = 12.7$ h) produced by $^{63}\text{Cu} (n, \gamma)^{64}\text{Cu}$ reaction (Figure 99).

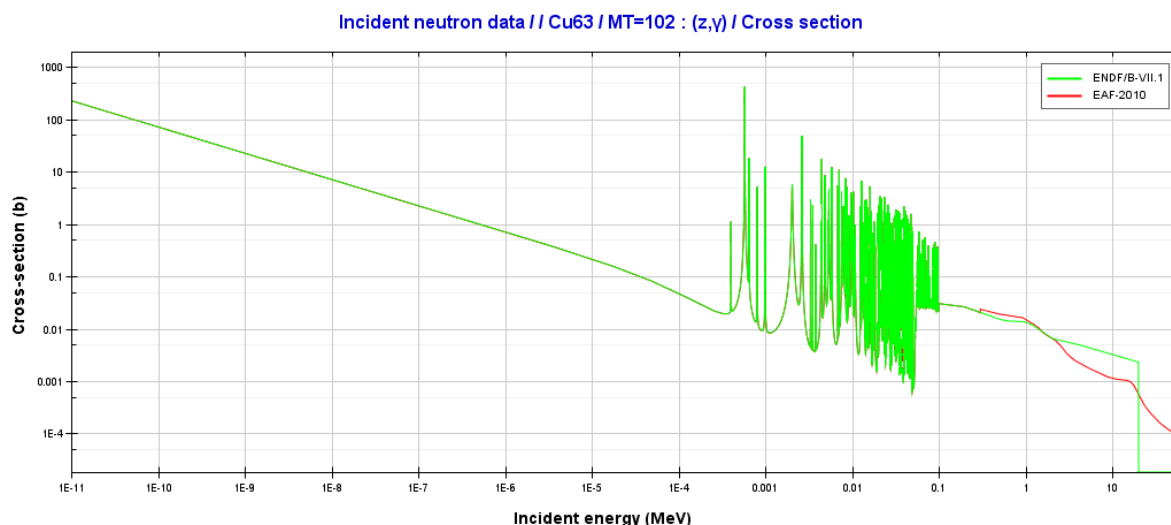


Figure 99: $^{63}\text{Cu} (n, \gamma)$ neutron Cross Section.

Natural Cu foils were irradiated with a 60 MeV proton only in two different positions in the Activator (Figure 100 & Table 34).

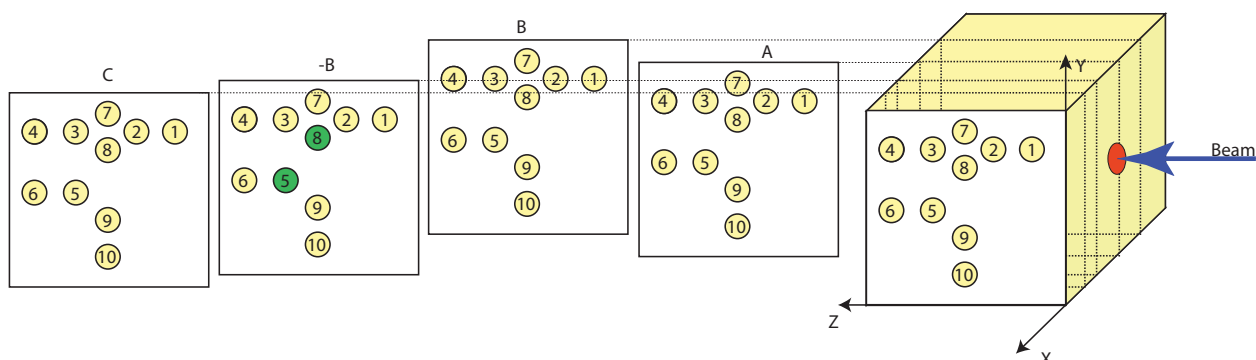


Figure 100: ^{Nat}Cu sample positions inside of the Activator (Runs 3² & 14).

Table 34: ^{Nat}Cu sample positions inside of the Activator for all runs

No	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
2	0.127	-B8	22.1	0.1454
16	0.127	-B5	22.6	0.1451

In the γ -ray spectrum (Figure 101), ^{64}Cu ($T_{1/2} = 12.7$ h) was identified at 1345.8 KeV.

² The beam was very unstable and interrupted two times during the irradiation. The real irradiation time without interruption was 3129 s. The average of beam intensity was 2.913 μA . And the standard deviation of the beam intensity was 0.71 μA .

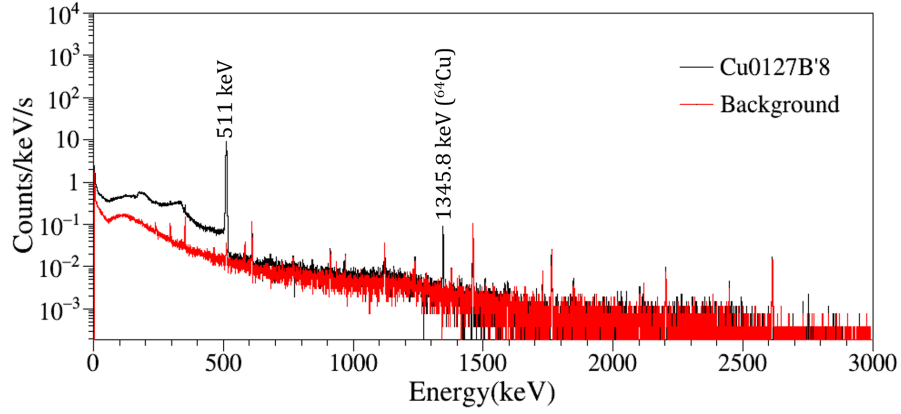


Figure 101: γ -ray spectrum from the irradiated Cu foil (run3, p_{beam} , $T_{cool} = 8.8$ h).

The experimental and MC simulation activities from $^{63}\text{Cu} (n, \gamma)^{64}\text{Cu}$ reaction is shown in Table 35 and Figure 102. We have only two activity values in the experimental data.

Table 35: Experimental and MC Simulation Activities for ^{64}Cu

Run #	Thickness (cm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
3	0.0127	-B8	22.1	1.03E+05	45.31	7.70E+04	2.48	4.98E+04	8.88
14	0.0127	-B5	22.6	8.28E+04	6.21	6.10E+04	1.68	4.94E+04	10.30

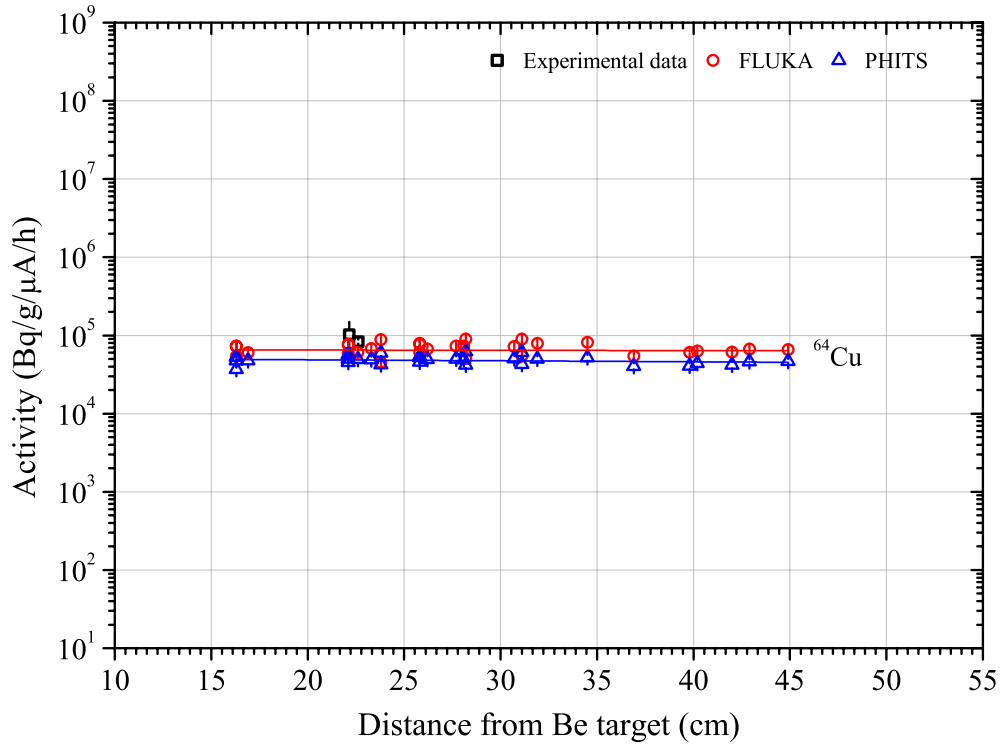


Figure 102: Measured activity for ^{64}Cu .

5.6.1.7 Natural Sc Activation Foil

Natural Sc has one stable isotope ^{45}Sc . We are interested in ^{46}Sc ($T_{1/2} = 83.79$ d) produced by the $^{45}\text{Sc}(n, \gamma)$ reaction (Figure 103) and ^{44}Sc ($T_{1/2} = 3.97$ h) and $^{44\text{m}}\text{Sc}$ ($T_{1/2} = 58.61$ h, third metastable state) are produced by $^{45}\text{Sc}(n, 2n)$ reaction (Figure 104).

Natural Sc foils were irradiated by 60 MeV protons, inside of the Activator (Figure 105 & Table 36). The average of beam intensity was $2.913 \mu\text{A}$ and the standard deviation of the beam intensity was $0.71 \mu\text{A}$. The A5 position is aligned in front to the beryllium target. In Figure 65, we can also observe ^{44}Sc , $^{44\text{m}}\text{Sc}$ apart from the two typical gamma lines of ^{46}Sc .

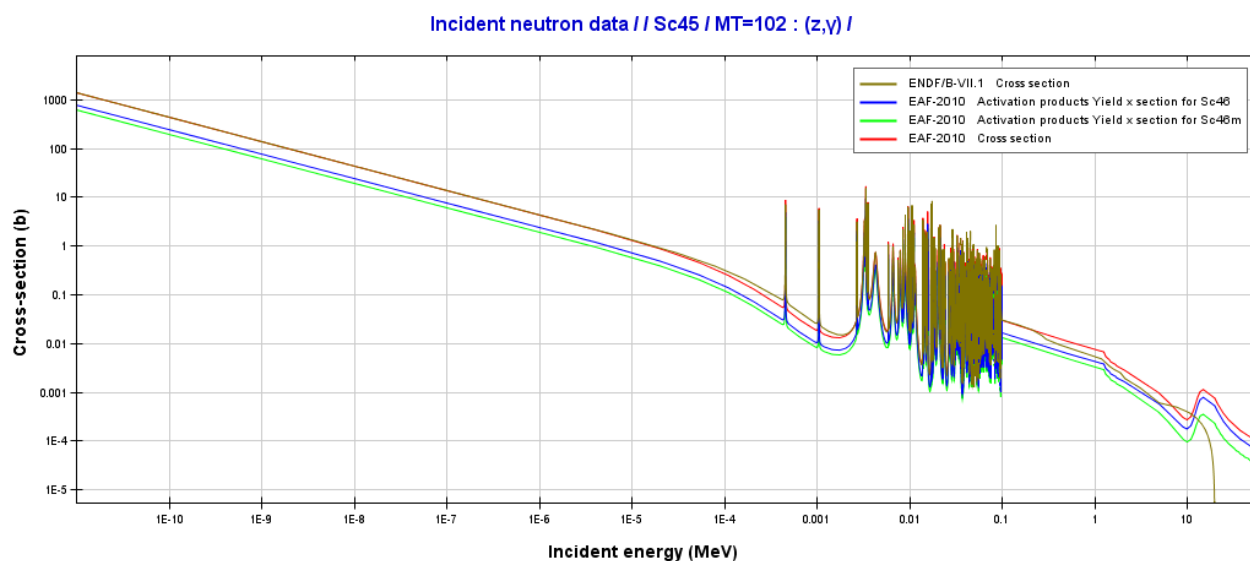


Figure 103: $^{45}\text{Sc}(n, \gamma)$ neutron Cross Section.

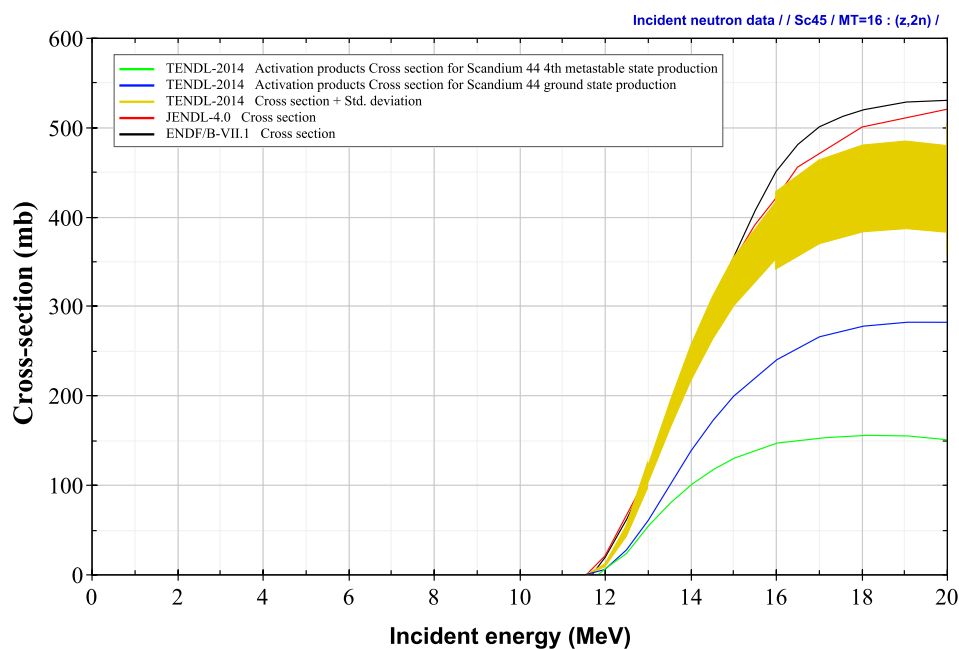


Figure 104: $^{45}\text{Sc}(n, 2n)$ neutron Cross Section.

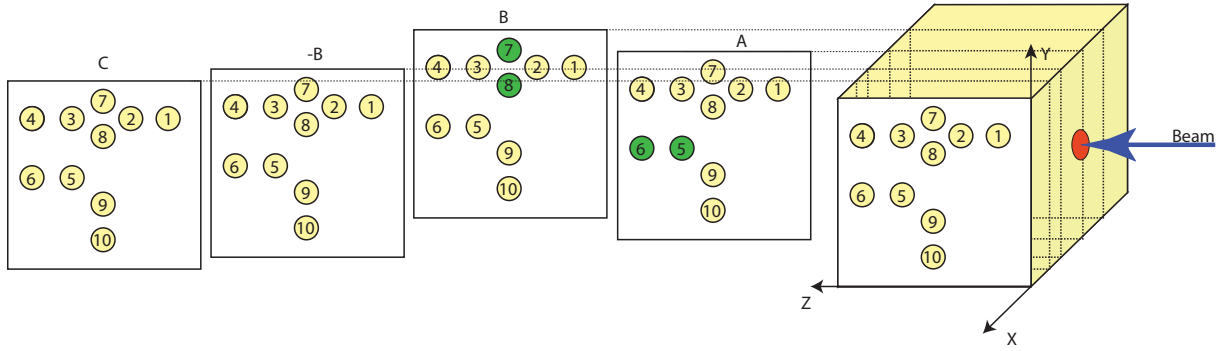


Figure 105: ^{Nat}Sc sample positions inside of the Activator (Run 3).

However in A6 position which is more distant from the Be target we can only observe the presence of the two gamma lines of ^{46}Sc (Figure 107).

Table 36: ^{Nat}Sc sample positions inside of the Activator for all runs

Run #	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
3	0.127	A5	16.9	0.01084
3	0.127	A6	36.9	0.00994
3	0.127	B7	28.2	0.01131
3	0.127	B8	22.1	0.01115

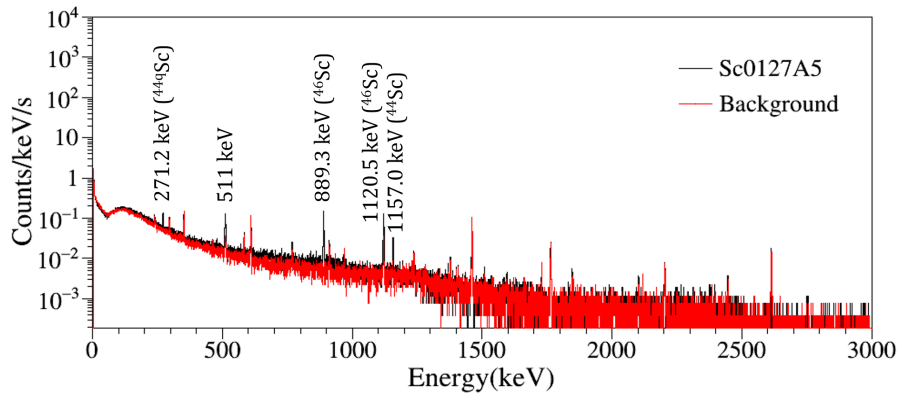


Figure 106: γ -ray spectrum from the irradiated ^{Nat}Sc foil (run03, p_{beam} , $T_{\text{cool}} = 23.9$ h).

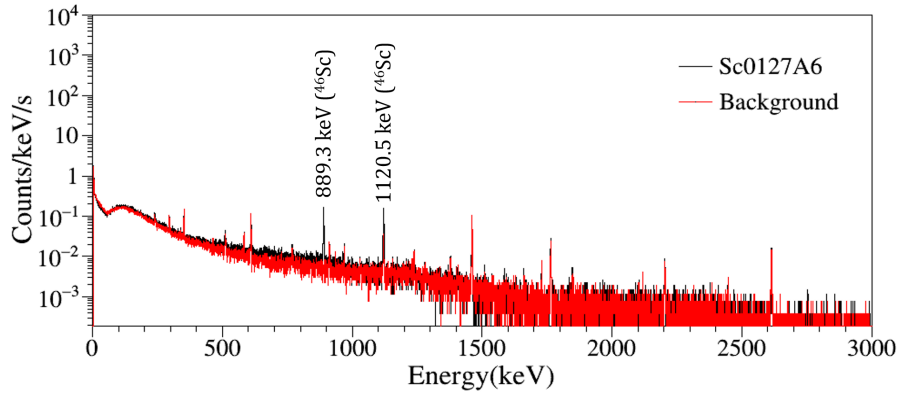


Figure 107: γ -ray spectrum from the irradiated Sc foil (run03, p_{beam} , $T_{\text{cool}} = 26.0$ h)

Table 37 & Figure 108 show the experimental measured activity for ^{46}Sc . 889.3 KeV γ -ray peak ($I_\gamma = 98.46\%$) was used for the activity calculation.

Table 37: Experimental and MC Simulation Activities for ^{46}Sc

Run #	Thickness (cm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
3	0.0127	A5	16.9	4.39E+03	45.24	2.20E+03	1.62		
3	0.0127	A6	36.9	5.48E+03	45.24	3.04E+03	2.70		
3	0.0127	B7	28.2	8.51E+03	45.23	5.46E+03	3.31		
3	0.0127	B8	22.1	6.72E+03	45.23	4.12E+03	3.08		

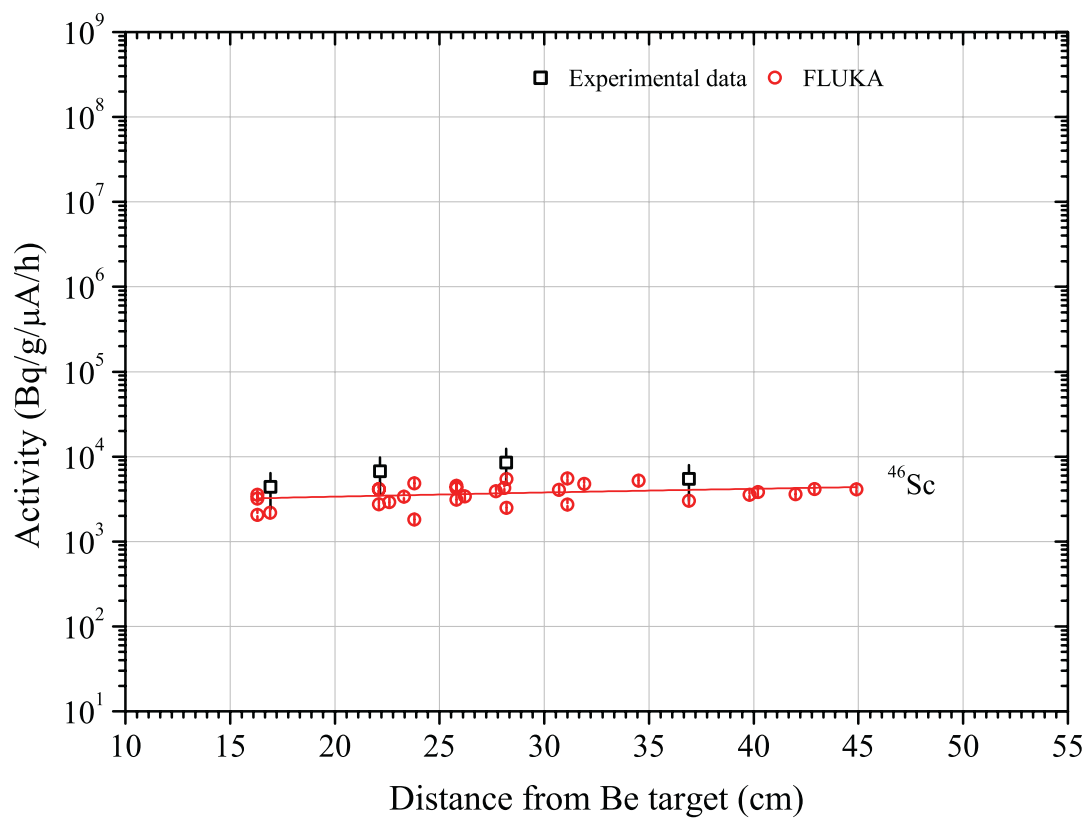


Figure 108: Measured activity for ^{46}Sc .

5.6.1.8 Activation Foils (In, Au, W & Al) Resume

In this section we present a comparison with the data taken with natural In, Au, W and Al activation foils.

The energies 1.46 eV, 4.96 eV, and 18.9 eV have been chosen because at these energies the Au, In, and W used in activation foils exhibit strong resonances in their neutron absorption cross-sections.

As seen in all previous calculations in the case of proton beam on Be target (Figure 109), experimental data of different activation foils are consistent with each other and are also consistent with the predictions of Monte Carlo using FLUKA and PHITS.

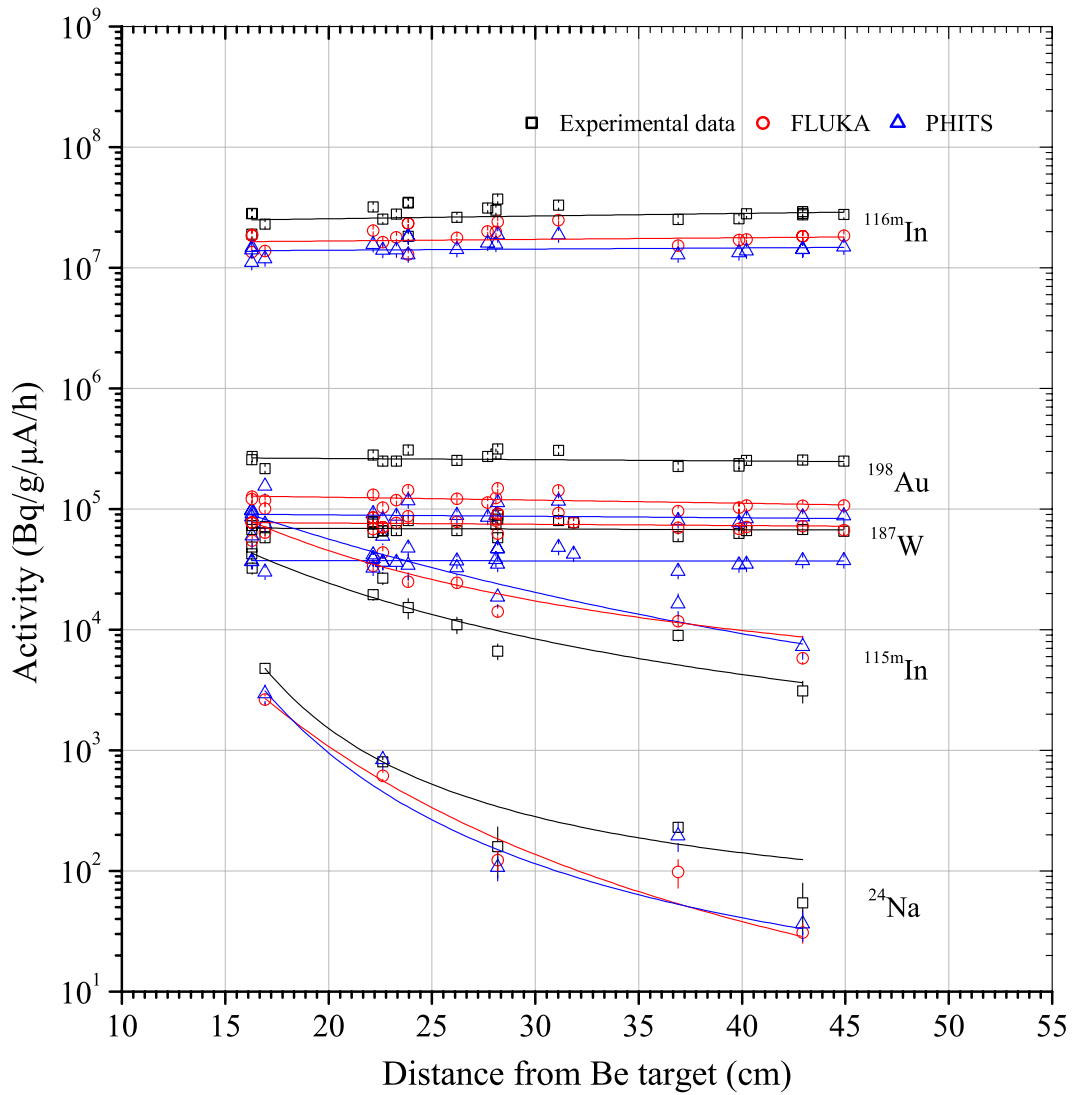


Figure 109: Measured activity for In, Au, W and Al activation foils (60 MeV p).

Figure 110 shows the activities for activation foils with a deuteron beam: Au, W and Al. The experimental results are shown together with the results of MC simulation with PHITS.

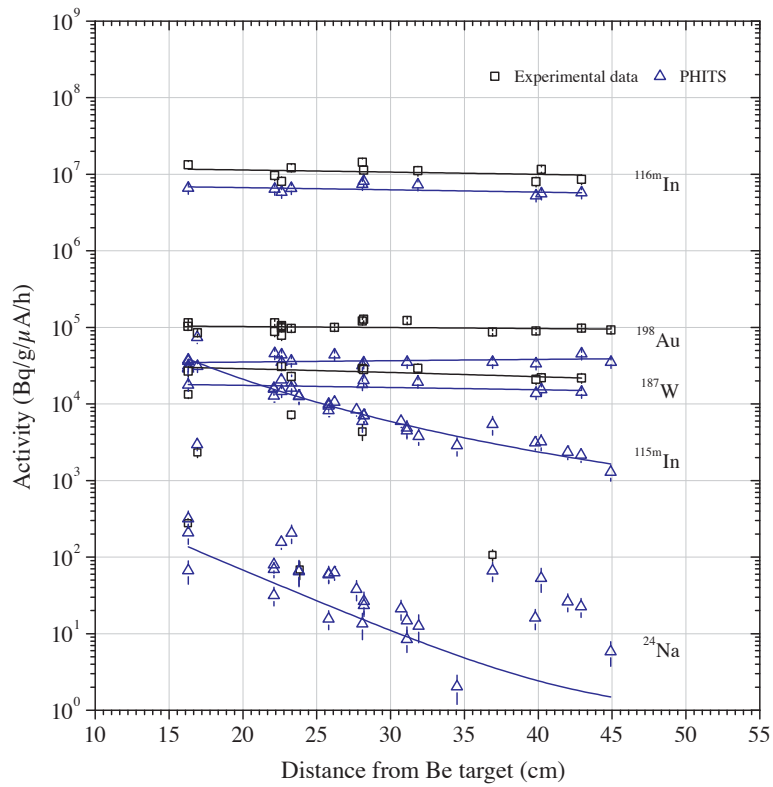


Figure 110: Measured activity for In, Au, W and Al activation foils (30 MeV d).

Figure 111 shows the results for the measured activities with proton and deuteron beams.

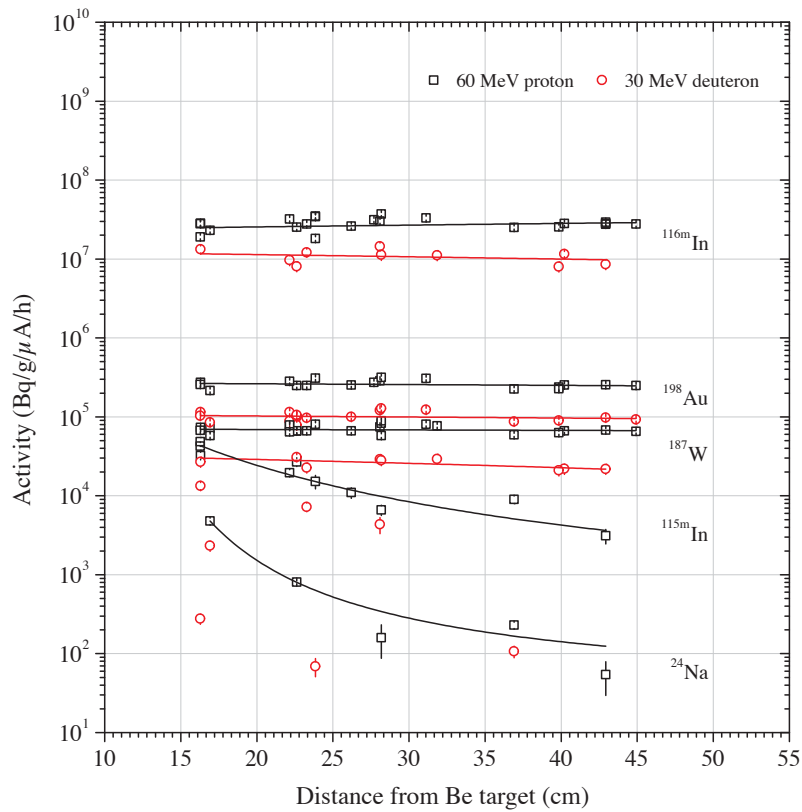


Figure 111: Measured activity for In, Au, W and Al activation foils.

5.7 Medical Isotopes

Five kinds of isotope samples were activated inside of the activator: ^{98}Mo (98.61%) metal powder, and $^{\text{nat}}\text{Mo}$, $^{\text{nat}}\text{Ho}$, $^{\text{nat}}\text{Lu}$, and $^{\text{nat}}\text{Re}$ metal foils with different thicknesses.

As it was mentioned in the Chapter 1, one of possible routes of the ^{99}Mo production is via the nuclear reaction $^{98}\text{Mo}(n, \gamma)^{99}\text{Mo}$. The isotope ^{99}Mo , which is created by an irradiation, emits β^- particles (100%) with the maximum energy 1210 KeV and the main γ -quanta with energy 181.1, 739.5 KeV (intensity of γ -lines $I_\gamma = 6.08\%$, 12.1% respectively). As a result of β^- decay ^{99}Mo ($T_{1/2} = 66$ h) transforms in $^{99\text{m}}\text{Tc}$ ($T_{1/2} = 6.02$ h), which emits photons with the basic energy 141 KeV ($I_\gamma = 87.7\%$).

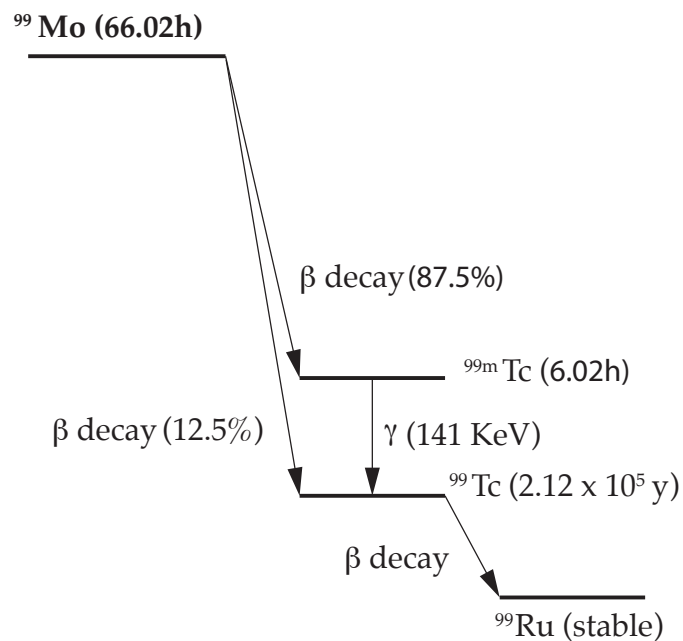


Figure 112: ^{99}Mo decay chain

Diagnostic imaging in nuclear medicine and $^{99\text{m}}\text{Tc}$ are synonymous for over 40 years. Almost 80% of the studies in nuclear medicine involve the use of $^{99\text{m}}\text{Tc}$ (almost 10% in the case of ^{18}F and 10% for the rest). Currently, most hospital radio pharmacies receive $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ generators on a weekly or twice weekly basis. Generators are eluted every morning and the $^{99\text{m}}\text{Tc}$ obtained is used to prepare a variety of radiopharmaceuticals labeled with. Most of these products have half-lives of six to eight hours after labeling and therefore may occur in one batch per day.

The main applications of $^{99\text{m}}\text{Tc}$ in nuclear medicine are in myocardial perfusion (heart patients) and bone metastases (cancer patients). Other uses of $^{99\text{m}}\text{Tc}$ are in brain (regional cerebral blood flow), liver (gastroenterology), lungs thyroid, and kidney imaging (A.N. Dovbnya et al., 2009).

Radionuclides considered suitable for therapy may be divided into three categories, i.e. beta-particle emitters, alpha particle emitters, and Auger and Coster-Kronig electron emitters.

Most of the radionuclides in routine use for therapy are beta emitters. Beta particles are of low Linear Energy Transfer (LET) and their Relative Biological effectiveness's (RBE) is unity. The range of beta particles is considerably greater than the diameter of a typical cell (5~20 μm), and thus, the whole cell and surrounding cells are irradiated.

The isotope ^{166}Ho ($T_{1/2} = 1.2 \text{ d}$) is primarily a β -emitter ($E_{\text{max}}, 1.85 \text{ MeV}$) with a maximum path length of 8.7 mm in soft tissue and 3.8 mm in bone and is produced in the nuclear reaction $^{165}\text{Ho}(n, \gamma)^{166}\text{Ho}$ with a major γ at 6.95 & 80.6 KeV.

The main applications of ^{166}Ho in nuclear medicine are as seed sources that can deliver high radiation doses to the tumor, while the short range of the beta particles limits damage to the adjacent normal tissue. A beta emitter ^{166}Ho seed has been also proposed for treating small HCC tumors (small hepatocellular carcinomas - bioglass ^{166}Ho) and for Multiple myeloma diseases arising from plasma cells in the bone marrow, with bony involvement leading to cortical bone destruction. The physical half-life, 1.2 d, is short enough to permit delivery of high-dose chemotherapy and reinfusion of cryopreserved peripheral blood stem cells within 6 to 10 d. This short time frame after myeloablative chemotherapy is advantageous, compared with high-dose ^{131}I or ^{90}Y therapy (Hazel B. et al.).

The isotope ^{177}Lu ($T_{1/2} = 6.6 \text{ d}$) is produced in the neutron capture reaction $^{176}\text{Lu}(n, \gamma)^{177}\text{Lu}$ with a major γ at 208.4 KeV and is an attractive radionuclide for various therapeutic applications (cancer therapy). In FESTA we are using $^{\text{Nat}}\text{Lu}$, where the content of ^{176}Lu is about 2.59%. Through successful clinical applications in the field of brachytherapy and Endoradiotherapy there is growing demand for the nuclide ^{177}Lu worldwide. The main applications of ^{177}Lu in nuclear medicine are for heart disease treatment (restenosis therapy).

The isotope ^{188}Re ($T_{1/2} = 17 \text{ h}$) is produced in the nuclear reaction $^{187}\text{Re}(n, \gamma)^{188}\text{Re}$. (Major γ at 59 KeV) and is a radionuclide in which there is widespread interest for therapeutic purposes because of its favorable physical characteristics.

The main applications of ^{188}Re in nuclear medicine are for cancer treatment/diagnostics, monoclonal antibodies, bone cancer pain relief, treatment of rheumatoid arthritis, treatment of prostate cancer and treating bone pain. Also in hematology, radio immunotherapy by means of ^{188}Re might play a role by selectively targeting the bone marrow in patients undergoing conditioning prior to hematopoietic stem cell transplantation. The feasibility of such an approach was proven using a Re-labeled monoclonal antibody directed toward the CD66-antigen. Recent studies have found also to be clinically efficient for lung neoplasms carcinoma, non-small-cell lung carcinoma, small cell, and neoplasm recurrence applications.

Regarding the interest of a wide application of these medical radioisotopes, IBEL SA has undergone a very specific survey in Switzerland (Annex 3) that has covered a wide

range of radionuclides for clinical and scientific research for which it was important to know about their availability, frequency of use and importance.

5.7.1 ^{Nat}Mo

Natural molybdenum has seven stable isotopes ⁹²Mo (14.84%), ⁹⁴Mo (9.25%), ⁹⁵Mo (15.92%), ⁹⁶Mo (16.68%), ⁹⁷Mo (9.55%), ⁹⁸Mo (24.13%), and ¹⁰⁰Mo (9.63%).

The main purpose was to activate ^{Nat}Mo samples inside of the activator using 60 MeV proton and 30 MeV deuteron beams. The samples were irradiated for 1 hour.

The average proton beam current was 4.13 μ A and 4.11 μ A for runs 8 & 9 respectively. The average deuteron beam current was 1.03 μ A for run 12.

In order to study the thickness effect of ^{Nat}Mo foils when irradiated, two types of thicknesses were considered: 0.1mm and 0.25 mm.

Foils of 0.1 mm and 0.25 thicknesses were also used joining 0.025 mm thickness foils together.

^{Nat}Mo foils were placed and irradiated in different positions inside of the Activator (Figure 113 & Figure 114).

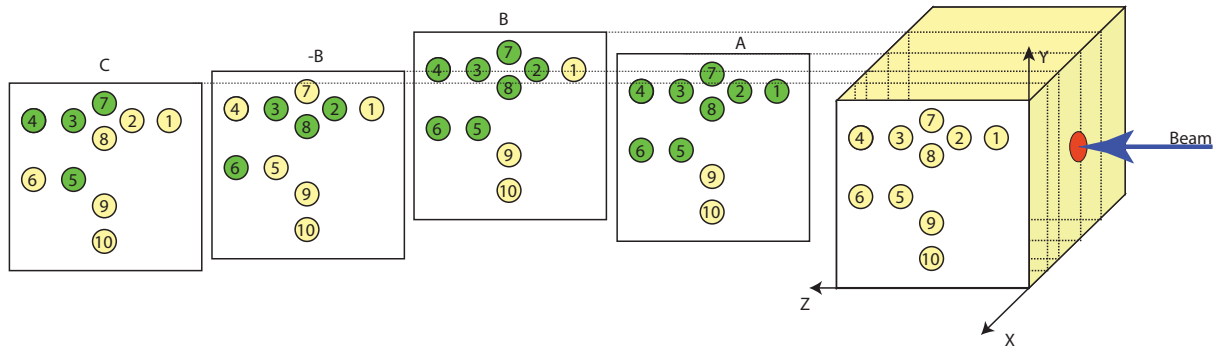


Figure 113: ^{Nat}Mo sample positions inside of the Activator (60 MeV p).

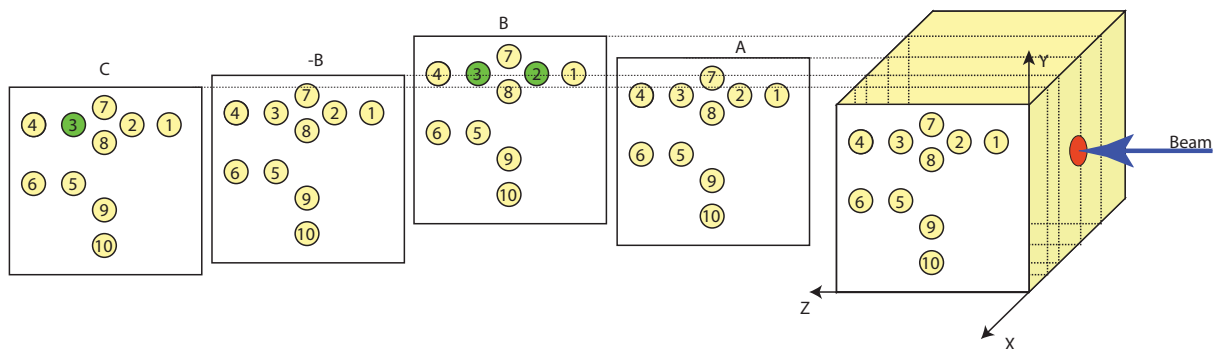


Figure 114: ^{Nat}Mo sample positions inside of the Activator (30 MeV d).

Table 38 & Table 39 show the sample positions inside of the Activator for p & d beams.

Table 38: ^{Nat}Mo sample positions inside of the Activator (60 MeV p)

Run	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
8	1	A3	23.3	0.832
8	1	B3	27.7	0.843
8	1	C3	30.7	0.889
9	1	A1	28.1	0.768
9	1	A2	16.3	0.959
9	1	A4	40.2	0.869
9	1	A5	16.9	0.854
9	1	A6	36.9	0.888
9	1	A7	23.8	0.846
9	1	A8	16.3	0.885
9	1	B4	42.9	0.858
9	1	B5	22.6	0.841
9	1	B6	39.8	0.855
9	1	B7	28.2	0.859
9	1	B8	22.1	0.874
9	1	C4	44.9	0.813
9	1	C5	26.2	0.833
9	1	C7	31.1	0.846
9	0.1	B2	22.1	0.09417
9	0.1	B3	27.7	0.09267
9	0.1	-B6	39.8	0.0912
9	0.1	-B8	22.1	0.09075
9	0.25	-B2	22.1	0.22573
9	0.25	-B3	27.7	0.22548
13	1	-B8	22.1	0.874

Table 39: ^{Nat}Mo sample positions inside of the Activator (30 MeV d)

Run	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
12	1	B2	22.1	0.846
12	1	B3	27.7	0.886
12	1	C3	30.7	0.798

^{99}Mo comes from $^{98}\text{Mo}(n, \gamma)^{99}\text{Mo}$ neutron capture reaction (Figure 115). ^{99m}Tc is the decay product of ^{99}Mo (β^- decay, 87.89 %) (Chadwick, M. et al.). ^{99}Mo ($T_{1/2} = 65.976$ h) and ^{99m}Tc ($T_{1/2} = 6.0067$ h) were identified in the gamma-ray spectra (Figure 116 & Figure 117).

The peak used for the measured activity estimation was: $E_\gamma = 739.5$ KeV, $I_\gamma = 12.26$ % (see Table 41; Figure 118, Figure 119 & Figure 120).

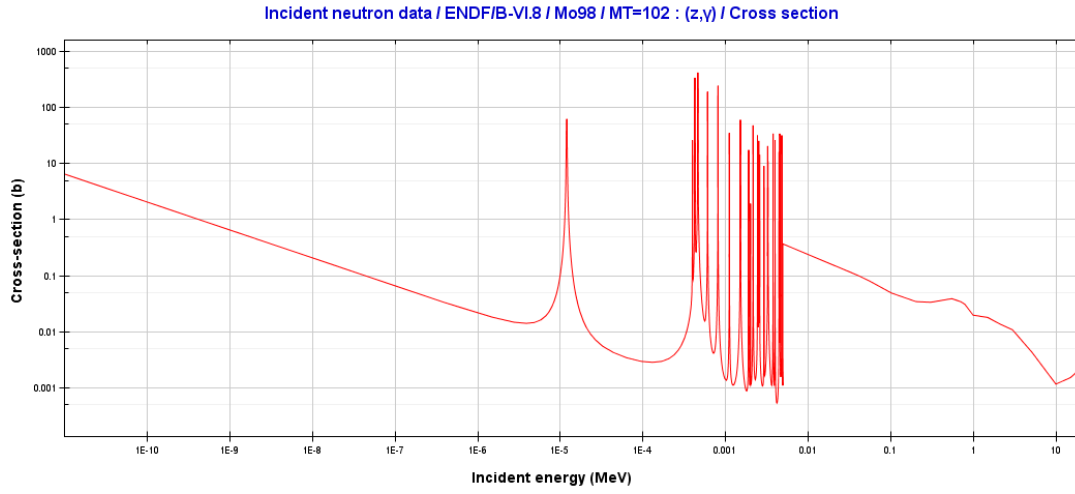


Figure 115: ^{98}Mo capture cross section.

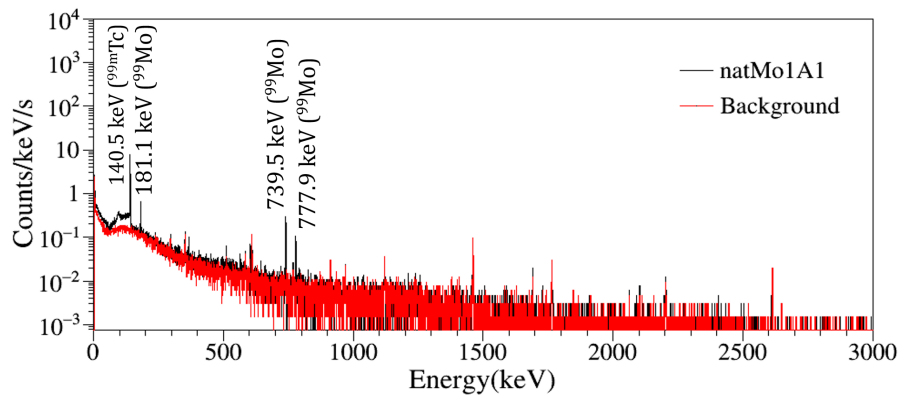


Figure 116: γ -ray spectrum from the irradiated $^{99\text{m}}\text{Tc}$ foil (run09, p_{beam} , $T_{\text{cool}} = 97.9\text{h}$).

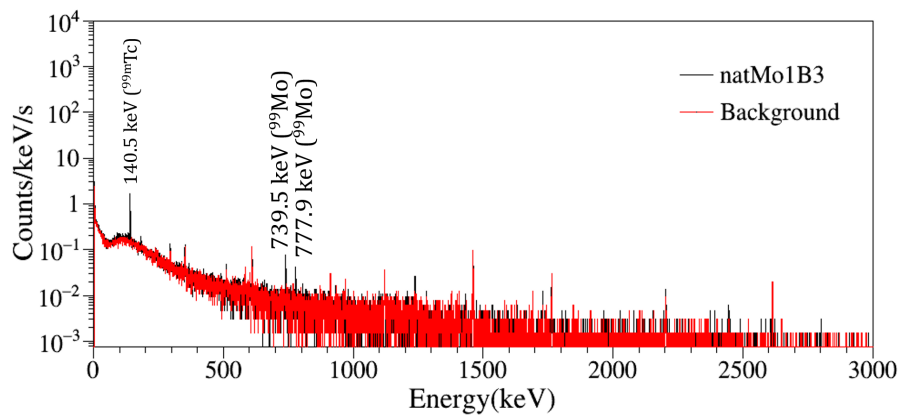


Figure 117: γ -ray spectrum from the irradiated $^{99\text{m}}\text{Tc}$ foil (run12, d_{beam} , $T_{\text{cool}} = 30.9\text{h}$).

Table 40: Experimental and MC Simulation Activities for ^{99}Mo (60 MeV p)

Run #	Thickness (mm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
8	1	A3	23.3	1.64E+03	4.4	2.14E+03	1	1.75E+03	10.6
8	1	B3	27.7	1.50E+03	4.41	1.99E+03	2.17	1.39E+03	11.5
8	1	C3	30.7	1.39E+03	4.42	1.80E+03	1.88	1.27E+03	9.81
9	1	A1	28.1	1.36E+03	4.41	1.74E+03	2.51	1.04E+03	9.95
9	1	A2	16.3	1.92E+03	4.28	2.41E+03	1.72	1.82E+03	10.7
9	1	A4	40.2	1.10E+03	4.45	1.29E+03	5.1	1.04E+03	12.4
9	1	A5	16.9	2.19E+03	4.33	2.73E+03	1.67	2.16E+03	10.7
9	1	A6	36.9	1.16E+03	4.43	1.44E+03	2.44	1.18E+03	11
9	1	A7	23.8	1.71E+03	4.34	2.19E+03	0.69	1.33E+03	11.4
9	1	A8	16.3	2.00E+03	4.31	2.56E+03	2.04	1.67E+03	10.8
9	1	B4	42.9	1.07E+03	4.47	1.25E+03	2.72	9.38E+02	10.2
9	1	B5	22.6	1.67E+03	4.34	2.14E+03	2.21	1.70E+03	11.3
9	1	B6	39.8	1.12E+03	4.48	1.33E+03	1.67	9.40E+02	9.71
9	1	B7	28.2	1.53E+03	4.38	1.92E+03	2.9	1.24E+03	10
9	1	B8	22.1	1.74E+03	4.33	2.09E+03	0.96	1.44E+03	10.9
9	1	C4	44.9	9.84E+02	4.51	1.13E+03	1.32	1.11E+03	10.5
9	1	C5	26.2	1.53E+03	4.36	2.04E+03	1.86	1.32E+03	10
9	1	C7	31.1	1.38E+03	4.39	1.74E+03	2	1.26E+03	9.54
9	0.1	B2	22.1	2.13E+03	5.47				
9	0.1	B3	27.7	1.84E+03	4.47				
9	0.1	-B6	39.8	1.34E+03	4.92				
9	0.1	-B8	22.1	2.13E+03	5.11				
9	0.25	-B2	22.1	1.88E+03	4.52				
9	0.25	-B3	27.7	1.73E+03	4.24				
13	1	-B8	22.1	1.72E+03	4.36	2.09E+03	0.96	1.44E+03	10.9

Table 41: Experimental and MC Simulation Activities for ^{99}Mo (30 MeV d)

Run #	Thickness (mm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
12	1	B2	22.1	5.47E+02	14.6	3.76E+04	0.38	5.06E+02	11.5
12	1	B3	27.7	4.83E+02	14.59	3.75E+04	0.66	5.49E+02	11.9
12	1	C3	30.7	4.45E+02	14.63	3.75E+04	0.67	6.17E+02	10.9

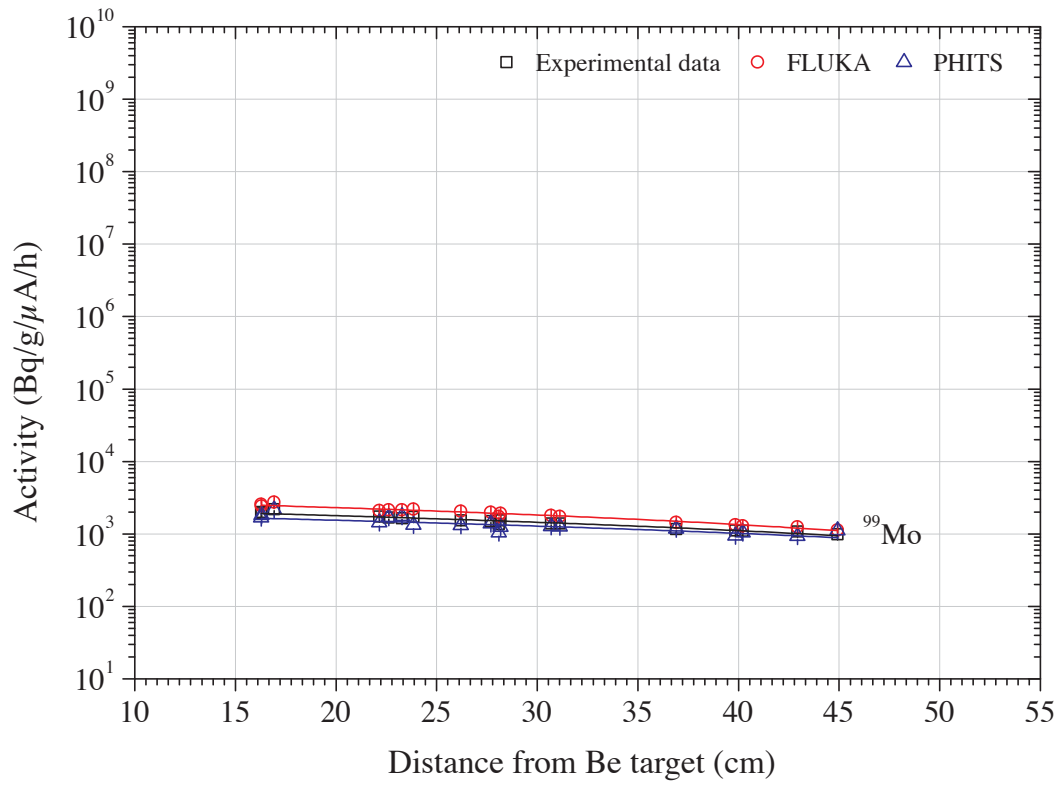


Figure 118: Measured activity for ^{99}Mo with 60 MeV p beam.

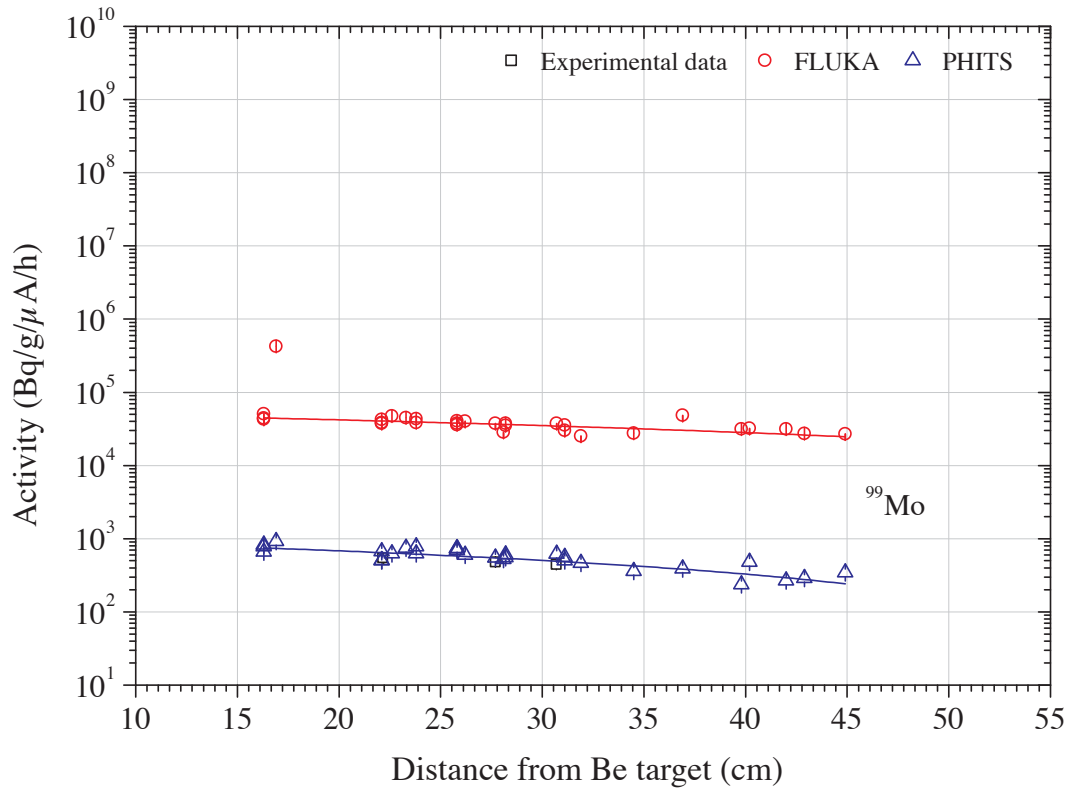


Figure 119: Measured activity for ^{99}Mo with a 30 MeV d beam.

In the previous figures (Figure 118 & Figure 119), in the case of irradiation of ^{nat}Mo with a proton beam, we can see that there is a satisfactory correspondence between the experimental results and the results of the MC simulation. This last was carried out with two MC codes: FLUKA & PHITS - both reproduce well the experimental results. However in the case of deuteron beam, the FLUKA code over estimates the experimental results of approximately 77 times higher on average. Figure 120 shows only the experimental results of the activity for proton and deuteron beams.

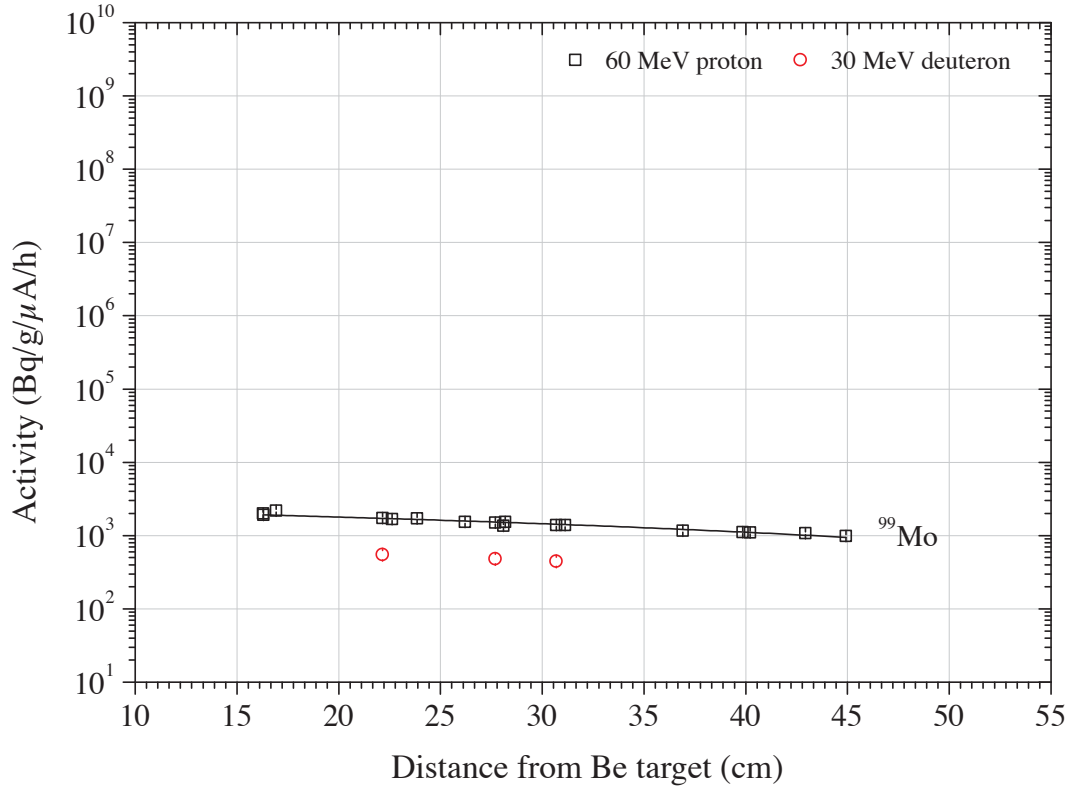


Figure 120: Measured activity for ^{99}Mo with 60 MeV protons & 30 MeV deuterons.

5.7.2 ^{98}Mo

The main purpose was to activate enriched ^{98}Mo metal powder (99.9%) inside of the activator using 60 MeV proton and 30 MeV deuteron beams.

The samples were irradiated for 1 hour. The average proton beam current of run13 and run14 was $1.01\ \mu\text{A}$. The average deuteron beam current of run18 and run19 was $0.83\ \mu\text{A}$ and $0.72\ \mu\text{A}$ respectively.

^{98}Mo samples were placed and irradiated in different positions inside of the Activator as is shown in Figure 121. The positions of the samples for both proton and deuteron beams were the same. Table 42 and Table 43 show the sample positions inside of the Activator for p & d beams respectively.

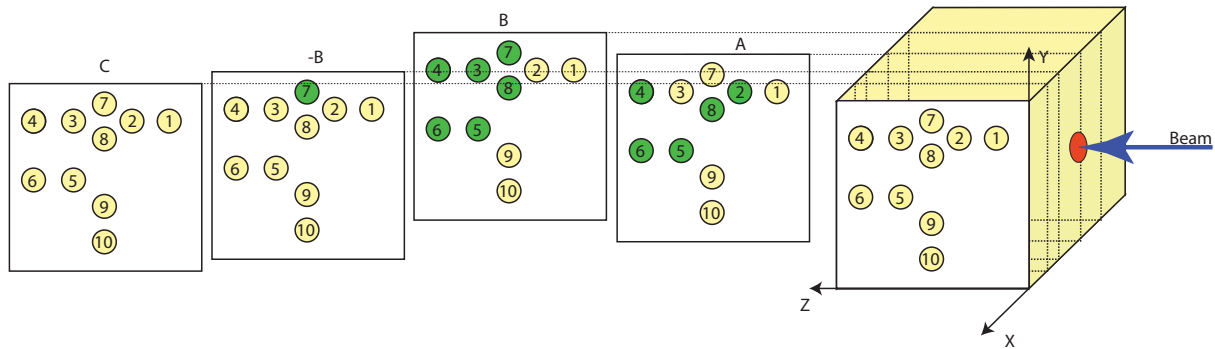


Figure 121: ^{98}Mo sample positions inside of the Activator.

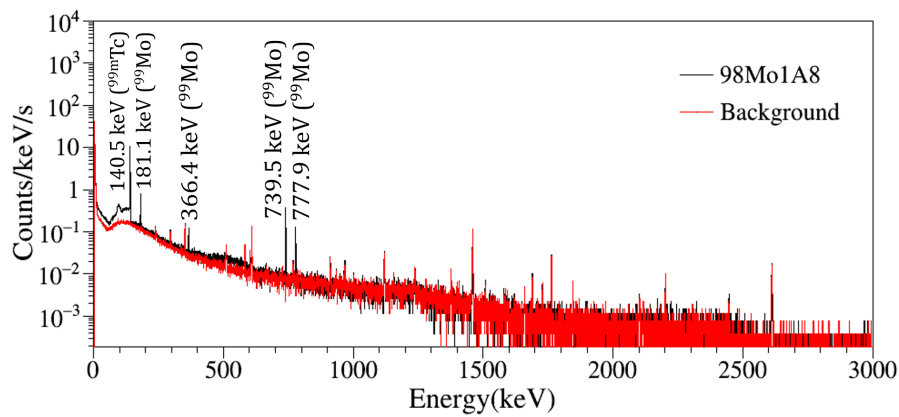
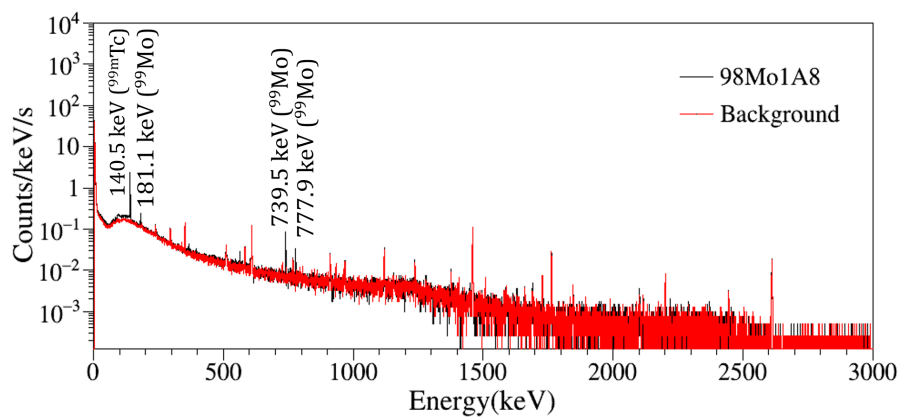
^{99}Mo ($T_{1/2} = 65.976\ \text{h}$) and $^{99\text{m}}\text{Tc}$ ($T_{1/2} = 6.0067\ \text{h}$) were identified in the gamma-ray spectra (Figure 122 & Figure 123). ^{99}Mo is produced via $^{98}\text{Mo}(n, \gamma)^{99}\text{Mo}$ neutron capture reaction. $^{99\text{m}}\text{Tc}$ is the decay product of ^{99}Mo (β^- decay, 87.89 %) (Chadwick et al., 2011).

Table 42: ^{98}Mo sample positions inside of the Activator for all runs

Run	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
13	1	A2	16.3	0.402
13	1	A4	40.2	0.401
13	1	A5	16.9	0.401
13	1	A6	36.9	0.399
13	1	A8	16.3	0.401
13	1	B3	27.7	0.404
13	1	B4	42.9	0.4
13	1	B5	22.6	0.399
13	1	B6	39.8	0.4
13	1	B7	28.2	0.403
13	1	B8	22.1	0.397
13	0.5	-B7	28.2	0.194
14	0.1	B7	28.2	0.044
14	0.25	-B7	28.2	0.113
16	1	B7	28.2	0.399

Table 43: ^{98}Mo sample positions inside of the Activator for all runs

Run	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
18	1	A2	16.3	0.402
18	1	A4	40.2	0.401
18	1	A5	16.9	0.401
18	1	A6	36.9	0.399
18	1	A8	16.3	0.401
18	1	B3	27.7	0.404
18	1	B4	42.9	0.4
18	1	B5	22.6	0.399
18	1	B6	39.8	0.4
18	1	B7	28.2	0.403
18	1	B8	22.1	0.397
18	0.5	-B7	28.2	0.194
19	0.25	-B7	28.2	0.113
19	0.1	B7	28.2	0.044

Figure 122: γ -ray spectrum from the irradiated ^{98}Mo (run13, p_{beam} , $T_{\text{cool}} = 44.4\text{h}$).Figure 123: γ -ray spectrum from the irradiated ^{98}Mo foil (run18, d_{beam} , $T_{\text{cool}} = 98.6\text{ h}$).

The main peak used for the activity measurement was: $E_{\gamma} = 739.5\text{ KeV}$, $I_{\gamma} = 12.26\%$ (Table 44 & Table 45; Figure 124, Figure 126, Figure 127 & Figure 129).

Table 44: Experimental and MC Simulation Activities for ^{99}Mo (60 MeV p)

Run #	Thickness (mm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
13	1	A2	16.3	7.40E+03	4.35	9510	1.78	5040	7.12
13	1	A4	40.2	4.64E+03	4.4	5150	3.76	2780	8.81
13	1	A5	16.9	7.64E+03	4.31	10000	1.35	5540	7.54
13	1	A6	36.9	4.73E+03	4.38	5810	1.63	3150	8.77
13	1	A8	16.3	7.69E+03	4.27	9920	1.99	5200	7.9
13	1	B3	27.7	5.97E+03	4.3	7690	1.74	3950	7.26
13	1	B4	42.9	4.28E+03	4.26	5120	2.19	2790	6.91
13	1	B5	22.6	6.83E+03	4.31	8380	2.89	4610	6.9
13	1	B6	39.8	4.69E+03	4.36	5430	1.07	2880	7.99
13	1	B7	28.2	6.33E+03	4.31	7660	2.06	4070	6.42
13	1	B8	22.1	6.98E+03	4.42	8360	0.9	4270	7.61
13	0.5	-B7	28.2	6.93E+03	4.3				
14	0.1	B7	28.2	6.97E+03	5.09				
14	0.25	-B7	28.2	7.29E+03	4.45				
16	1	B7	28.2	6.46E+03	4.44				

Table 45: Experimental and MC Simulation Activities for ^{99}Mo (30 MeV d)

Run #	Thickness (mm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
18	1	A2	16.3	3.47E+03	6.79	167000	0.18	2130	9.65
18	1	A4	40.2	1.85E+03	7.33	125000	0.38	1370	10.6
18	1	A5	16.9	3.28E+03	6.64	261000	0.51	2140	8.28
18	1	A6	36.9	2.02E+03	7.56	144000	0.33	1330	10.3
18	1	A8	16.3	3.60E+03	7.21	194000	0.25	2250	9.7
18	1	B3	27.7	2.64E+03	7.01	149000	0.4	1770	7.5
18	1	B4	42.9	1.74E+03	6.8	111000	0.26	982	9.74
18	1	B5	22.6	2.86E+03	7.08	177000	0.23	2020	9.11
18	1	B6	39.8	1.96E+03	7.59	126000	0.29	944	9.37
18	1	B7	28.2	2.89E+03	7.44	150000	0.19	1840	8.6
18	1	B8	22.1			1.65E+05	0.2	1.92E+03	7.39
18	0.5	-B7	28.2	2.97E+03	7.06				
19	0.25	-B7	28.2	3.09E+03	8.44				
19	0.1	B7	28.2	3.01E+03	10.75				

The results presented in the above tables confirm the results obtained with ^{99}Mo , that is the FLUKA overestimate by a factor almost 80 the induced activity with 30 MeV deuterons.

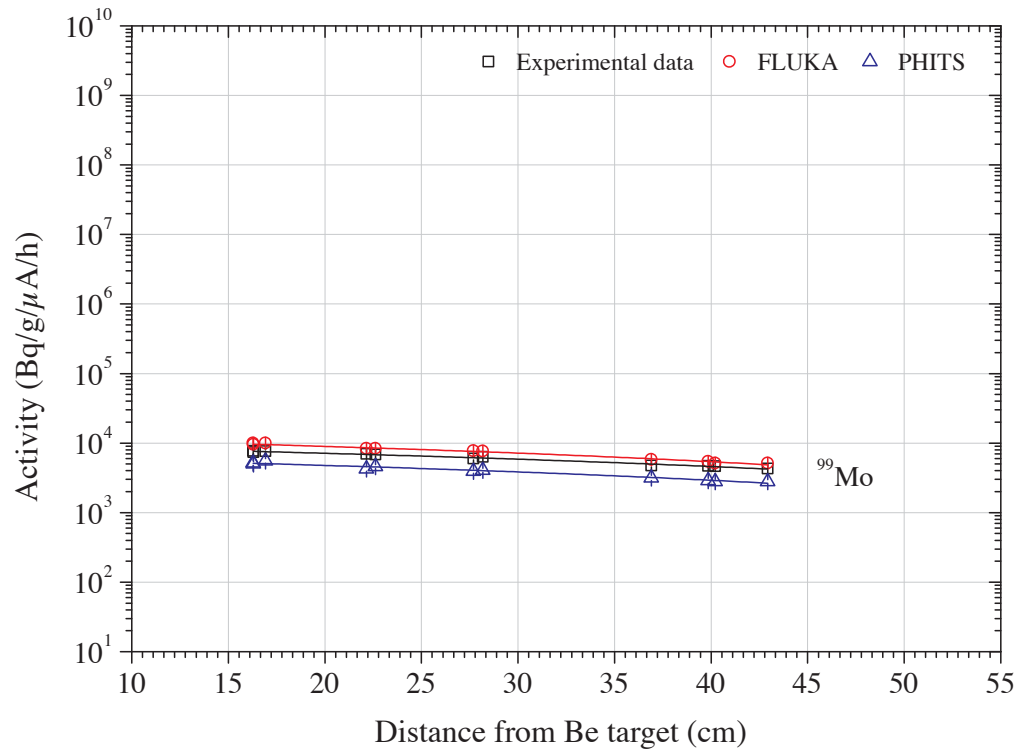


Figure 124: Measured activity for ^{99}Mo (60 MeV p).

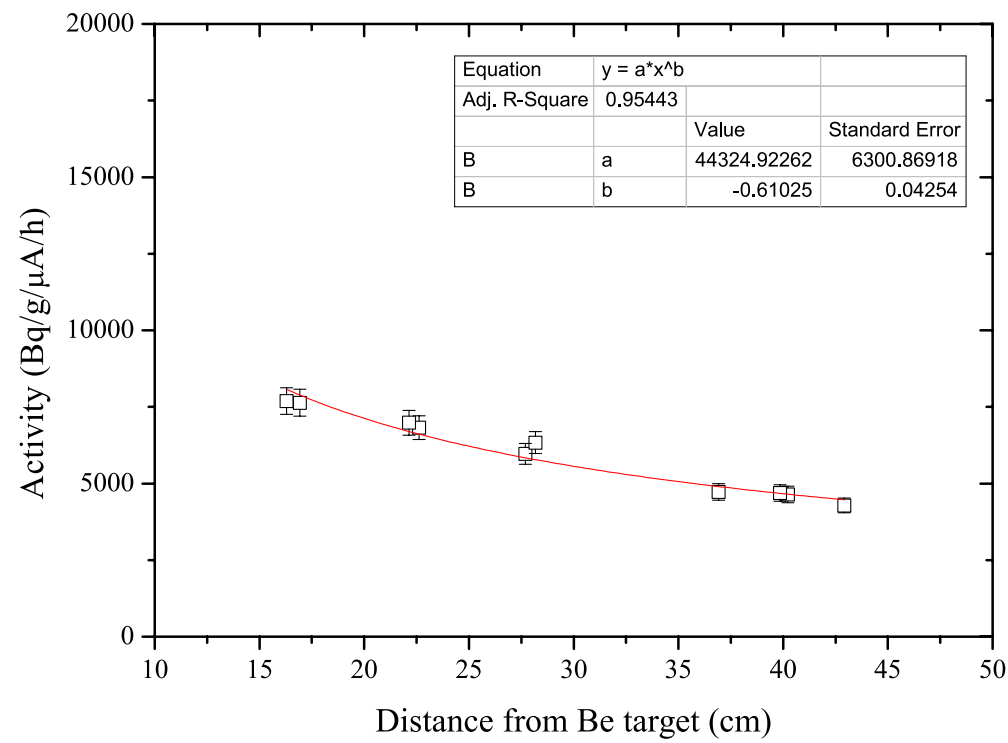


Figure 125: Measured activity (fitting curve) for ^{99}Mo (60 MeV p).

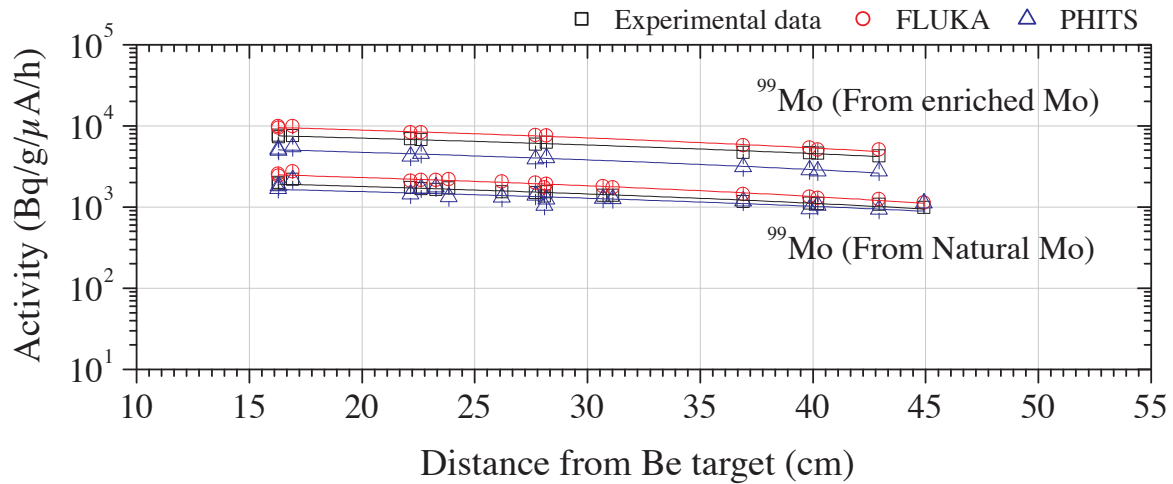


Figure 126: Measured activity for ^{99}Mo & $^{\text{Nat}}\text{Mo}$ (60 MV p).

If one corrects for the concentrations of ^{98}Mo in $^{\text{Nat}}\text{Mo}$ sample (that is only 24%) then the specific activity in Bq/gr of ^{98}Mo is the same in both cases. This indicates that ^{98}Mo has the same capture probability in both materials. This should be the case since $^{\text{Nat}}\text{Mo}$, the capture resonance of ^{100}Mo is shielding the one of ^{98}Mo for neutrons in the energy region close to 500 eV and therefore we should be getting much less captures in ^{98}Mo when mixed with ^{100}Mo . The fact that this is not happening means that the neutron in the diffuser have a much higher energies and the captures in ^{98}Mo are taking place at a much higher energy that is well above 1 KeV.

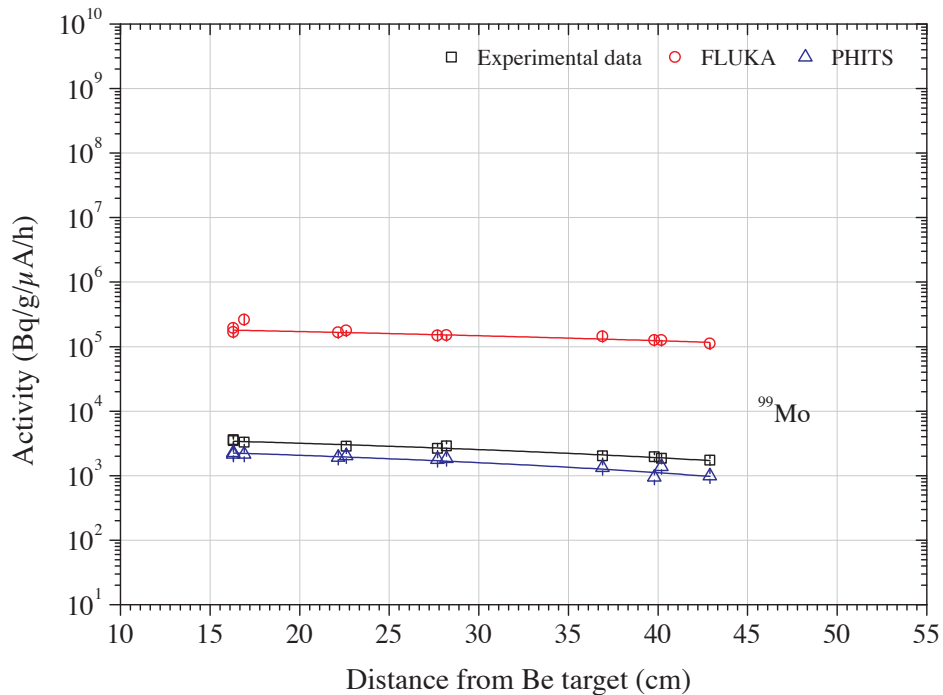


Figure 127: Measured activity for ^{99}Mo (30 MeV d).

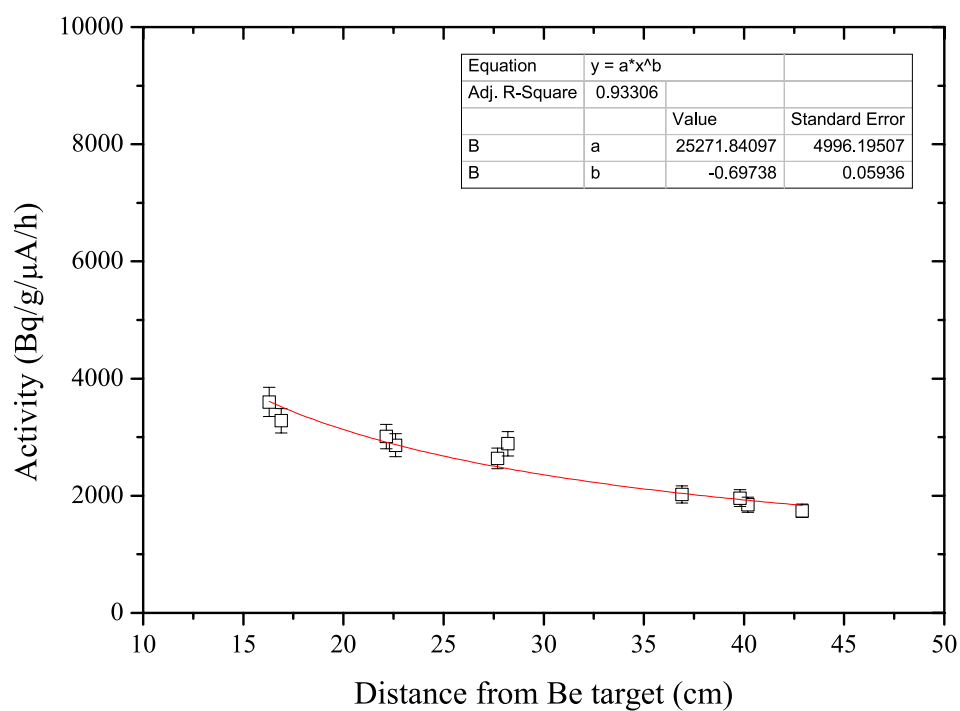


Figure 128: Measured activity (fitting curve) for ^{99}Mo (30 MeV d).

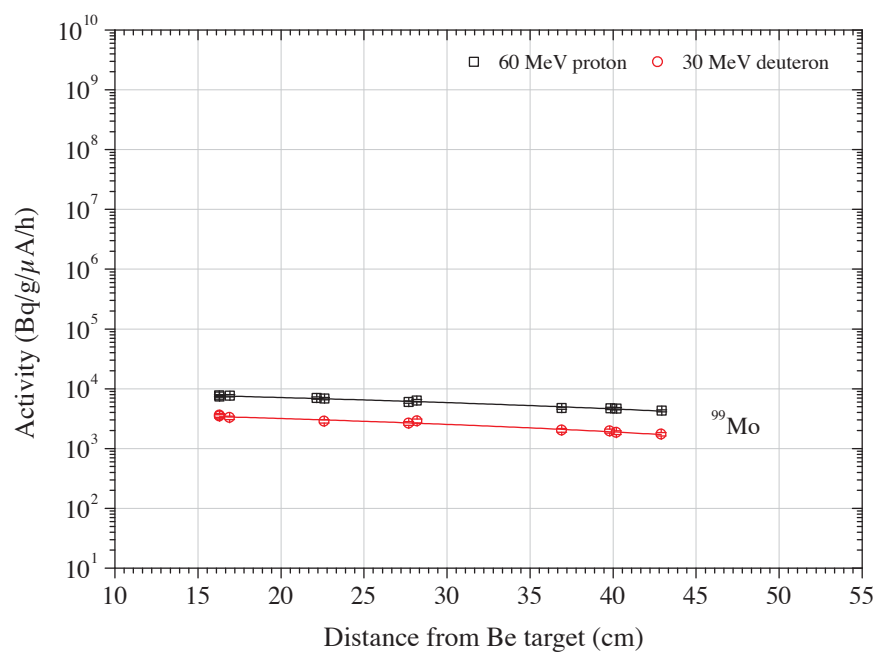


Figure 129: Measured activity for ^{99}Mo with 60 MeV protons & 30 MeV deuterons.

5.7.3 ^{nat}Lu

^{nat}Lu has two stable isotopes ^{175}Lu (97.41%) and ^{176}Lu (2.59%). ^{177}Lu ($T_{1/2} = 6.647$ d) was identified in the gamma-ray spectra. ^{177}Lu comes from $^{176}\text{Lu}(n, \gamma)^{177}\text{Lu}$ reaction (Figure 130).

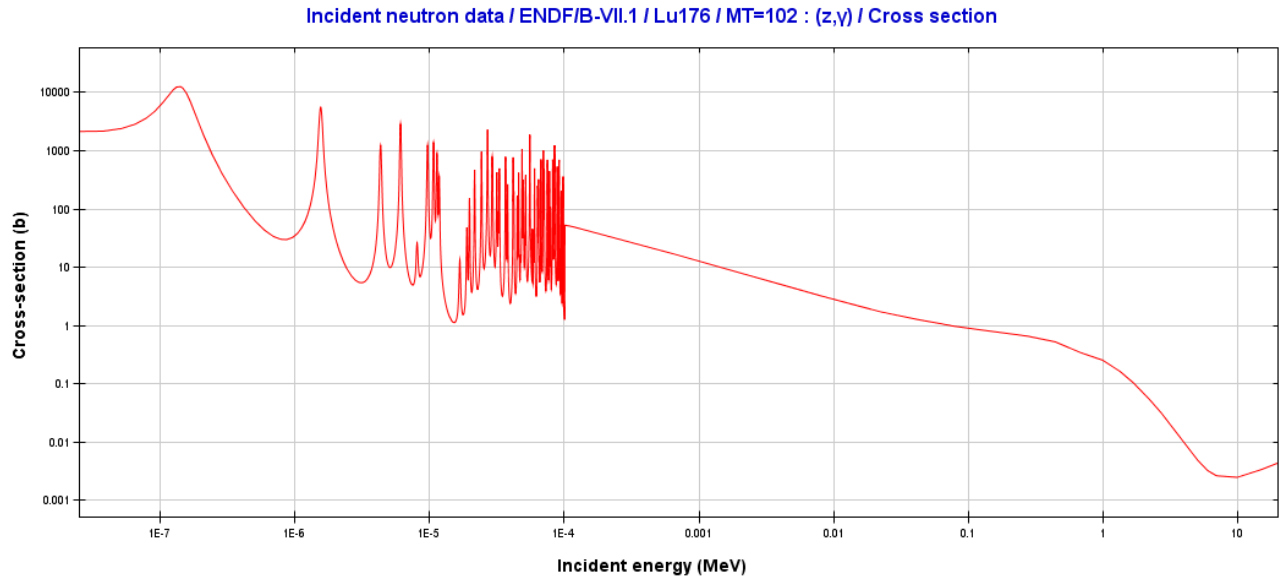


Figure 130: $^{176}\text{Lu}(n, \gamma)$ cross section reaction

^{nat}Lu samples were placed and irradiated in different positions inside of the Activator (Figure 131 & Figure 132) and (Table 46 & Table 47).

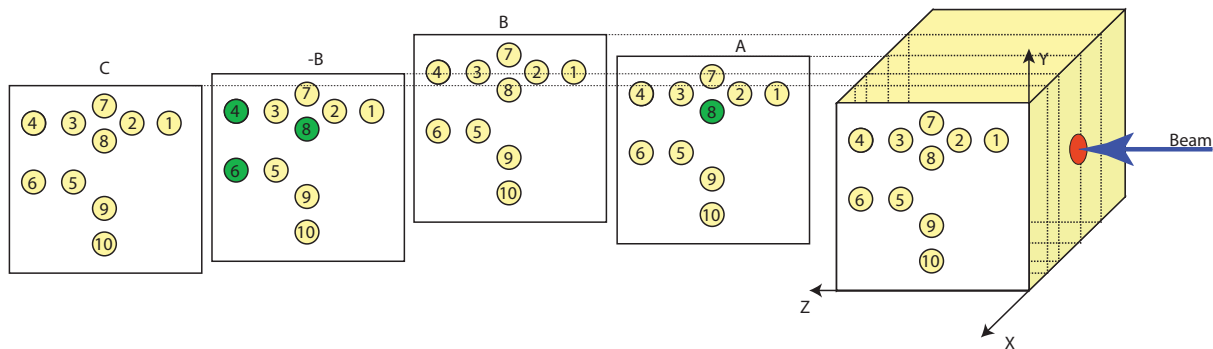


Figure 131: ^{nat}Lu sample positions inside of the Activator (60 MeV p).

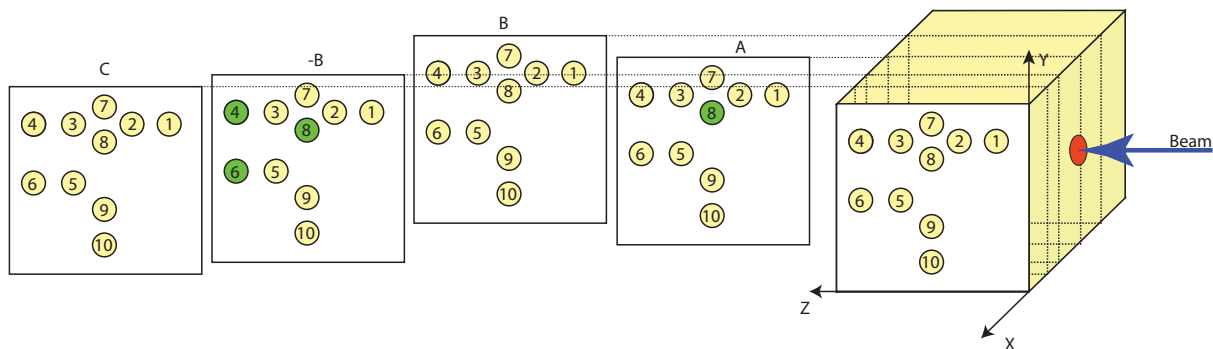


Figure 132: ^{nat}Lu sample positions inside of the Activator (30 MeV d).

Table 46: ^{Nat}Lu sample positions inside of the Activator (60 MeV p)

Run	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
14	1	A8	16.3	0.708
14	1	-B4	42.9	0.744
14	1	-B6	39.8	0.658
14	1	-B8	22.1	0.736

Table 47: ^{Nat}Lu sample positions inside of the Activator (30 MeV d)

Run	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
19	1	A8	16.3	0.758
19	1	-B4	42.9	0.76
19	1	-B6	39.8	0.764
19	1	-B8	22.1	0.667

One main peak was used for calculation ($E_\gamma = 208.4 \text{ KeV}$, $I_\gamma = 10.36 \%$).

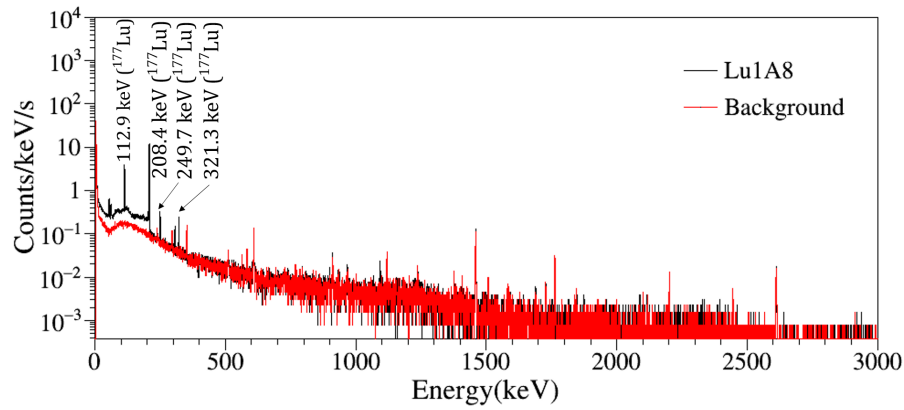
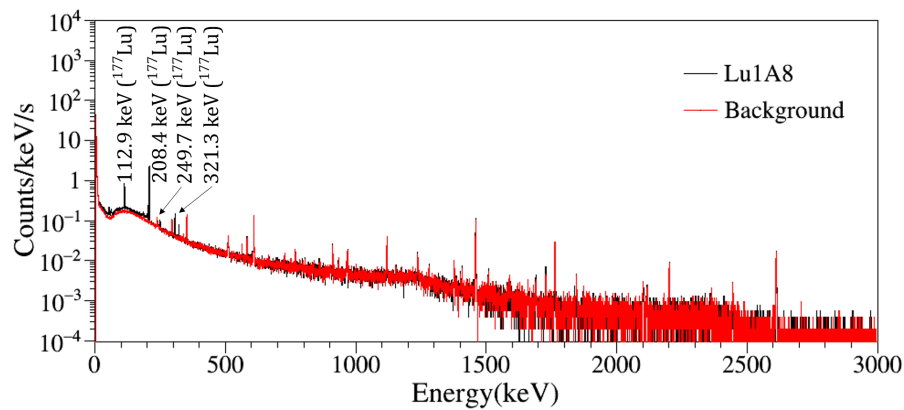
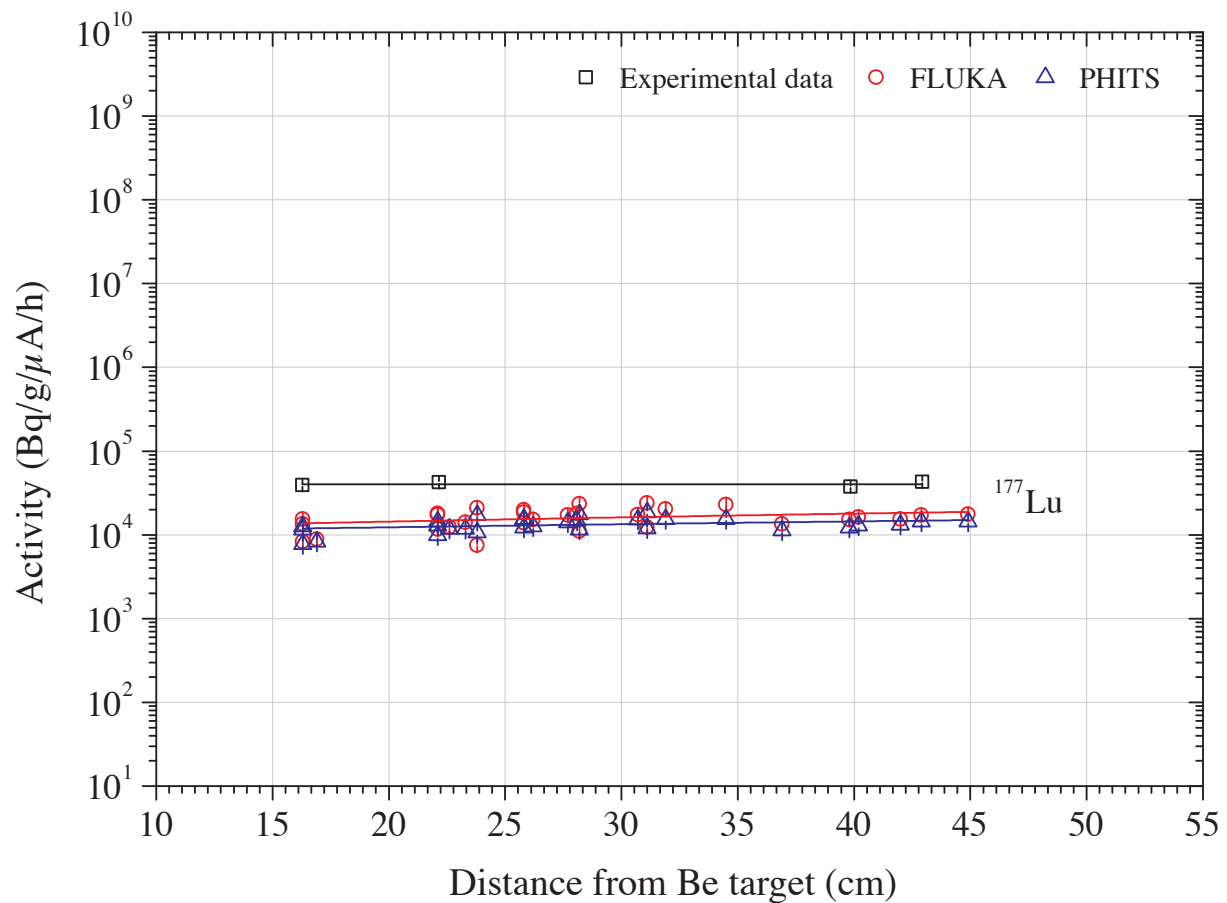
Figure 133: γ -ray spectrum from the irradiated ^{Nat}Lu foil (run14, p_{beam} , $T_{\text{cool}} = 52.0\text{h}$).Figure 134: γ -ray spectrum from the irradiated ^{Nat}Lu foil (run19, d_{beam} , $T_{\text{cool}} = 187.5 \text{ h}$).

Table 48: Experimental and MC Simulation Activities for ^{177}Lu (60 MeV p)

Run #	Thickness (mm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
14	1	A8	16.3	3.91E+04	3.78	1.34E+04	3.68	1.15E+04	6.29
14	1	-B4	42.9	4.30E+04	3.78	1.71E+04	2.31	1.43E+04	6.76
14	1	-B6	39.8	3.76E+04	3.78	1.51E+04	4.18	1.21E+04	6.06
14	1	-B8	22.1	4.23E+04	3.78	1.73E+04	1.73	1.29E+04	6.68

Table 49: Experimental and MC Simulation Activities for ^{177}Lu (30 MeV d)

Run #	Thickness (mm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
19	1	A8	16.3	1.56E+04	7.48	2.58E+05	0.172	4.75E+03	7.82
19	1	-B4	42.9	1.75E+04	7.46	4.24E+05	0.234	5.66E+03	6.45
19	1	-B6	39.8	1.49E+04	7.47	3.61E+05	0.123	4.82E+03	11.6
19	1	-B8	22.1	1.83E+04	7.47	3.38E+05	0.18	6.05E+03	9.14

Figure 135: Measured activity for ^{177}Lu for 60 MeV p.

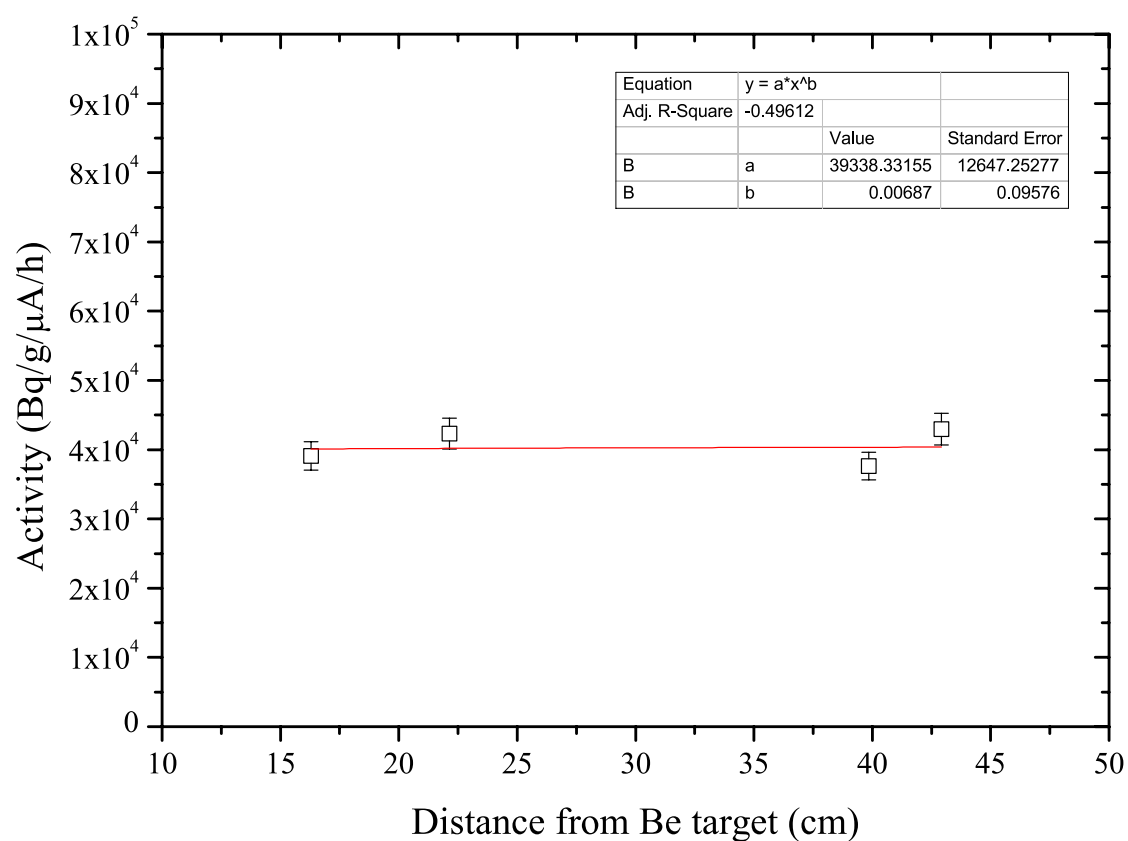


Figure 136: Measured activity (fitting curve) for ^{177}Lu for 60 MeV p.

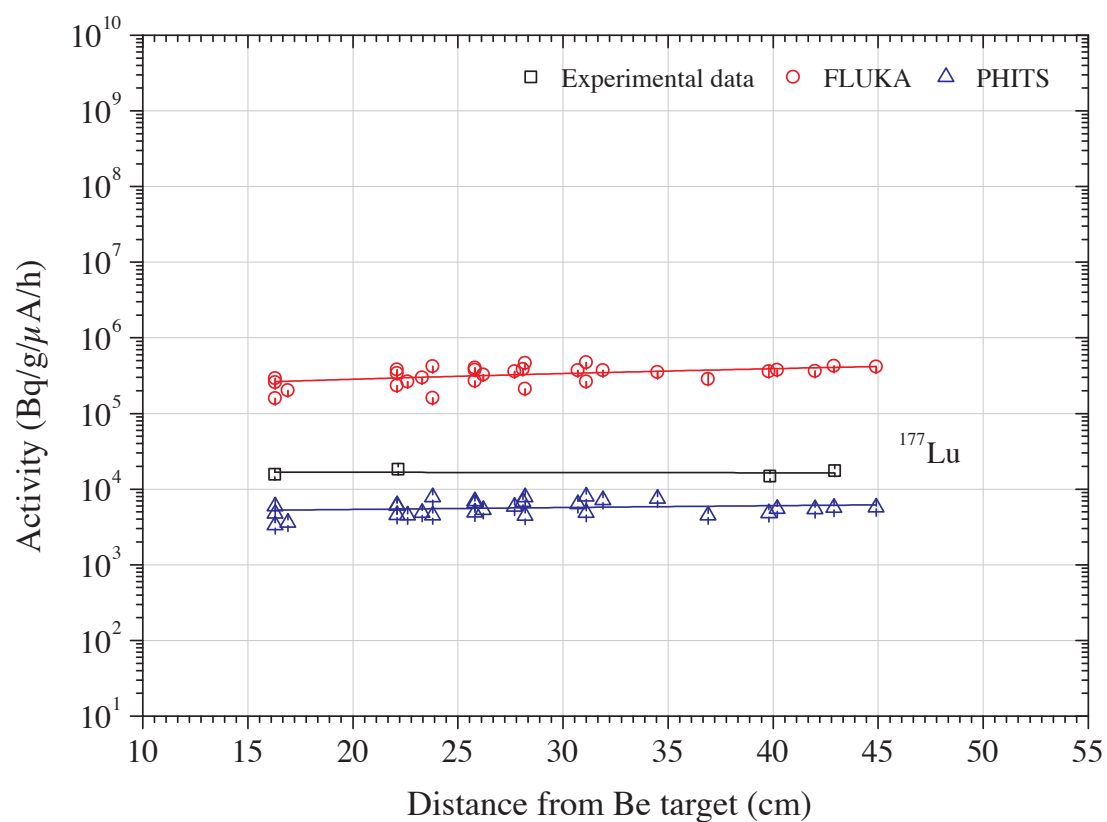


Figure 137: Measured activity for ^{177}Lu for 30 MeV d beams.

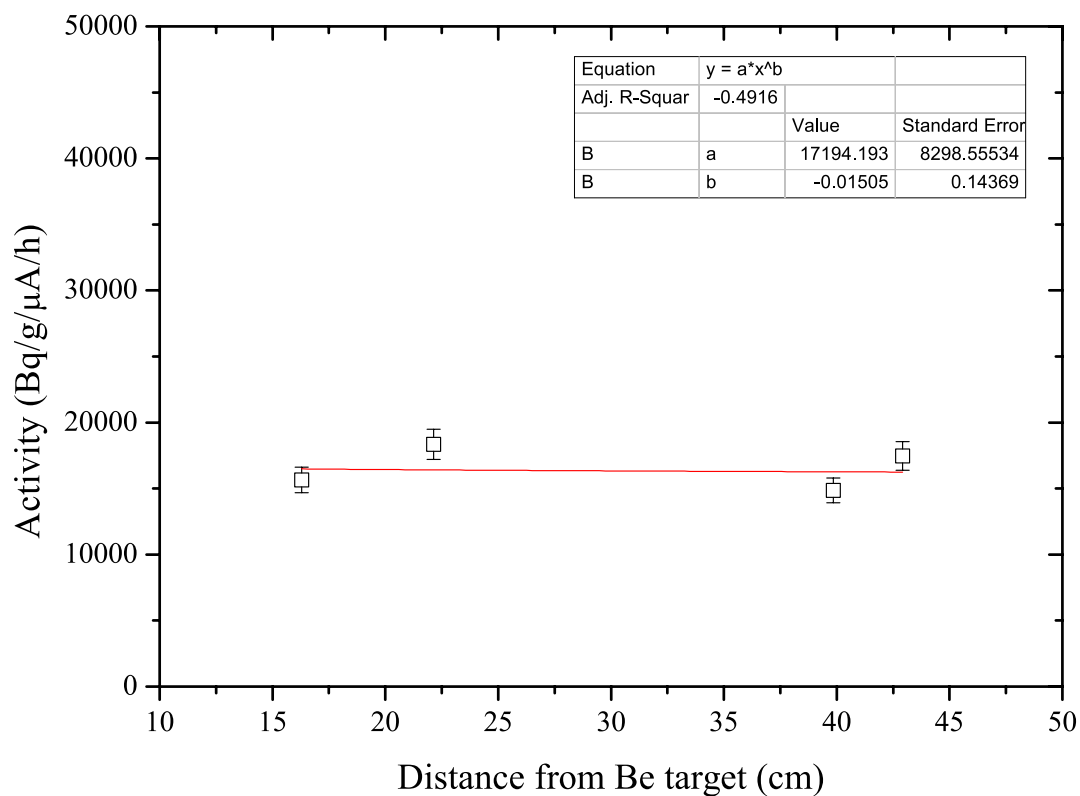


Figure 138: Measured activity (fitting curve) for ^{177}Lu for 30 MeV d beams.

The activity increases with distance, meaning that in the case of Lu, are more sensitive to thermal neutrons, which are obviously in the periphery of the diffuser lead or in other words, near the reflector.

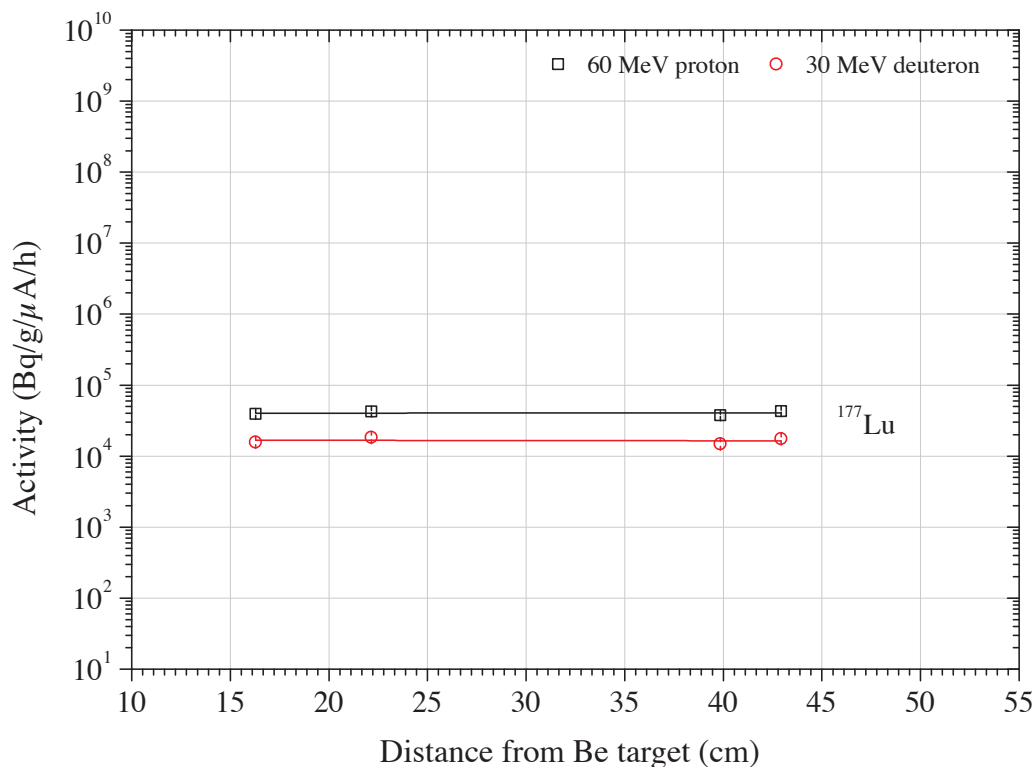


Figure 139: Measured activity for ^{177}Lu with 60 MeV protons & 30 MeV deuterons.

5.7.4 ^{Nat}Re

^{Nat}Re has two stable isotopes ¹⁸⁵Re (37.4%) and ¹⁸⁷Re (62.6%). ¹⁸⁶Re comes from ¹⁸⁵Re(n, γ)¹⁸⁶Re reaction and ¹⁸⁸Re comes from ¹⁸⁷Re(n, γ)¹⁸⁸Re reaction.

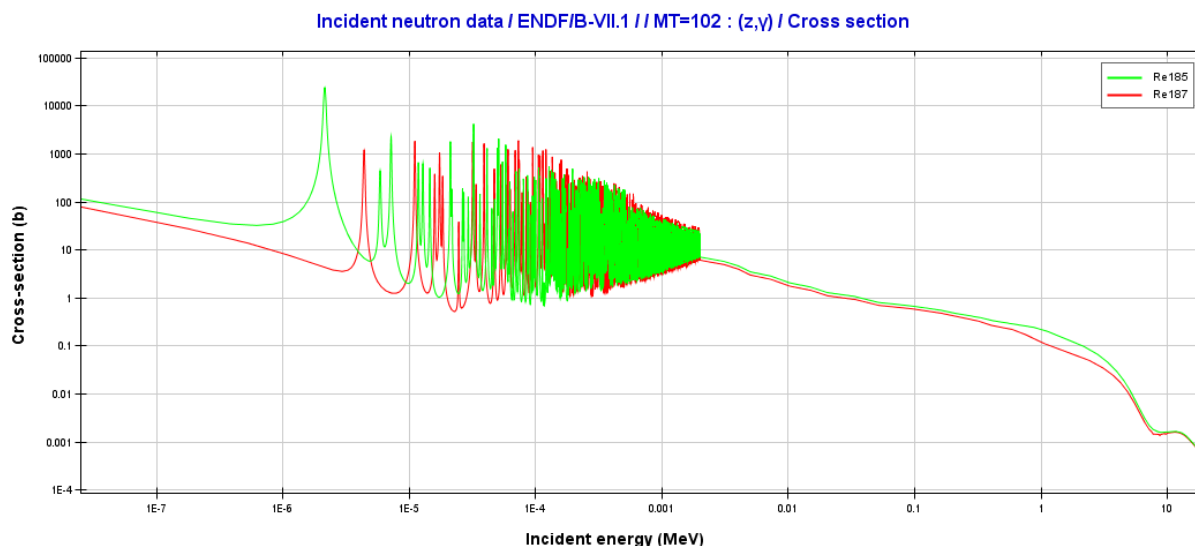


Figure 140: ¹⁸⁷Re(n, γ) capture cross section.

^{Nat}Re samples were placed and irradiated either for proton and deuteron beams, only in four positions within the Activator (Figure 141 & Table 50)

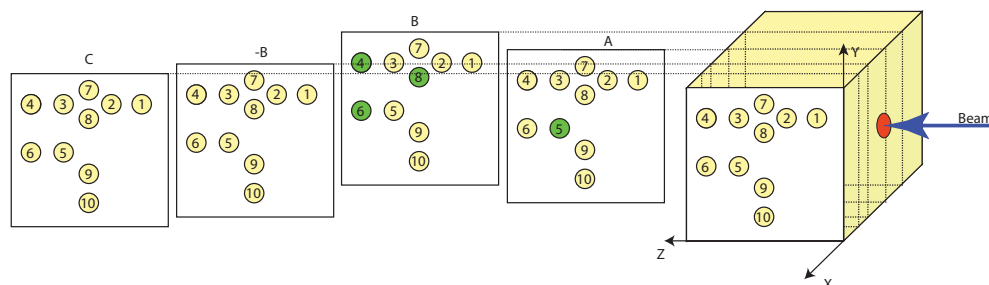


Figure 141: ^{Nat}Re sample positions inside of the Activator (60 MeV p & 30 MeV d).

Table 50: ^{Nat}Re sample positions inside of the Activator

	Run	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
p beam	14	1	A5	16.9	1.476
	14	1	B4	42.9	1.424
	14	1	B6	39.8	1.472
	14	1	B8	22.1	1.424
d beam	19	1	A5	16.9	1.476
	19	1	B4	42.9	1.415
	19	1	B6	39.8	1.725
	19	1	B8	22.1	1.195

¹⁸⁶Re ($T_{1/2}$ = 3.7186 d) and ¹⁸⁸Re ($T_{1/2}$ = 17.003 h) were identified in the γ -ray spectrum (Figure 142 & Figure 143). The peak used for activity calculation of ¹⁸⁸Re was at E_γ = 477.99 KeV, I_γ = 1.08 %. In this report, only ¹⁸⁸Re was analyzed (Table 51).

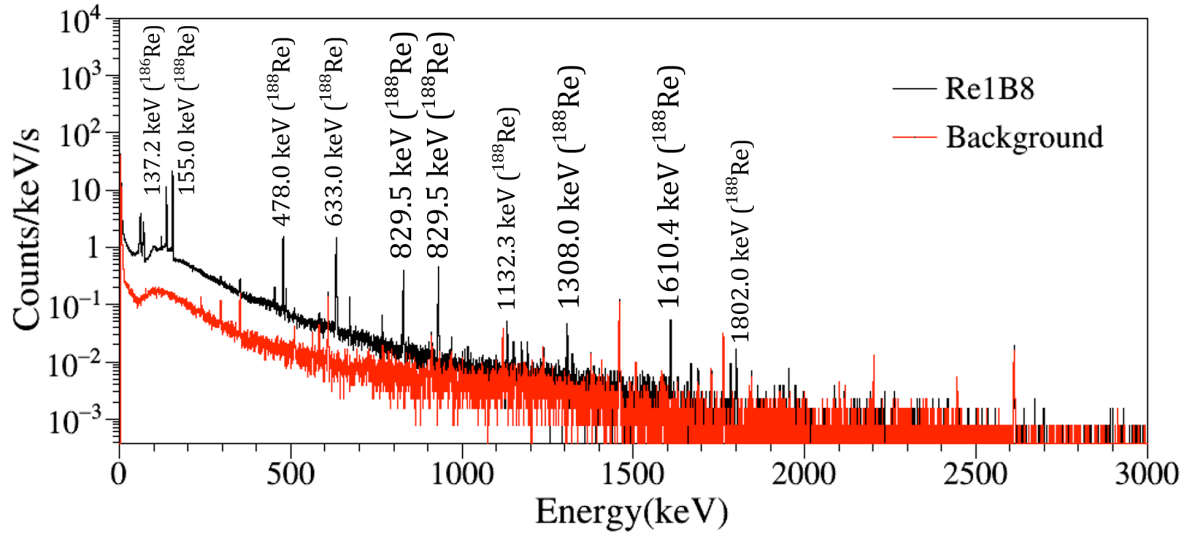


Figure 142: γ -ray spectrum from the irradiated ^{186}Re foil (run14, p_{beam} , $T_{\text{cool}} = 48.8\text{h}$).

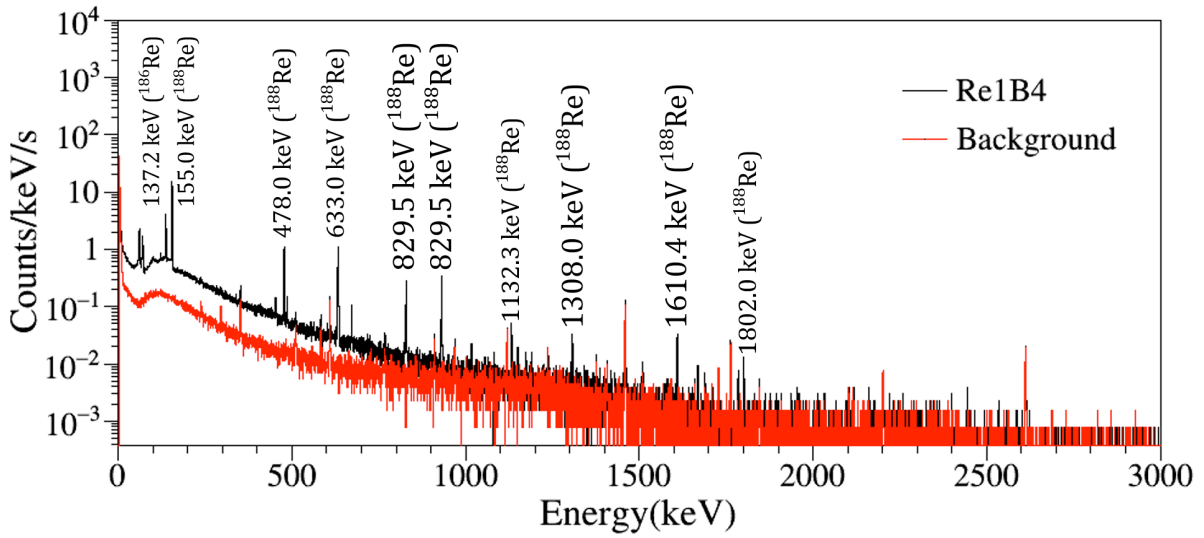


Figure 143: γ -ray spectrum from the irradiated ^{186}Re foil (run19, d_{beam} , $T_{\text{cool}} = 25.4\text{h}$).

Table 51: Experimental and MC Simulation Activities for ^{188}Re

	Run #	Thickness (mm)	Position	d (cm)	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
p beam	14	1	A5	16.9	2.38E+05	4.07	2.16E+05	0.86	1.42E+05	5.85
	14	1	B4	42.9	2.45E+05	4.06	1.99E+05	1.47	1.43E+05	4.72
	14	1	B6	39.8	2.27E+05	4.07	1.89E+05	1.09	1.34E+05	5.26
	14	1	B8	22.1	2.85E+05	4.04	2.56E+05	0.83	1.60E+05	6.08
d beam	19	1	A5	16.9	9.85E+04	7.51	4.51E+06	0.09	6.14E+04	6.01
	19	1	B4	42.9	1.11E+05	7.69	4.70E+06	0.24	5.52E+04	5.02
	19	1	B6	39.8	9.14E+04	7.67	4.54E+06	0.12	5.33E+04	9.3
	19	1	B8	22.1	1.27E+05	7.69	5.09E+06	0.16	7.52E+04	7.06

¹⁸⁶Re results are in progress because it requires a self-absorption corrections.

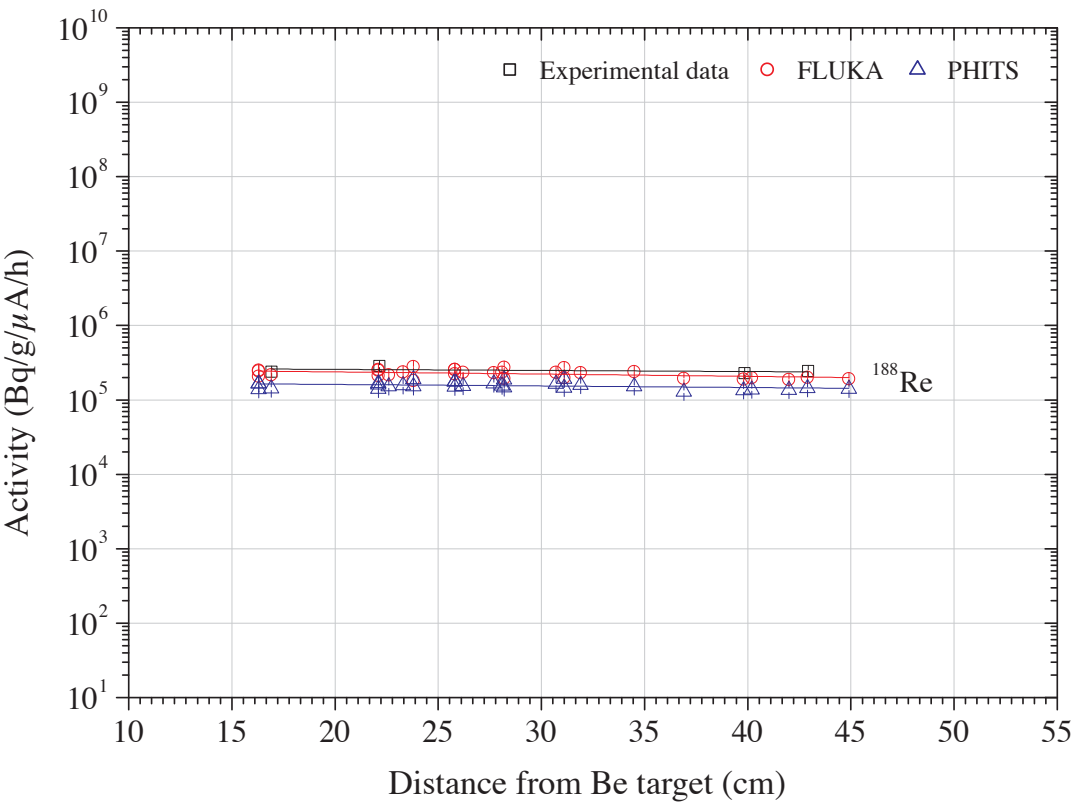


Figure 144: Induced activity for ¹⁸⁸Re with a 60 MeV proton beam.

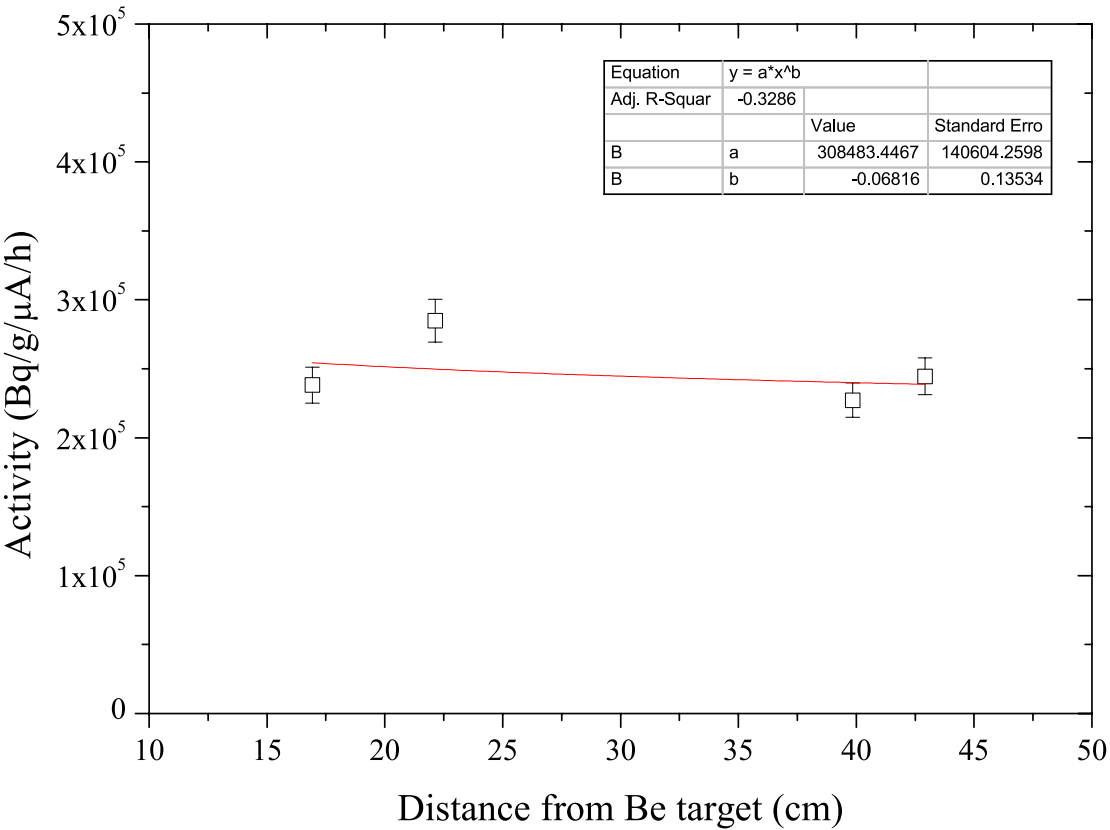


Figure 145: Induced activity (fitting curve) for ¹⁸⁸Re with a 60 meV proton beam.

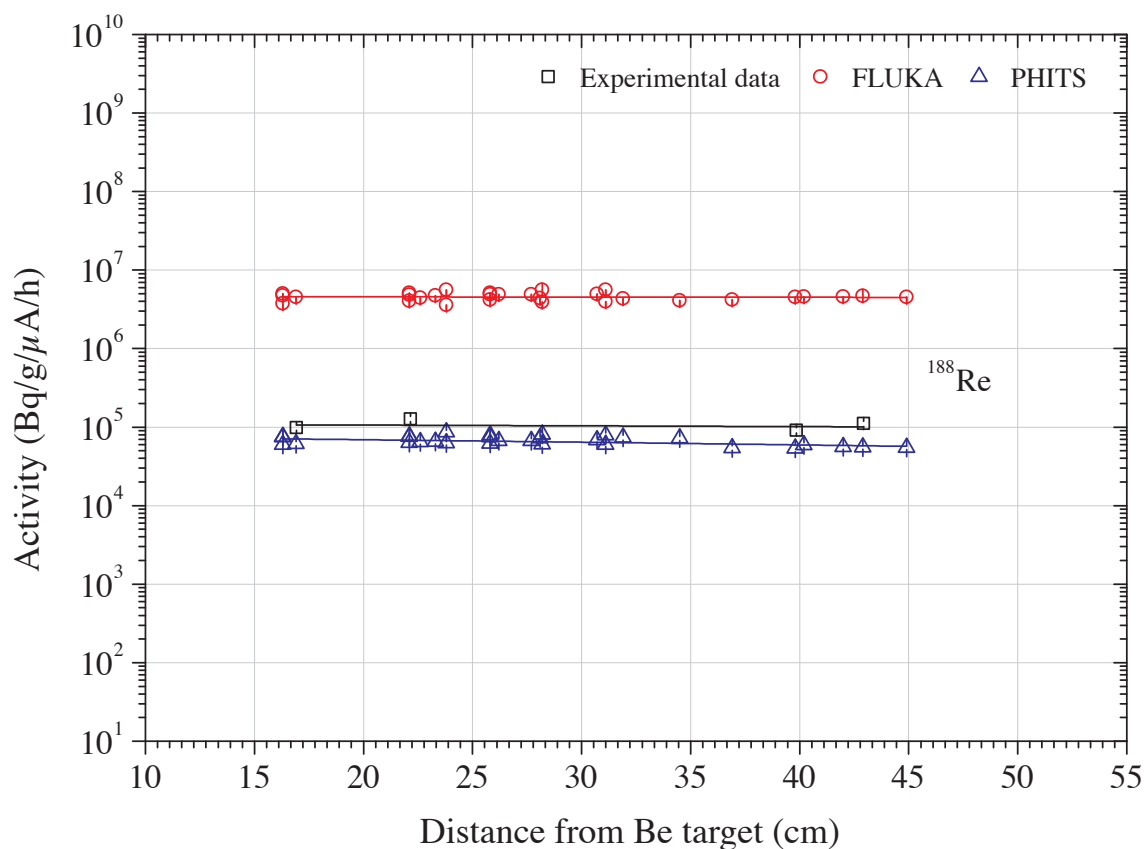


Figure 146: Induced activity for ^{188}Re with a 30 MeV deuteron beam.

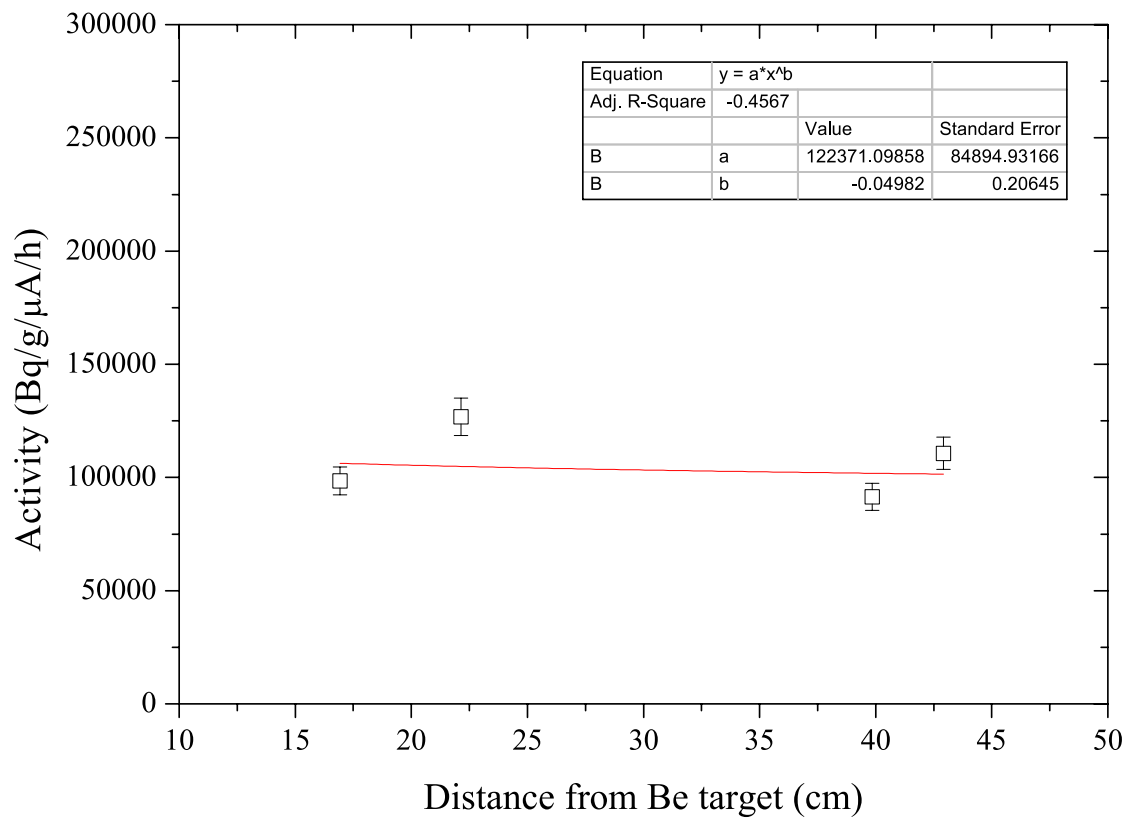


Figure 147: Induced activity (fitting curve) for ^{188}Re with a 30 MeV deuteron beam.

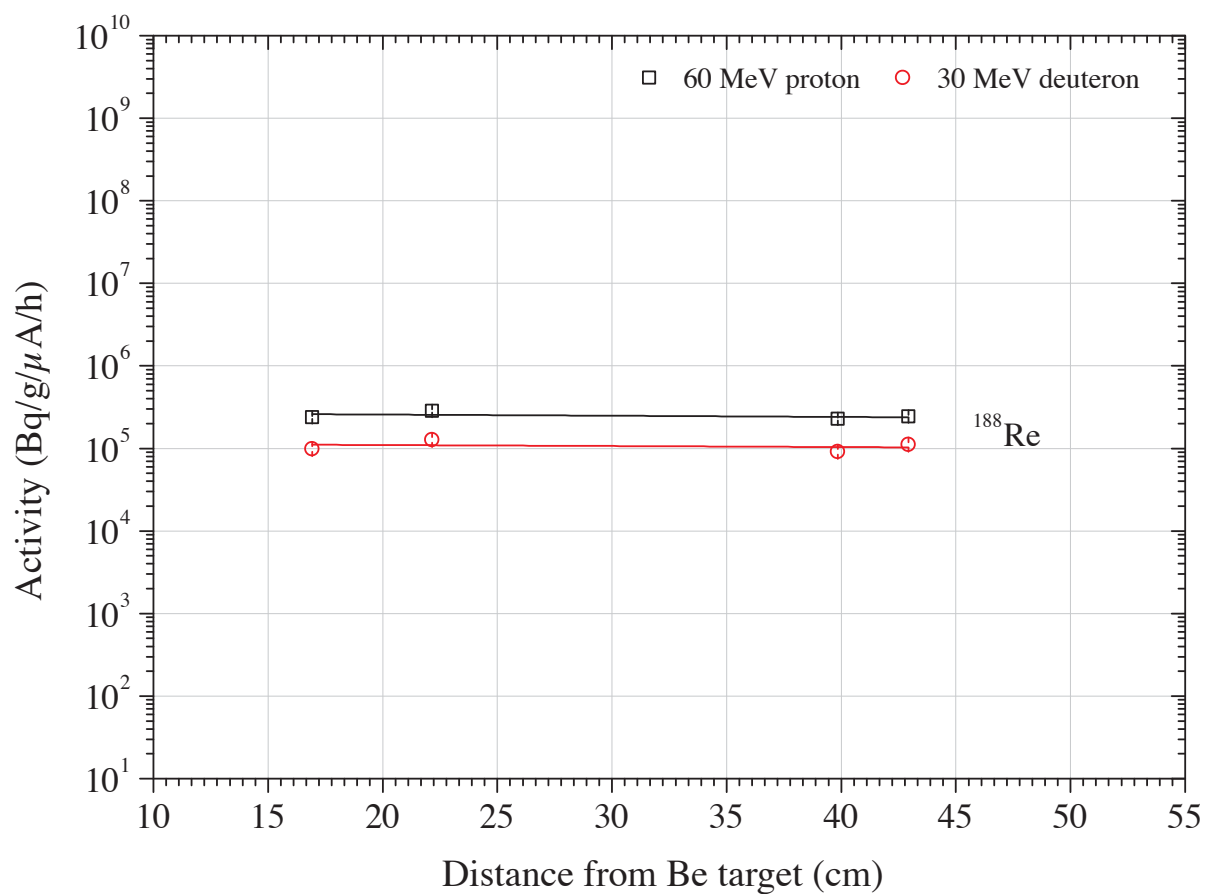


Figure 148: Measured activity for ^{188}Re with 60 MeV protons & 30 MeV deuterons.

5.7.5 ^{165}Ho

^{165}Ho has only one stable isotope ^{165}Ho . ^{166}Ho is produced by $^{165}\text{Ho}(n, \gamma)^{166}\text{Ho}$ reaction (Figure 149).

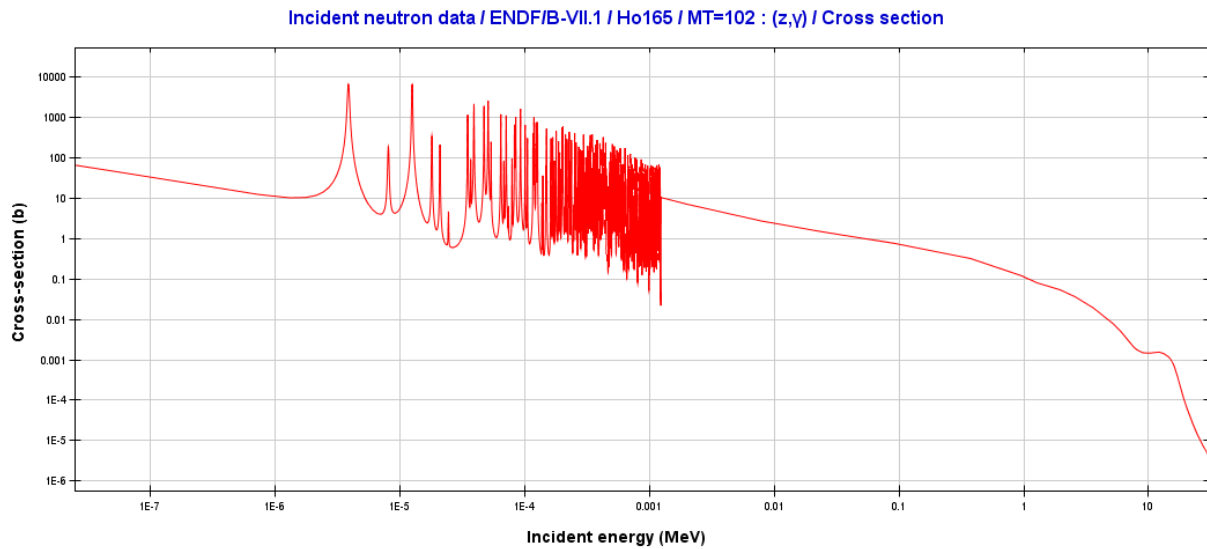


Figure 149: ^{165}Ho cross section

^{165}Ho samples were placed and irradiated in different positions within the Activator (Figure 150, Figure 151 & Table 52).

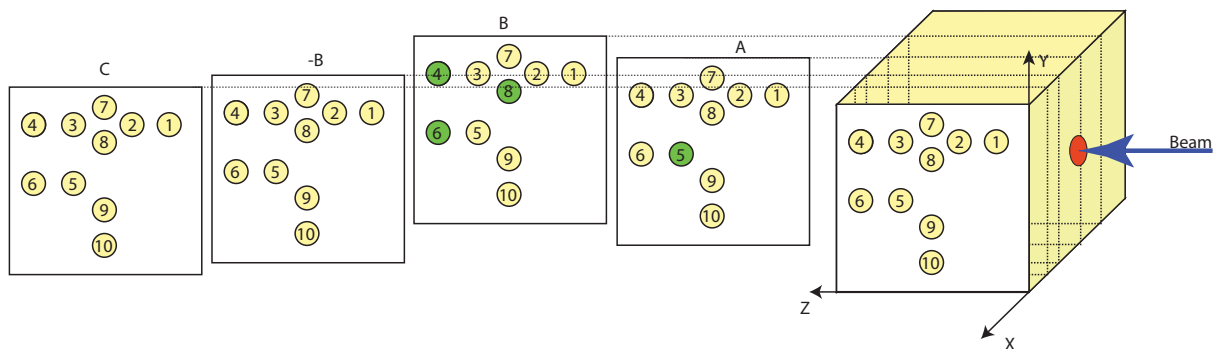


Figure 150: ^{165}Ho sample positions inside of the Activator (60 MeV p).

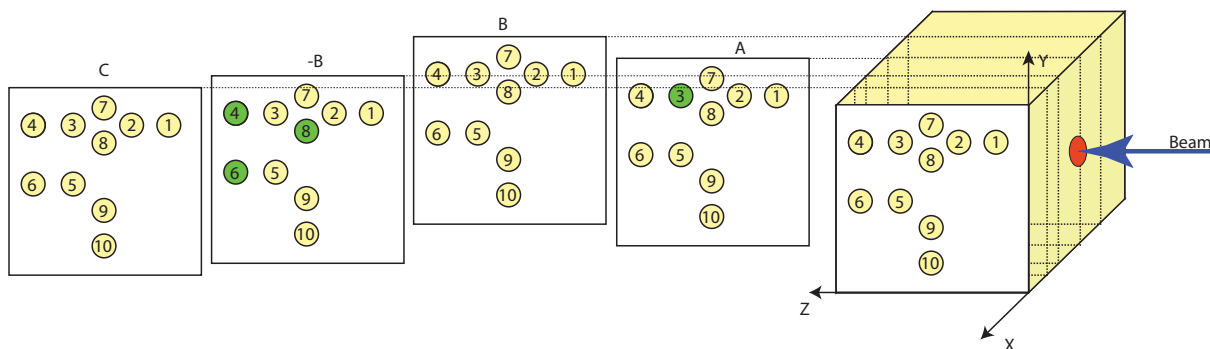
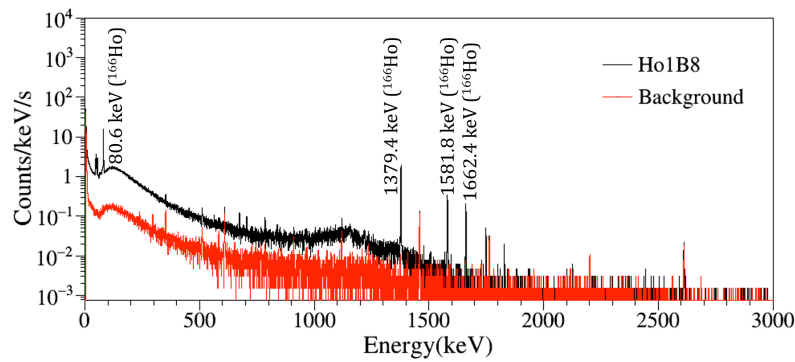
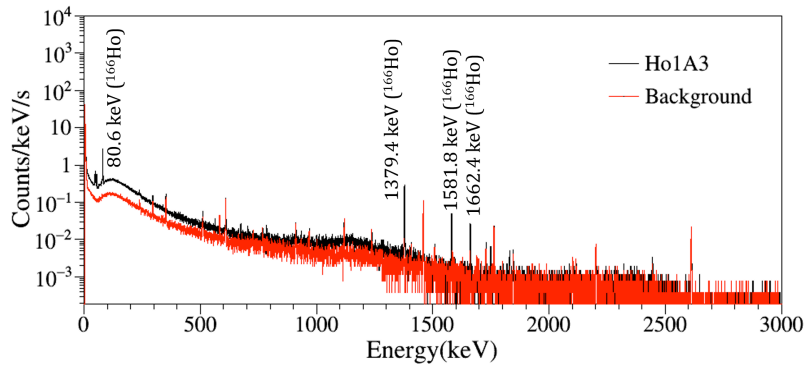


Figure 151: ^{165}Ho sample positions inside of the Activator (30 MeV d).

Table 52: ^{Nat}Ho sample positions inside of the Activator for all runs

	Run	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
p beam	15	1	A5	16.9	0.623
	15	1	B4	42.9	0.697
	15	1	B6	39.8	0.69
	15	1	B8	22.1	0.591
d beam	18	1	A3	23.3	0.548
	18	1	-B4	42.9	0.597
	18	1	-B6	39.8	0.566
	18	1	-B8	22.1	0.431

In the γ -ray spectrum, ^{166}Ho ($T_{1/2} = 26.824$ h) was identified. In Figure 152 & Figure 153, we can remark the peak that was used for the activity calculation ($E_\gamma = 1379.4$ KeV, $I_\gamma = 0.922$ %).

Figure 152: γ -ray spectrum from the irradiated ^{Nat}Ho foil (run15, p_{beam} , $T_{cool} = 3.1$ h).Figure 153: γ -ray spectrum from the irradiated ^{Nat}Ho foil (run18, d_{beam} , $T_{cool} = 26.8$ h).Table 53: Experimental and MC Simulation Activities for ^{166}Ho

	Run #	Thickness (mm)	Position	d (cm)	Exp Activity (Bq/g/h/ μ A)	Err (%)	PHITS Activity (Bq/g/h/ μ A)	Err (%)
p beam	15	1	A5	16.9	3.77E+05	4.34	130000	4.95
	15	1	B4	42.9	3.91E+05	4.34	131000	4.2
	15	1	B6	39.8	3.74E+05	4.33	122000	4.1
	15	1	B8	22.1	4.74E+05	4.33	150000	5.02
d beam	18	1	A3	23.3	1.85E+05	6.53	64200	6.23
	18	1	-B4	42.9	1.60E+05	6.61	51800	7.27
	18	1	-B6	39.8	1.40E+05	6.69	49800	6.8
	18	1	-B8	22.1	1.95E+05	6.67	68400	5.84

In the present report the MC simulation calculations for ^{166}Ho were conducted with PHITS and not with FLUKA since the code does not include the cross section of ^{166}Ho .

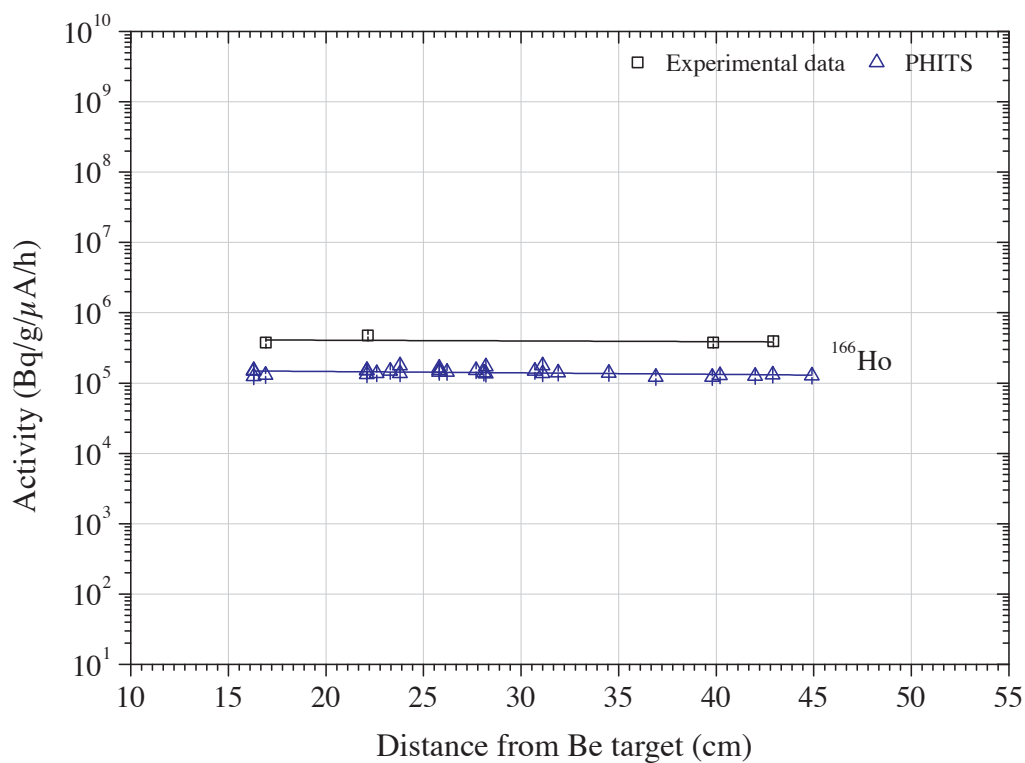


Figure 154: Measured activity for ^{166}Ho for p beam.

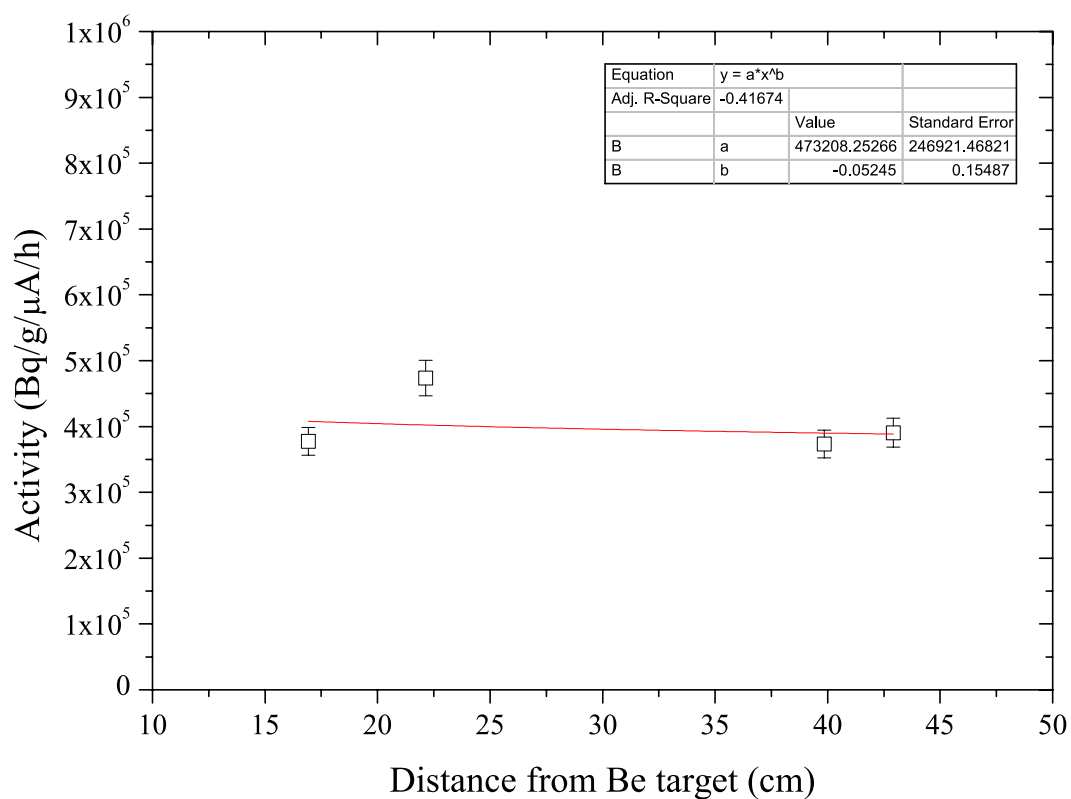


Figure 155: Measured activity (fitting curve) for ^{166}Ho for p beam.

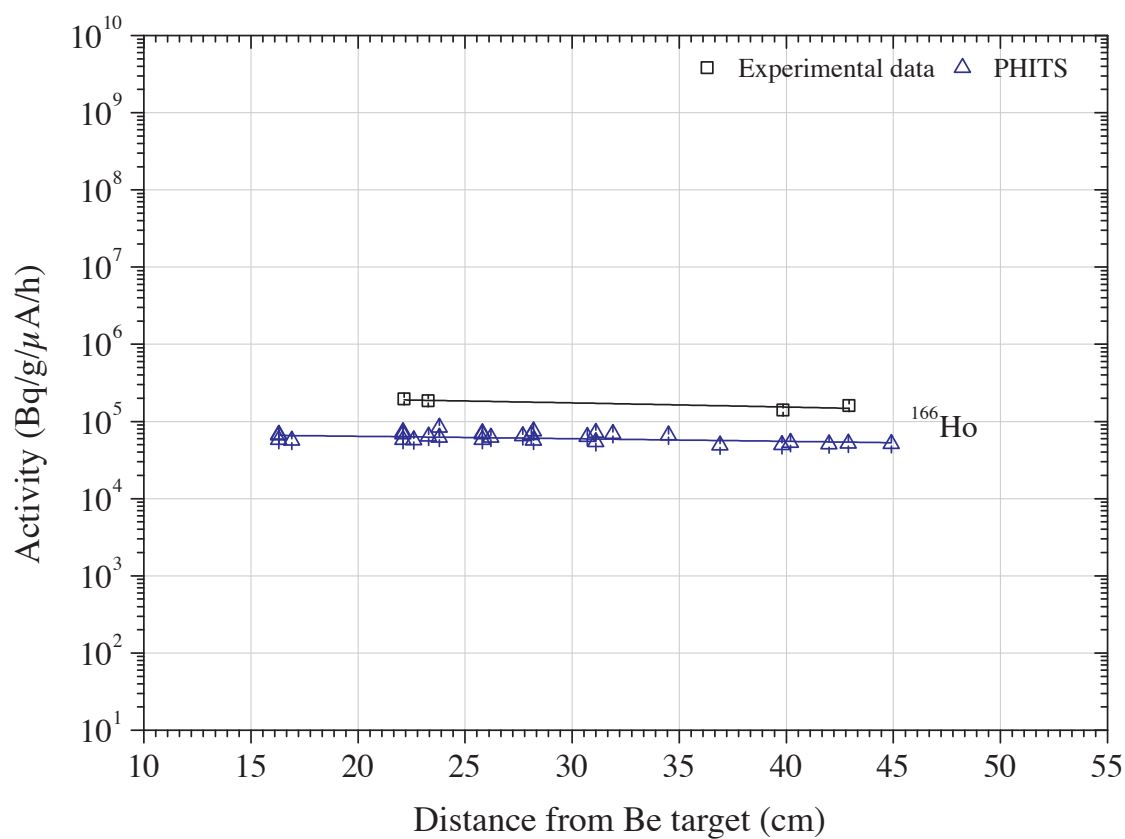


Figure 156: Measured activity for ^{166}Ho for d beam.

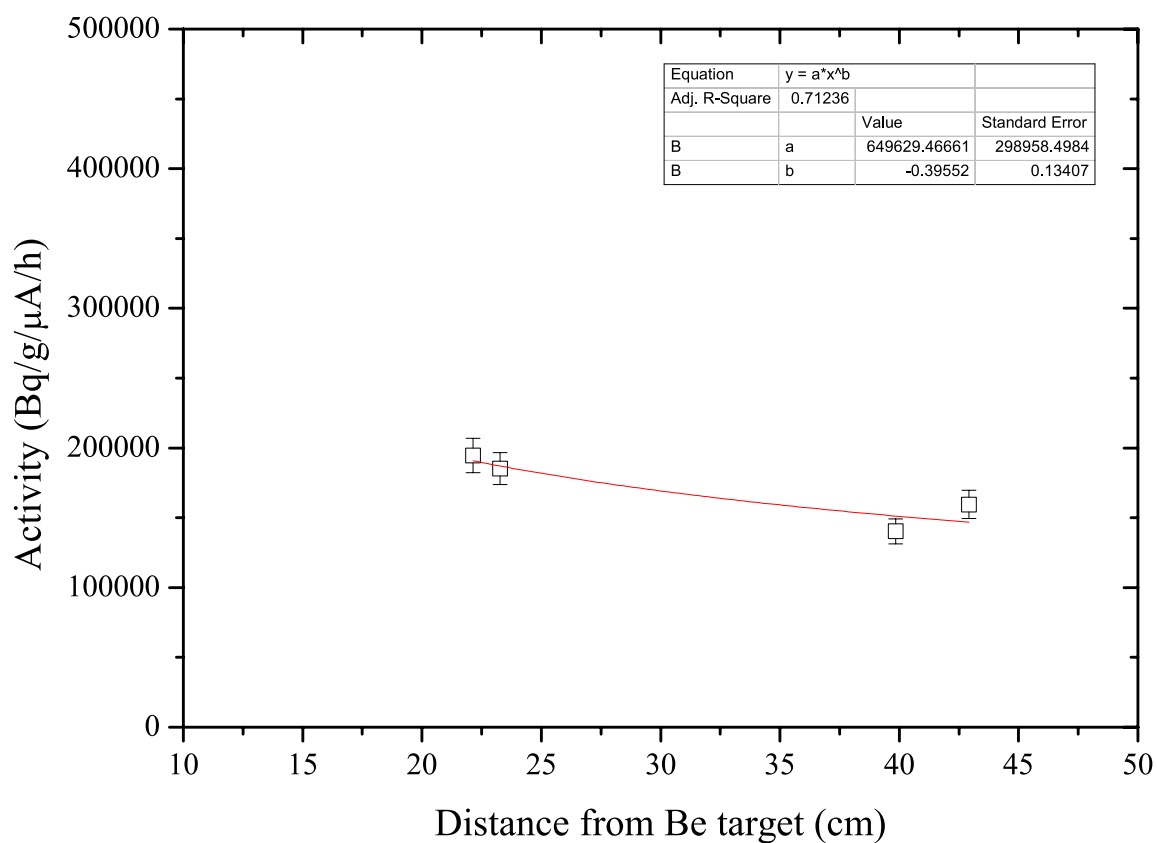


Figure 157: Measured activity (fitting curve) for ^{166}Ho for d beam.

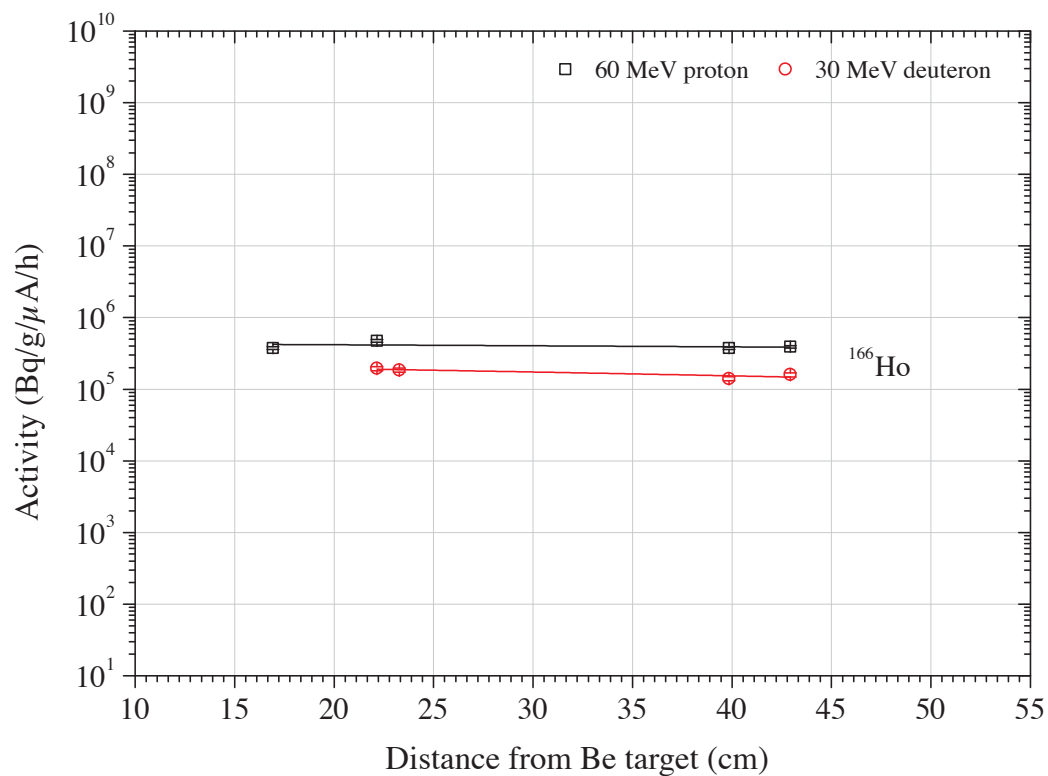


Figure 158: Measured activity for ^{166}Ho with 60 MeV protons & 30 MeV deuterons.

5.7.6 Medical isotopes Resume

In FESTA experiment we have used several medical isotopes (^{Nat}Ho , ^{Nat}Re , ^{Nat}Lu , enriched ^{98}Mo and ^{Nat}Mo) and in this report is presented the experimental results from the measurements obtained after the irradiation with proton and deuteron beams. The figures show the measured activities for all medical isotopes and the calculated MC activities except for ^{Nat}Ho were it was used only PHITS code.

The table shows

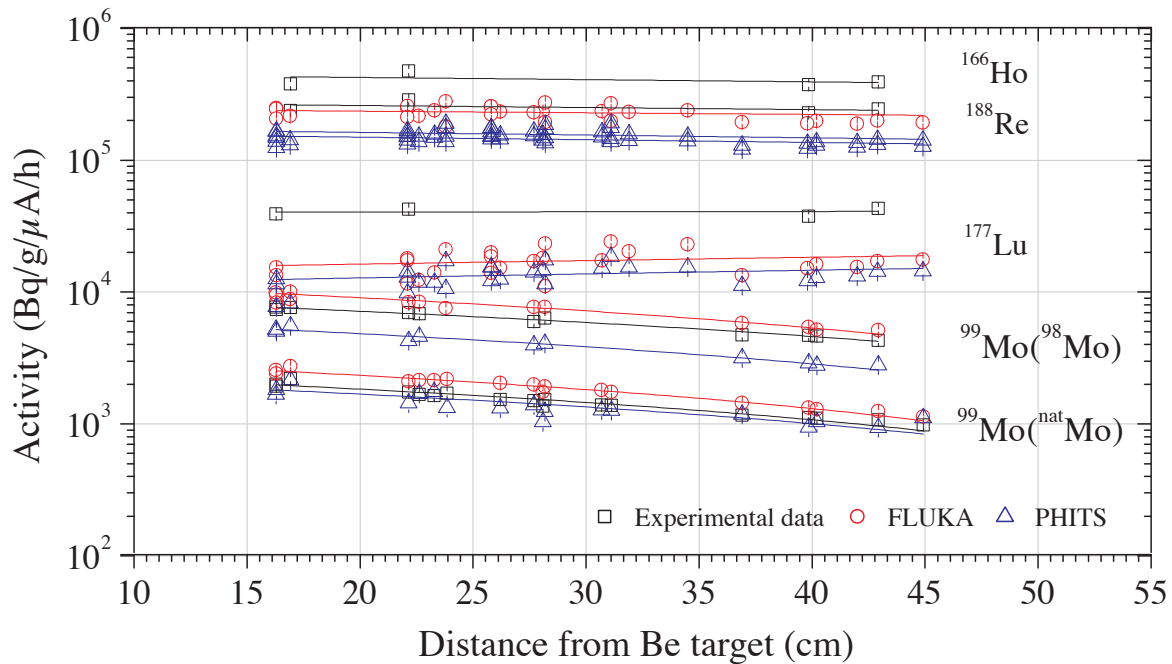


Figure 159: Measured activity for ^{Nat}Ho , ^{Nat}Re , ^{Nat}Lu , enriched ^{98}Mo and ^{Nat}Mo (60 MeV p).

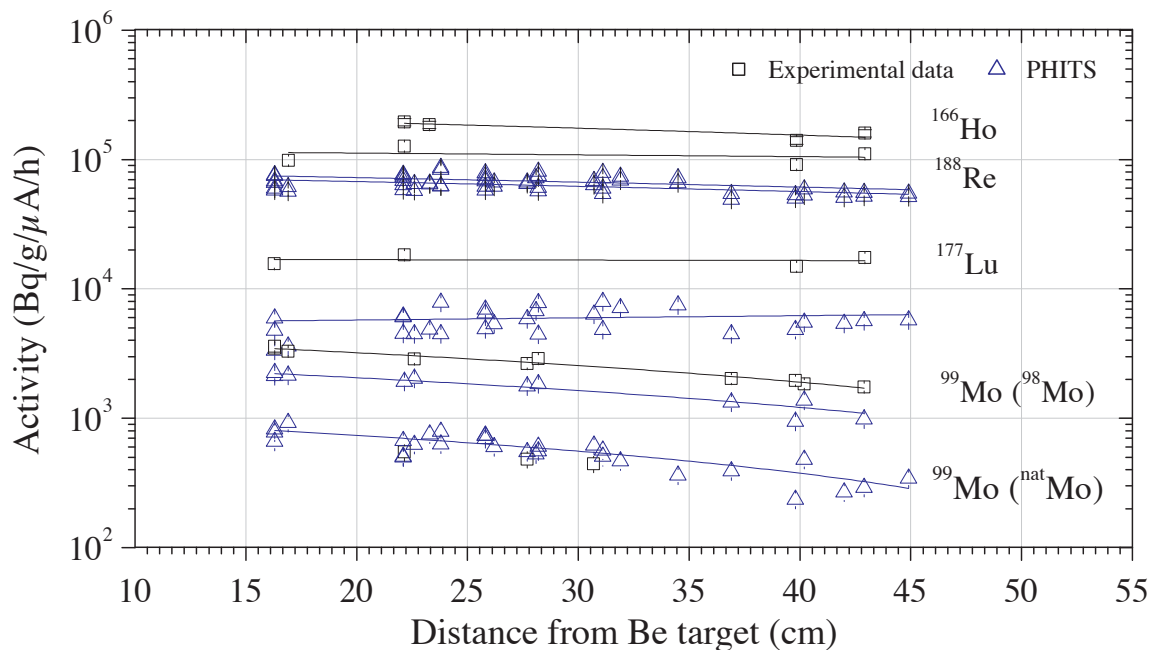


Figure 160: Measured activity for ^{Nat}Ho , ^{Nat}Re , ^{Nat}Lu , enriched ^{98}Mo and ^{Nat}Mo (30 MeV d).

5.7.7 Thickness Effect

Thickness effect studies at 60 MeV proton beam, were conducted in FESTA experiment with ^{nat}W (run08 & run09), ^{nat}Mo (run08 & run09) and ^{98}Mo metallic powder (run13, run14 & run16). The results of the activity calculation at different thickness placed in several positions in the three layers (A, B & C) within the Activator are shown in Figure 161 & Figure 162. The thinner the sample in mg/cm^2 , the higher the resonance integral and thus the induced activity.

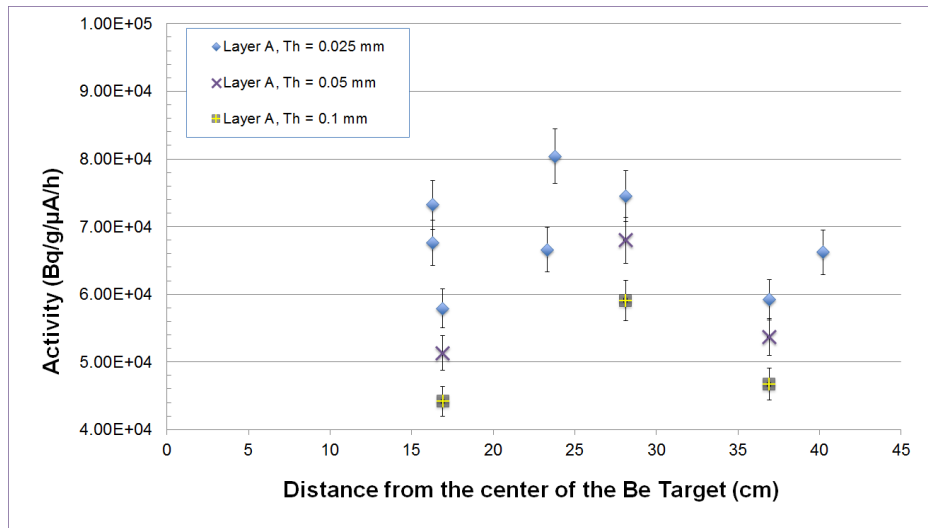


Figure 161: ^{nat}W & activity for different thicknesses.

Somehow one can see that in case of ^{nat}Mo with increasing the sample thickness, the activity decreases slightly. 0.1 mm thickness ^{nat}Mo foil have ~20% more specific activity than 1 mm thickness ^{nat}Mo foil. In the case of enriched ^{98}Mo powder, 0.25 mm thickness ^{98}Mo powder have ~10% more specific activity than 1 mm thickness ^{98}Mo powder.

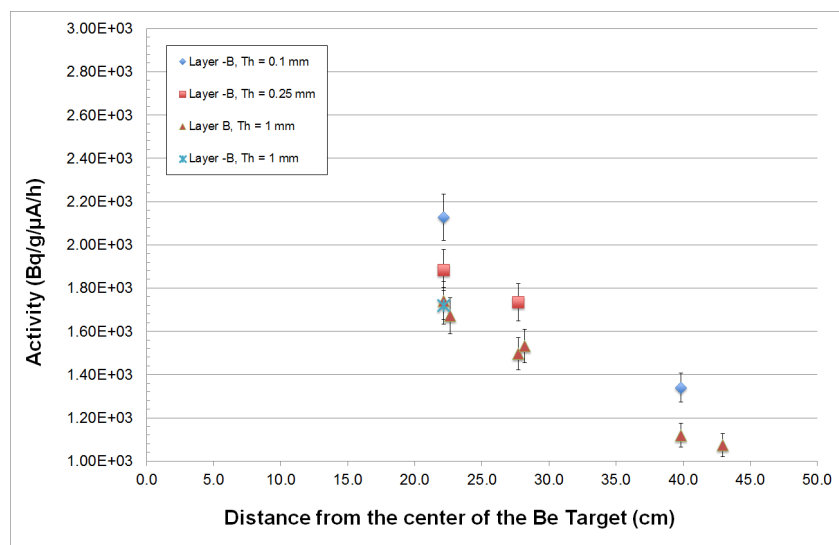


Figure 162: ^{nat}Mo activity for different thicknesses.

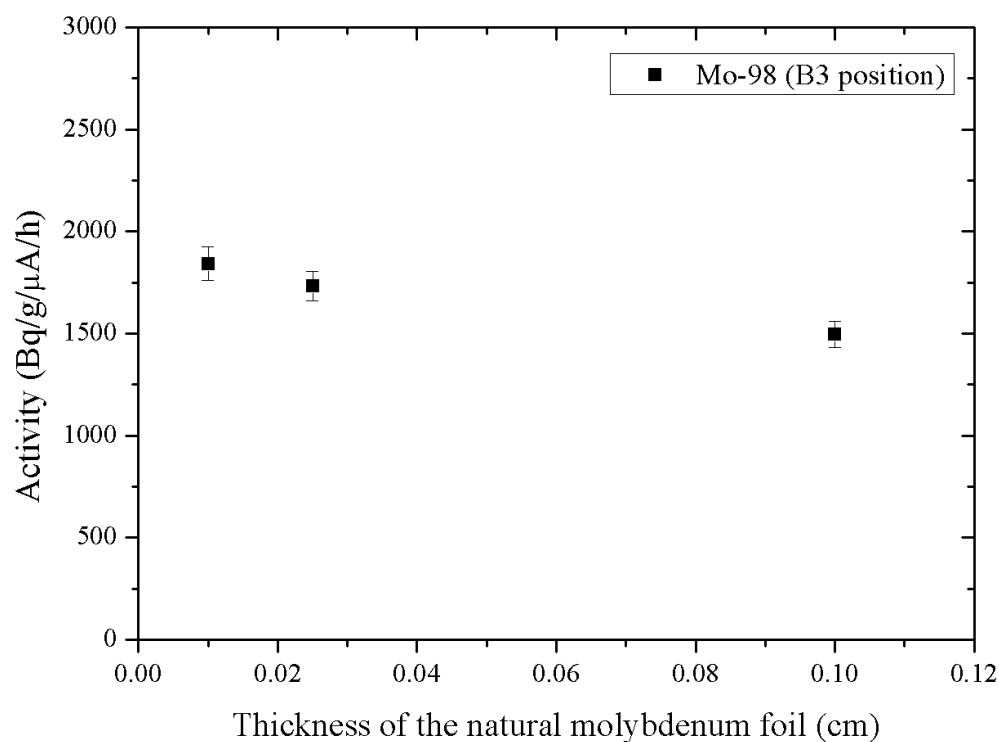


Figure 163: Activity of ^{98}Mo for different thicknesses (^{nat}Mo , p_{beam}).

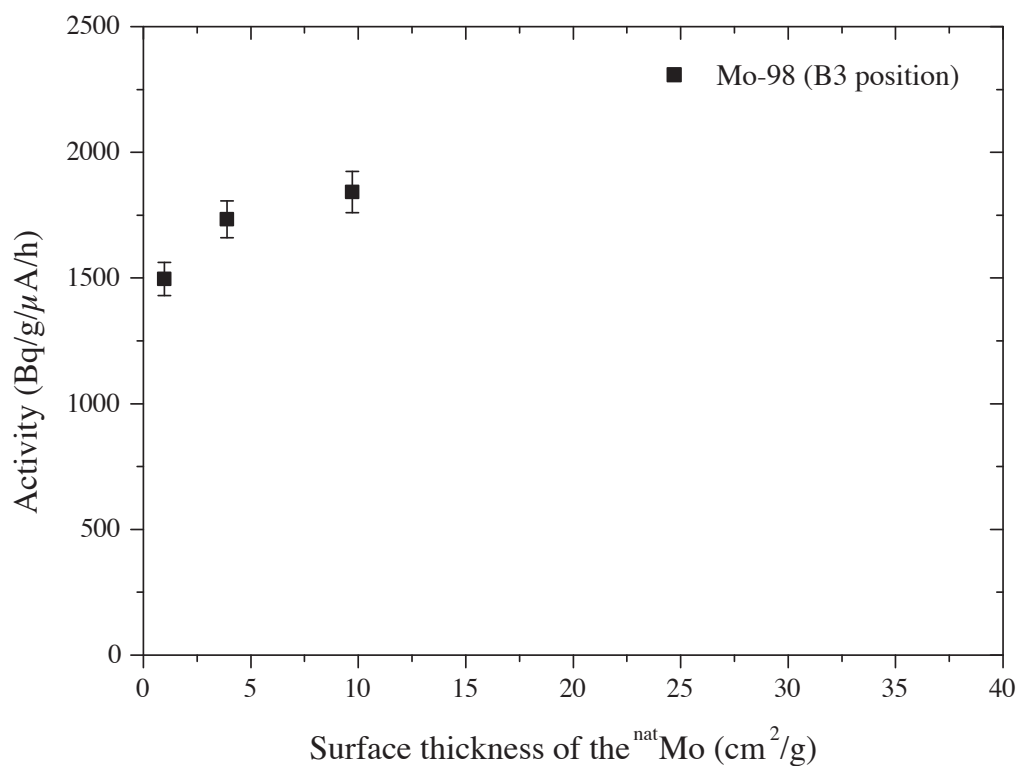


Figure 164: Activity of ^{98}Mo for different surface thicknesses (^{nat}Mo , p_{beam}).

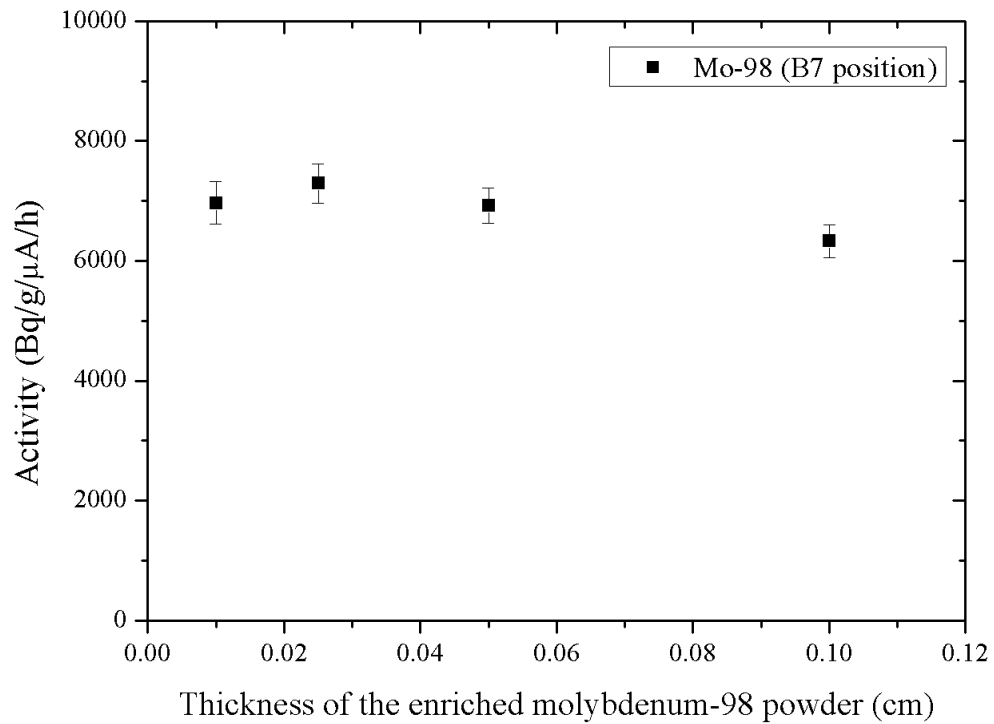


Figure 165: Activity of ^{98}Mo for different thicknesses (^{99}Mo , p_{beam}).

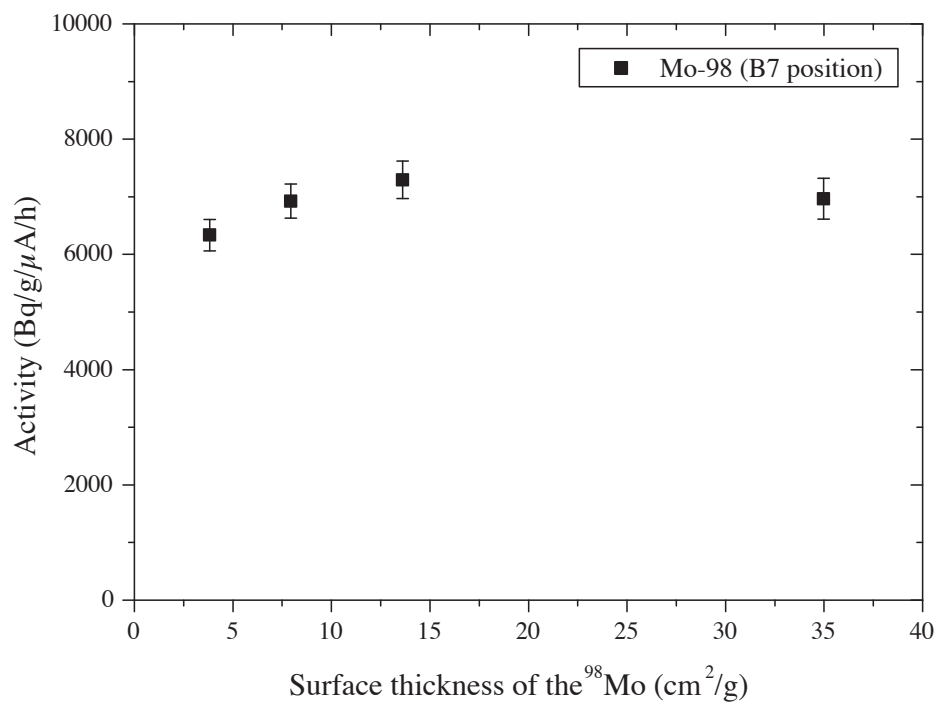


Figure 166: Activity of ^{98}Mo for different surface thicknesses (^{99}Mo , p_{beam}).

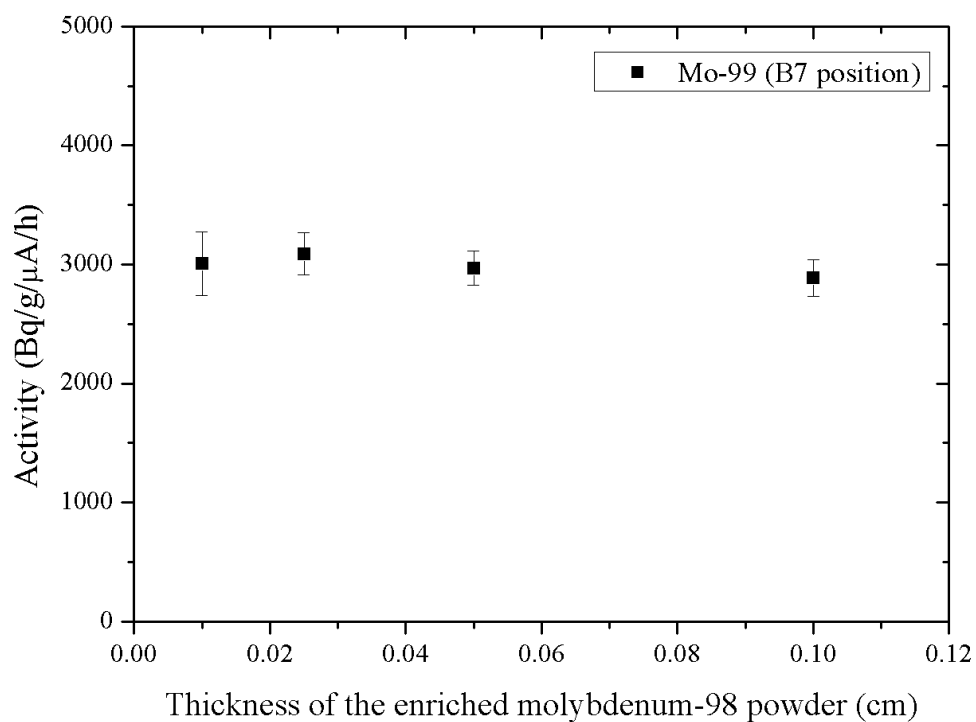


Figure 167: Activity of ^{98}Mo for different thicknesses (^{99}Mo , d_{beam}).

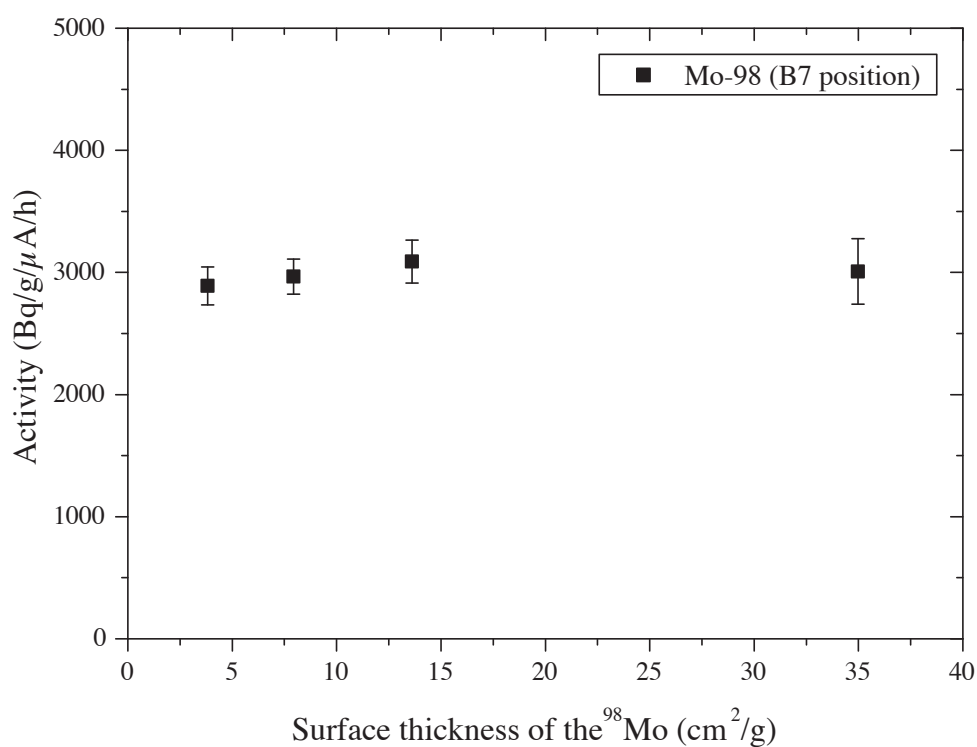


Figure 168: Activity of ^{98}Mo for different surface thicknesses (^{99}Mo , d_{beam}).

In the case with the W samples in layer A, there is a difference of the order of almost 30% (Figure 169).

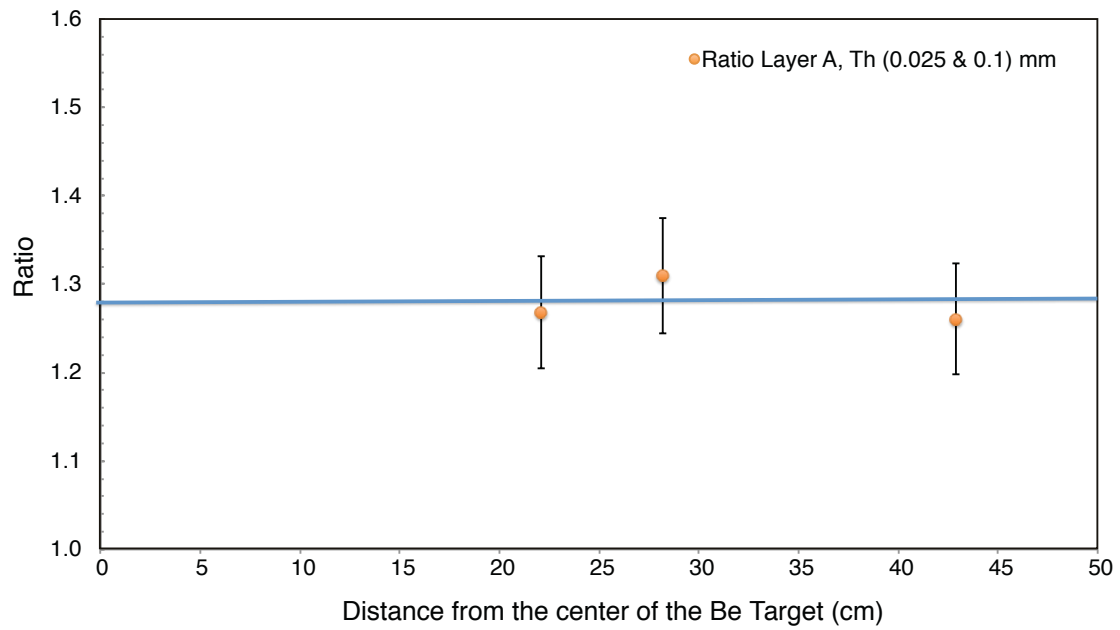


Figure 169: ^{187}W activity ratio for different thicknesses in Layer A.

Therefore, experimental testing of the effect of thickness in the irradiated samples leaves a possibility of finding the optimal thickness for better performance of neutron capture by adiabatic resonance in the activator.

5.7.8 Effect of the Diffuser support (iron)

A test to verify the asymmetry of neutron capture at 60 MeV protons, on the top and bottom region of the activator with respect to the center of the target (22.1 cm & 28.2 cm) was held in the run02 & run 20.

Three activation foils were used: ^{nat}Zr (Run2, Layer A), ^{nat}In (Run20, Layer A), and ^{nat}W (Run20, Layer B). We can compare the activities of 5 radioisotopes: ^{116m}In , ^{115m}In , ^{187}W , ^{97}Zr , and ^{89}Zr produced by $^{115}\text{In}(n, \gamma)^{116m}\text{In}$, $^{115}\text{In}(n, n')^{115m}\text{In}$, $^{186}\text{W}(n, \gamma)^{187}\text{W}$, $^{96}\text{Zr}(n, \gamma)^{97}\text{Zr}$, and $^{90}\text{Zr}(n, 2n)^{89}\text{Zr}$ respectively.

There is apparently a clear evidence of significant loss of neutrons in the bottom part of the Activator, which obviously cause a decrease in the neutron capture. One reason for this problem is due to the iron table, which supports the lead block, whose legs pierce the Activator, outward, allowing an important neutron leak through them. It was estimated that a possible optimization of the Activator to correct this problem might improve the neutron capture.

Table 54 Experimental and MC Activities

Run #	Thickness (cm)	Position	d (cm)	Radioisotope	Exp Activity (Bq/g/h/ μA)	Err (%)	FLUKA Activity (Bq/g/h/ μA)	Err (%)	PHITS Activity (Bq/g/h/ μA)	Err (%)
2	0.0127	A7	23.8	^{97}Zr	9.59E+02	8.7	5.84E+02	8.46	7.13E+02	26.5
2	0.0127	A8	16.3	^{97}Zr	9.22E+02	8.88	6.04E+02	6.97	6.83E+02	27.6
2	0.0127	A9	16.3	^{97}Zr	8.88E+02	8.75	6.26E+02	5.87	2.99E+02	26.4
2	0.0127	A10	23.8	^{97}Zr	7.80E+02	8.7	5.28E+02	5.03	6.12E+02	24.4
2	0.0127	A7	23.8	^{89}Zr	9.63E+01	9.91				
2	0.0127	A8	16.3	^{89}Zr	5.55E+02	8.83				
2	0.0127	A9	16.3	^{89}Zr	5.40E+02	8.73				
2	0.0127	A10	23.8	^{89}Zr	9.48E+01	9.78				
20	0.0127	A7	23.8	^{116m}In	3.45E+07	3.71	2.33E+07	2.19	1.81E+07	8.12
20	0.0127	A8	16.3	^{116m}In	2.80E+07	3.72	1.85E+07	3.1	1.41E+07	7.7
20	0.0127	A9	16.3	^{116m}In	1.91E+07	3.73	1.35E+07	2.9	1.11E+07	7.92
20	0.0127	A10	23.8	^{116m}In	1.82E+07	3.74	1.29E+07	3.53	1.28E+07	8.16
20	0.0127	A7	23.8	^{115m}In	1.50E+04	32.4	2.67E+04	4.22	3.31E+04	25.9
20	0.0127	A8	16.3	^{115m}In	4.85E+04	12.1	7.74E+04	1	9.64E+04	16.5
20	0.0127	A9	16.3	^{115m}In	4.24E+04	11	7.72E+04	1.14	9.79E+04	17.6
20	0.0127	A10	23.8	^{115m}In	1.52E+04	19	2.50E+04	1.5	3.42E+04	23.9
20	0.025	B7	28.2	^{187}W	8.92E+04	3.78	8.73E+04	2.95	4.75E+04	8.38
20	0.025	B8	22.1	^{187}W	7.88E+04	3.8	7.69E+04	3.79	3.69E+04	8.97
20	0.025	B9	22.1	^{187}W	6.43E+04	3.83	6.44E+04	3.24	2.83E+04	9.24
20	0.025	B10	28.2	^{187}W	5.77E+04	3.84	6.13E+04	5.16	3.45E+04	7.52

As shown in Figure 170, the activity at positions 22.1 cm from the Be target show a slight difference, which is becoming more important as a function of the distance from the target.

Table 55: Ratio of the Activity in in different holes.

Run #	Layer	Radioisotope	Activity(Hole10)/Activity(Hole7)			Activity(Hole9)/Activity(Hole8)		
			Exp	FLUKA	PHITS	Exp	FLUKA	PHITS
2	A	^{97}Zr	0.81 $\pm$ 0.10	0.90 $\pm$ 0.09	0.86 $\pm$ 0.31	0.96 $\pm$ 0.12	1.04 $\pm$ 0.09	0.44 $\pm$ 0.17
2	A	^{89}Zr	0.98 $\pm$ 0.14			0.97 $\pm$ 0.12		
20	A	^{116m}In	0.53 $\pm$ 0.03	0.55 $\pm$ 0.02	0.71 $\pm$ 0.08	0.68 $\pm$ 0.04	0.73 $\pm$ 0.03	0.79 $\pm$ 0.09
20	A	^{115m}In	1.01 $\pm$ 0.38	0.94 $\pm$ 0.04	1.03 $\pm$ 0.36	0.87 $\pm$ 0.14	1.00 $\pm$ 0.02	1.02 $\pm$ 0.25
20	B	^{187}W	0.65 $\pm$ 0.03	0.70 $\pm$ 0.04	0.73 $\pm$ 0.08	0.82 $\pm$ 0.04	0.84 $\pm$ 0.04	0.77 $\pm$ 0.10

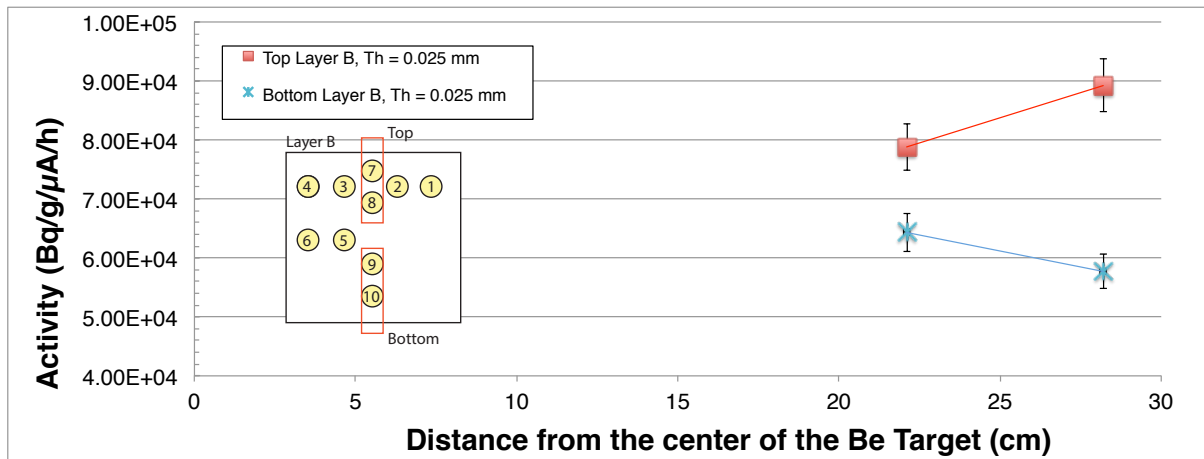


Figure 170: ^{238}W activity in top and bottom positions in Layer B.

It is very difficult to judge from the activity measurements since a variation of the activity as a function of distance does not necessarily imply a similar variation of the neutron flux since energy of neutrons and this neutron cross section also vary as a function of the distance. For this study it is necessary to compare fluxes and not just the measured activities.

5.7.9 Neutron leakage outside of the Activator

^{nat}In activation foils at 60 MeV protons, were used in order to check the neutron leakage from the holes (4 & 6) of the Activator. The sample holders simply were shifted up to 15 cm from the Activator and ^{nat}In samples were placed near to the these holes as is shown in the picture (Figure 171). Natural indium foils were irradiated near to these holes 4 and 6.



Figure 171: ^{nat}In sample positions outside the Activator near to holes 4 & 6.

The sample after irradiation was measured at a distance, $d = 0$ cm from the HPGe detector, due to its very low activity. In fact to have the final result, it was necessary to make a measurement with the standard source at the same distance, however that was not done.

The conclusion is that there is little neutron leakage from the Activator.

5.7.10 Moderation and reflector effects

In FESTA a special run (run16) with 60 MeV proton beam was conducted to test the effects of graphite moderation and reflection.

To this end there were conducted two specific studies, as follow:

1. Check in holes 4 & 6 the carbon (graphite) reflector effect using ^{nat}W activation foils & ^{nat}Mo samples, displacing the sample holders 15 cm out from the Activator in order to allocate the activation foils in the graphite layer region (Figure 172), unlike the previous runs, where samples were located in the region of the lead diffuser.

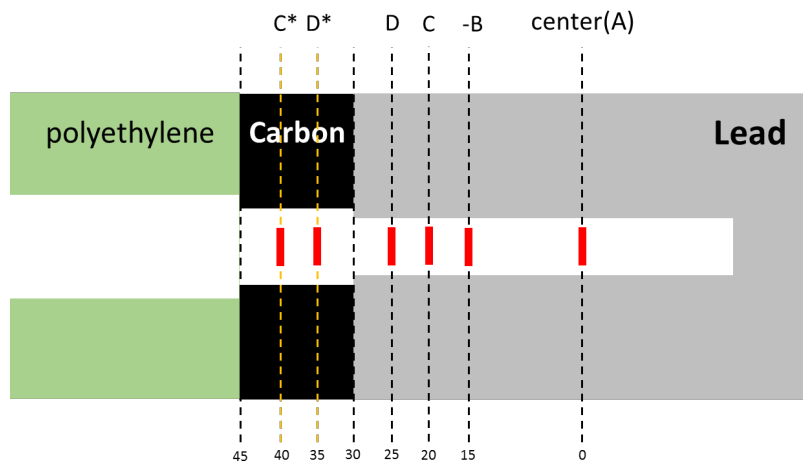


Figure 172: Sample positions in hole 4 and 6.

The activities are shown in the Tables and in Figure 173 for ^{nat}W and ^{nat}Mo .

Table 56: Experimental Activitiy for ^{nat}W (0.25 mm thickness)

Sample code	Position	Distance (cm)	Activity (Bq/g/ μ A/h)	Error (%)
W025A4	A4	40.2	6.98E+04	4.51
W025B'4	-B4	42.9	6.97E+04	4.52
W025C*4	C*4	56.7	8.06E+04	4.52
W025C4	C4	44.9	7.01E+04	4.52
W025D*4	D*4	53.3	8.30E+04	4.52
W025D4	D4	47.4	7.13E+04	4.52

Table 57: Experimental Activitiy for ^{nat}Mo (1 mm thickness)

Sample code	Position	Distance (cm)	Activity (Bq/g/ μ A/h)	Error (%)
natMo1A6	A6	35.3	1.22E+03	4.42
natMo1B'6	-B6	38.3	1.13E+03	4.65
natMo1C*6	C*6	53.3	6.51E+02	4.51
natMo1C6	C6	40.5	1.12E+03	4.7
natMo1D*6	D*6	49.7	8.75E+02	4.35
natMo1D6	D6	43.3	1.05E+03	4.48

One can notice that there is a slight increase of the activity in the case of ^{nat}W when the samples are placed in the graphite region, which can be explained as the result of the contribution of the captures conducted by the slowing down escape of fast neutrons (W has major resonance cross section at 18.9 eV).

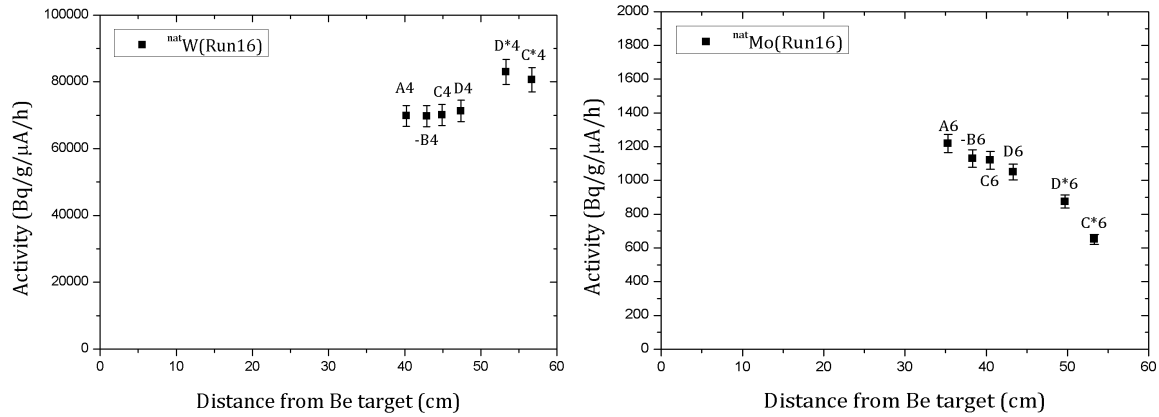


Figure 173: The activity of the irradiated ^{nat}W & ^{nat}Mo foils (Hole 4).

In the case of ^{nat}Mo the measured activity in the carbon region decreases and that could be explained because of the absence of epithermal neutrons, which are easily captured by ^{98}Mo . However, a more reliable explanation is based on the fact that the activity is proportional to Φ and σ , and when going to graphite, both are crucial to the induced activity. Thus, the analysis of the fluence in the activator is required.

2. Check the moderation effect.

The purposes of this study were to increase locally the moderation of the neutrons close to the samples. This was done by replacing lead bricks by small graphite bricks inside of the hole.

In Run16, the graphite bricks were placed inside of the hole7 and hole8 instead of lead bricks. However the samples (^{98}Mo , ^{nat}Ho , ^{nat}Re , and ^{nat}Lu) have the same activity as for the normal geometry (graphite + lead bricks).

The activities in both configurations are compared in the Table 58 and Figure 174. Results indicate that the samples are not affected by this modification.

Table 58: Comparison with Measured activity

Run #	Material	Position	Distance (cm)	Mass (g)	Exp Activity (Bq/g/h/μA)	Err (%)
16	^{98}Mo	B7	28.2	0.399	6.46E+03	4.44
13	^{98}Mo	B7	28.2	0.403	6.33E+03	4.31
16	^{nat}Ho	-B8	22.1	0.542	4.39E+05	4.32
15	^{nat}Ho	B8	22.1	0.591	4.74E+05	4.33
16	^{nat}Re	B8	22.1	1.43	2.80E+05	4.07
14	^{nat}Re	B8	22.1	1.424	2.85E+05	4.04
16	^{nat}Lu	A8	16.3	0.75	3.81E+04	3.87
14	^{nat}Lu	A8	16.3	0.708	3.91E+04	3.78

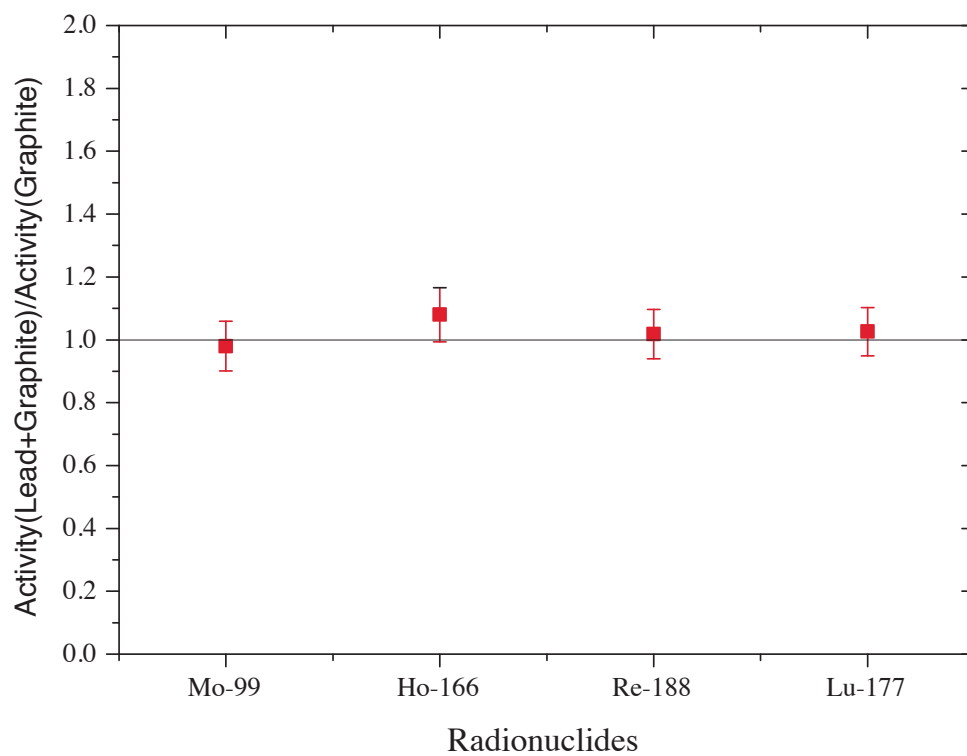


Figure 174: The activity of the irradiated ^{Nat}W & ^{Nat}Mo foils (Hole 4).

5.7.11 Symmetry in B & -B layers

A cross checks of the symmetry between layer B & -B can be processed with FESTA results as it is shown in the Table 59. These are the results using a 60 MeV proton beam at different positions.

Table 59: Experimental Activity.

Run #	Sample Code	Thickness (cm)	Position	Distance (cm)	Mass (g)	EXP (Bq/g/h/ μ A)	err(%)
13	116mIn	0.0127	-B4	42.9	0.02871	2.75E+07	5.91
5	116mIn	0.0127	B4	42.9	0.0279	2.83E+07	5.91
20	116mIn	0.0127	B4	42.9	0.0324	2.91E+07	6.05
13	198Au	0.0051	-B6	39.8	0.0273	2.27E+05	5.28
6	198Au	0.0051	B6	39.8	0.0273	2.39E+05	5.36
13	natMo	0.1	-B8	22.1	0.874	1.72E+03	5.73
9	natMo	0.1	B8	22.1	0.874	1.74E+03	5.71
13	natW	0.025	-B5	22.6	0.359	6.62E+04	5.91
7	natW	0.025	B5	22.6	0.402	6.60E+04	5.91

One can notice that it was obtained almost same results of the activity of the samples placed at the same distance from the target in both layers (B & -B).

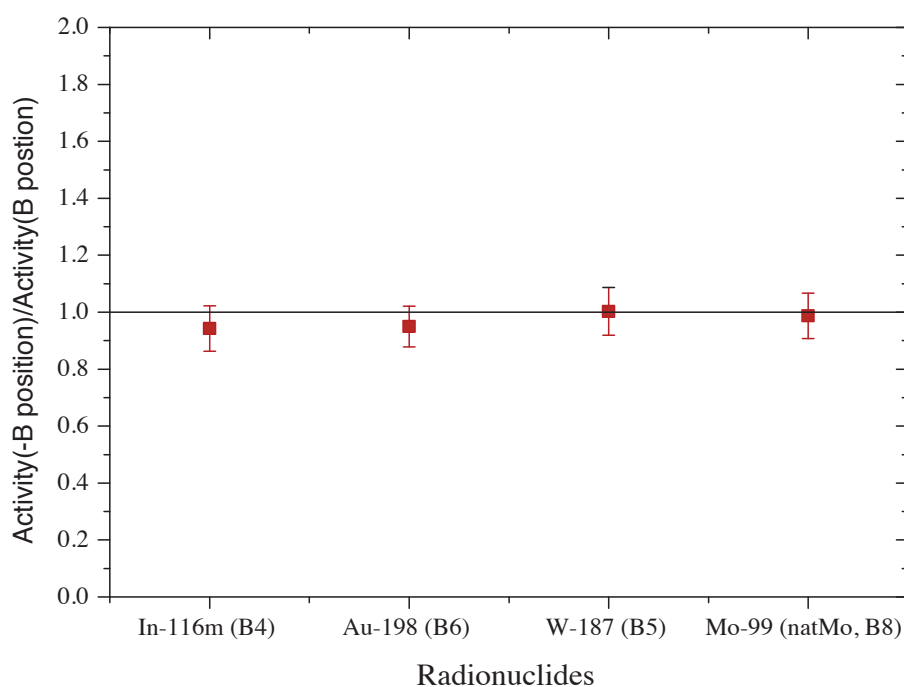


Figure 175: Activity ratio of the irradiates samples in layers B & -B.

5.7.12 Neutron source estimation

In Run17, Hole 5 was left open and four samples (^{nat}In , ^{nat}Au , ^{nat}W , and ^{nat}Al) were placed close to the hole to check the neutron leakage.

Table 60: Characteristics of the samples

No	Material	Thickness (mm)	Position	Distance from Be target (cm)	Mass (g)
1	^{nat}In	0.127	A6	36.9	0.03362
2	^{nat}In	0.127	F5	79.8	0.2606
3	^{nat}Au	0.051	F5	79.8	0.11878
4	^{nat}W	0.25	F5	79.8	0.406
5	^{nat}Al	0.762	F5	79.8	0.2606

The measured activity for the samples in front of hole5 (Table 61) was about 10^2 times smaller than the samples inside the activator (Table 62). The radionuclides ^{24}Na produced by $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ was not detected due to their low activity.

Table 61: Experimental Activity for the samples

Position	Distance (cm)	Radionuclides	Activity (Bq/g/ $\mu\text{A/h}$)	Error (%)
A6	36.9	^{116m}In	$2.38\text{E}+07$	4.62
F5	79.8	^{116m}In	$1.65\text{E}+05$	4.63
F5	79.8	^{198}Au	$1.01\text{E}+03$	3.88
F5	79.8	^{187}W	$2.75\text{E}+02$	4.98
F5	79.8	^{24}Na		

Table 62: Comparison with Measured activity in Hole 5

Position	A5		B5		C5		F5	
Distance (cm)	16.9		22.6		26.2		79.8	
Radionuclides	Activity (Bq/g/ $\mu\text{A/h}$)	Error (%)	Activity (Bq/g/ $\mu\text{A/h}$)	Error (%)	Activity (Bq/g/ $\mu\text{A/h}$)	Error (%)	Activity (Bq/g/ $\mu\text{A/h}$)	Error (%)
^{116m}In	$2.31\text{E}+07$	4.76	$2.53\text{E}+07$	5.69	$2.62\text{E}+07$	5.69	$1.65\text{E}+05$	4.62
^{198}Au	$2.16\text{E}+05$	4.2	$2.49\text{E}+05$	4.19	$2.53\text{E}+05$	4.2	$1.01\text{E}+03$	4.63
^{187}W	$5.79\text{E}+04$	4.56	$6.60\text{E}+04$	4.56	$6.64\text{E}+04$	4.56	$2.75\text{E}+02$	5.04

The Table 63 shows the activity of ^{116m}In in the same position (run4). It shows a very slight difference of the order of 5%. That could be explained by the neutron losses (escaping) across the open hole.

Table 63: Experimental Activity for ^{nat}In (run4)

Position	Distance (cm)	Radionuclides	Activity (Bq/g/ $\mu\text{A/h}$)	Error (%)
A6	36.91354765	^{116m}In	$2.51\text{E}+07$	4.77

6 Benchmark of MC Simulation codes

The production of neutrons in a Be target was one of the challenges of high importance for FESTA, as well as the production of radioisotopes with a high neutron flux to facilitate the use of the ARC method.

For the validation of the MC code to the activator, in order to accurately reproduce the neutron transport by the impact of a deuteron beam on a Be target, it was conducted a comparison of several codes as FLUKA, PHITS and GEANT4.

6.1 Protons on Be Target

Experimental measurements of neutron flux produced by proton beams (23-80 MeV) on targets Be and Pb targets is disclosed in Tilquin paper (I. Tilquin, 2005). The experimental setup of the experiment (Figure 176), with a pencil beam (zero width), has been used for the simulation, while in FESTA, the beryllium target had the following dimensions: a diameter of 50.3 mm with 37 mm thickness. The proton beam profile in FESTA had a horizontal diameter of 15 mm width and a vertical diameter of 30 mm high. The results of the simulation are shown in Table 64.

The neutron production values by using FLUKA are typically 25 ~ 55% less compared to the experimental data (Figure 177).

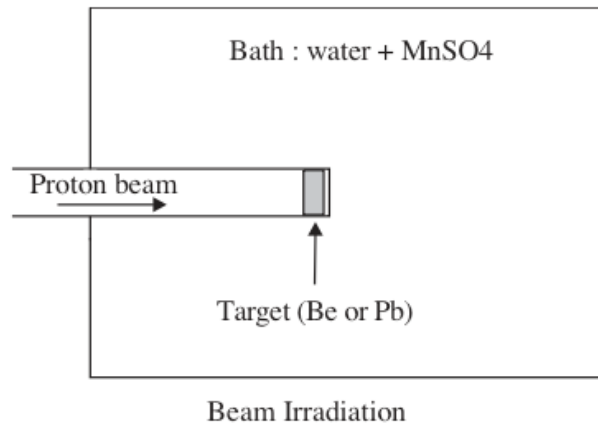


Figure 176: Experimental setup of Tilquin et al.

Table 64: Data/Simulation results

Ep (MeV)	Experiment (n/s/μA)	Error (%)	FLUKA (n/s/μA)	Error (%)	Exp./FLUKA	Error (%)
23	1.17E+11	8.49	9.25E+10	1.14	1.26	8.57
35	2.29E+11	8.41	1.71E+11	0.98	1.34	8.47
45	3.22E+11	7.97	2.52E+11	0.6	1.28	7.99
55	4.53E+11	8.45	3.48E+11	0.62	1.3	8.47
65	6.85E+11	6.2	4.56E+11	0.55	1.5	6.22
70	7.27E+11	7.89	5.08E+11	0.62	1.43	7.91
75	8.80E+11	5.57	5.67E+11	0.38	1.55	5.58
80	8.45E+11	8.52	6.20E+11	0.43	1.36	8.53

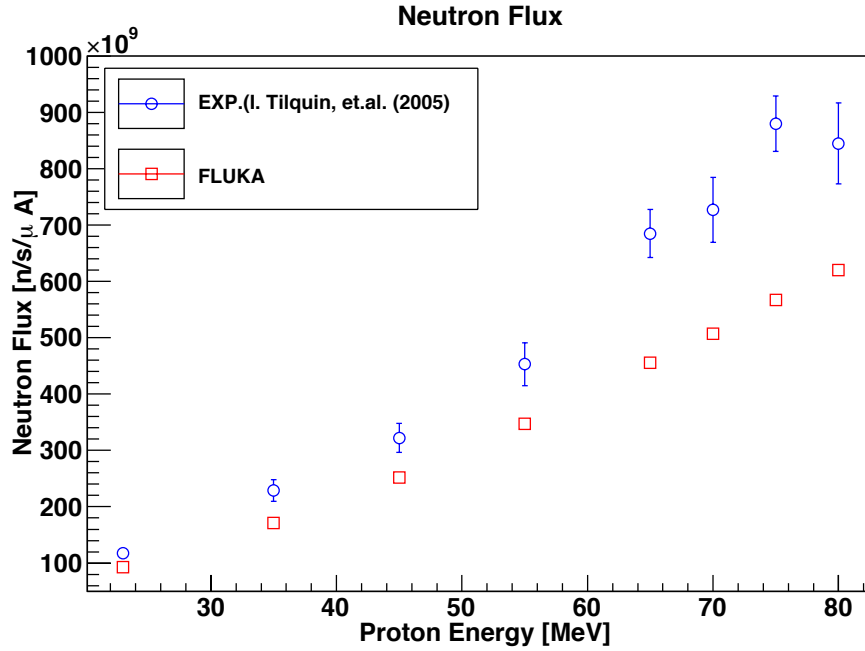


Figure 177: Neutron Flux as function of the proton energy.

6.2 Deuterons on Be Target

For the deuteron beam simulation, the report (M. Majerle, 2010) is the preferred reference. It is originally for the benchmark of McDeLi (or McDeLicious) code, but it includes several other previous experimental data. Figure 178 shows the experimental data, where we can observe large discrepancies at deuteron energies around 30 MeV ~ 40 MeV. For example Amols' data shows lower values at 35 MeV than Meulders' data at 33 MeV (J. P. Meulders et al.). Aoki's experimental data (T. Aoki et al.) at deuteron energy 25 MeV in 2004 was used for this benchmark work.

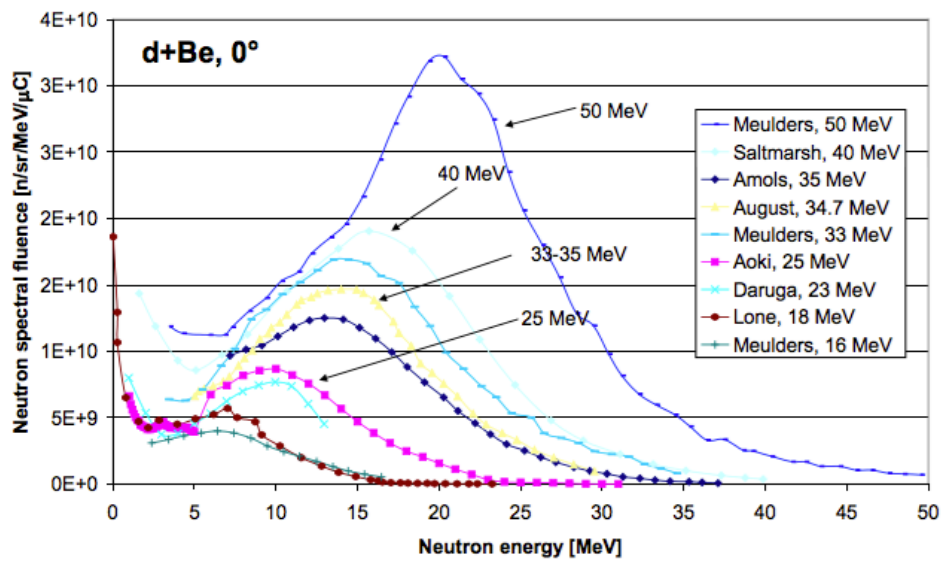


Figure 178: Neutron fluence in the forward direction of the experimental data (M. Majerle, 2010).

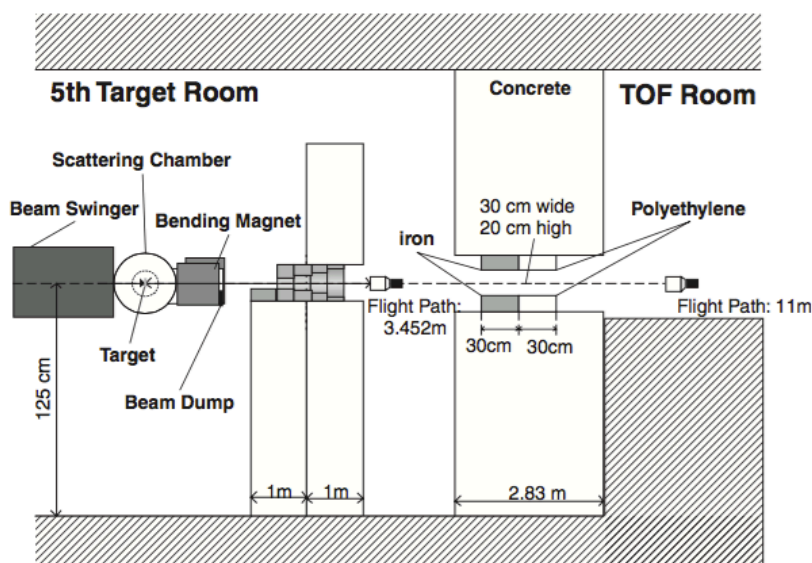


Figure 179: View of the experimental setup.

Short description of Aoki et. al. data

Neutron emission spectra in the (d, n) reactions on thick ^9Be target were measured for 25 MeV deuterons at the Tohoku University AVF cyclotron facility (Figure 179). The beryllium target was $\sim 3\text{mm}$ thick to stop the incident beam within the targets. Target area size was $2\text{ cm} \times 2\text{ cm}$. The spectra were measured with the time-of-flight method at 10 laboratory angles between 0° and 90° using two NE213 detector systems.

Short description of KA Weaver data

In the paper of KA Weaver (Keith Allen Weaver), Neutrons from deuteron bombardment of light nuclei, it is described an experiment that uses a 18 MeV deuteron beam on a Be target of 5.08 cm in diameter and 2.56 mm thick, weighing 9.57 g. The results of this experiment are shown in Figure 180.

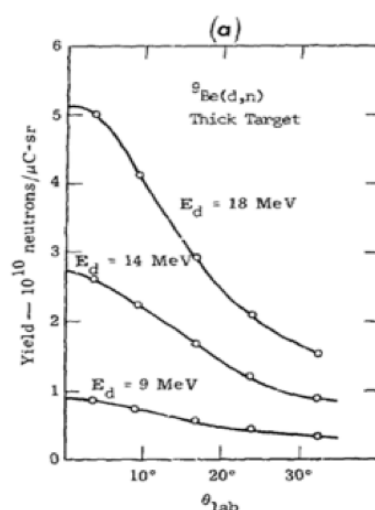


Figure 180: Deuteron Beam on Be Target @ 18 MeV (Experimental results).

The neutron yield from the Be target at different angles obtained with FLUKA is shown in Figure 181.

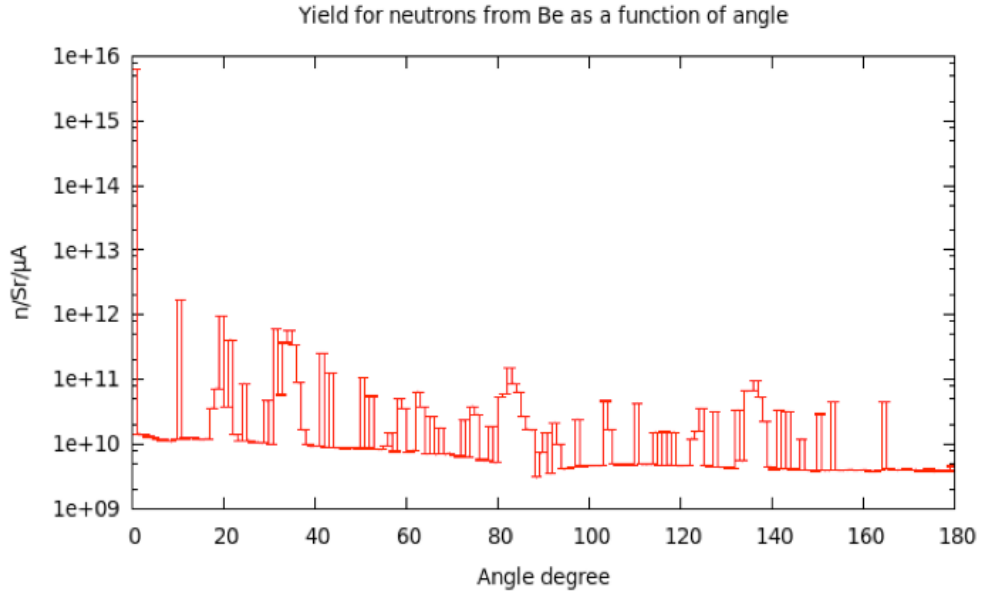


Figure 181: Deuterons on Be Target @ different angles (FLUKA).

One can observe the presence of abnormal peaks at different angles. For other two angles at 3.5° and 16.7° FLUKA simulation were also conducted on a Be target, see Figure 182.

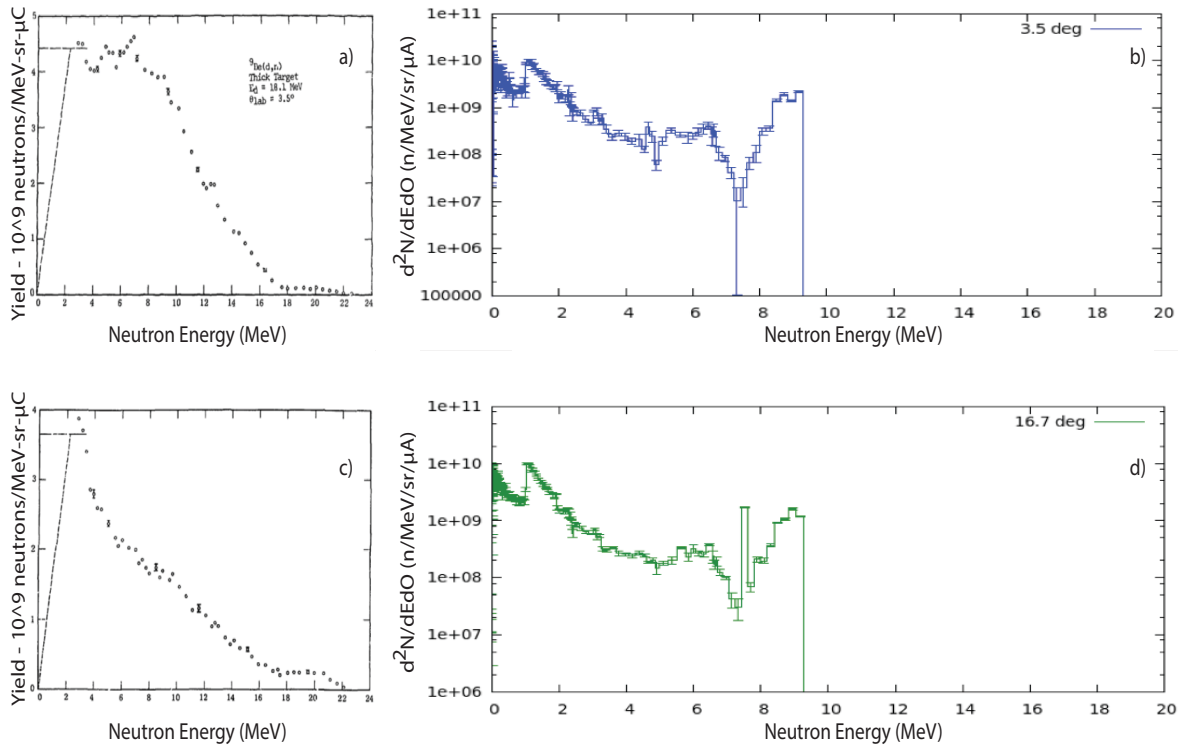


Figure 182: Deuterons on Be Target @ 3.5° (a-Data, b-FLUKA) & 16.7° (c-Data & d-FLUKA).

Spectrum of neutrons from 18.1 MeV deuterons on a thick target is shown in Figure 183.

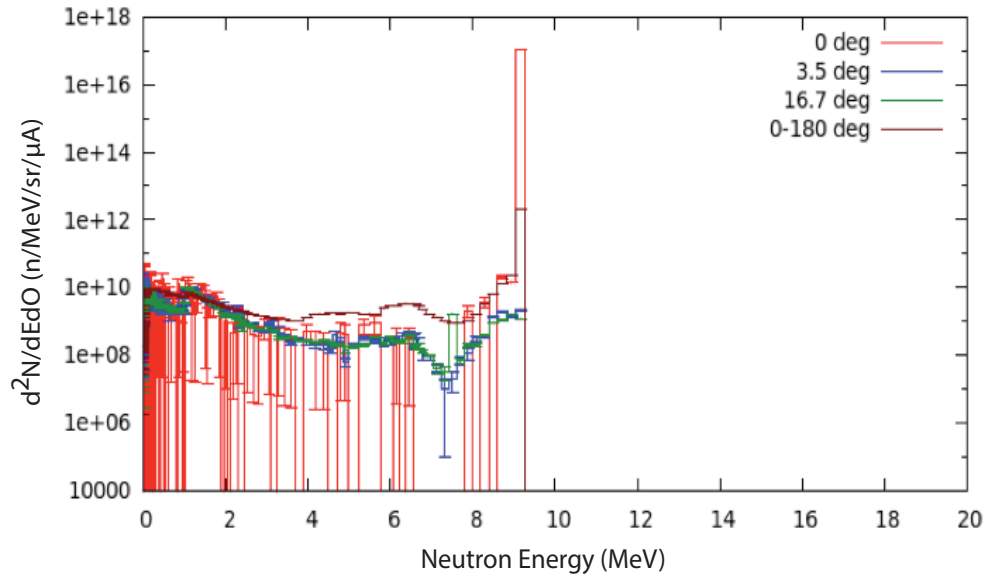


Figure 183: Deuterons on Be Target @ different angles (FLUKA).

In the same way as they were conducted in the experiment disclosed in Weaver KA, i.e., 19 MeV deuteron beam on a target lithium (disk diameter of 5 cm and 2 x 2.5 mm thick) and 18.1 deuteron beam into a carbon target (disk diameter of 5 cm and 6.4 mm thickness), FLUKA simulations were obtained @ 18.1 MeV deuterons to the same targets (Figure 184).

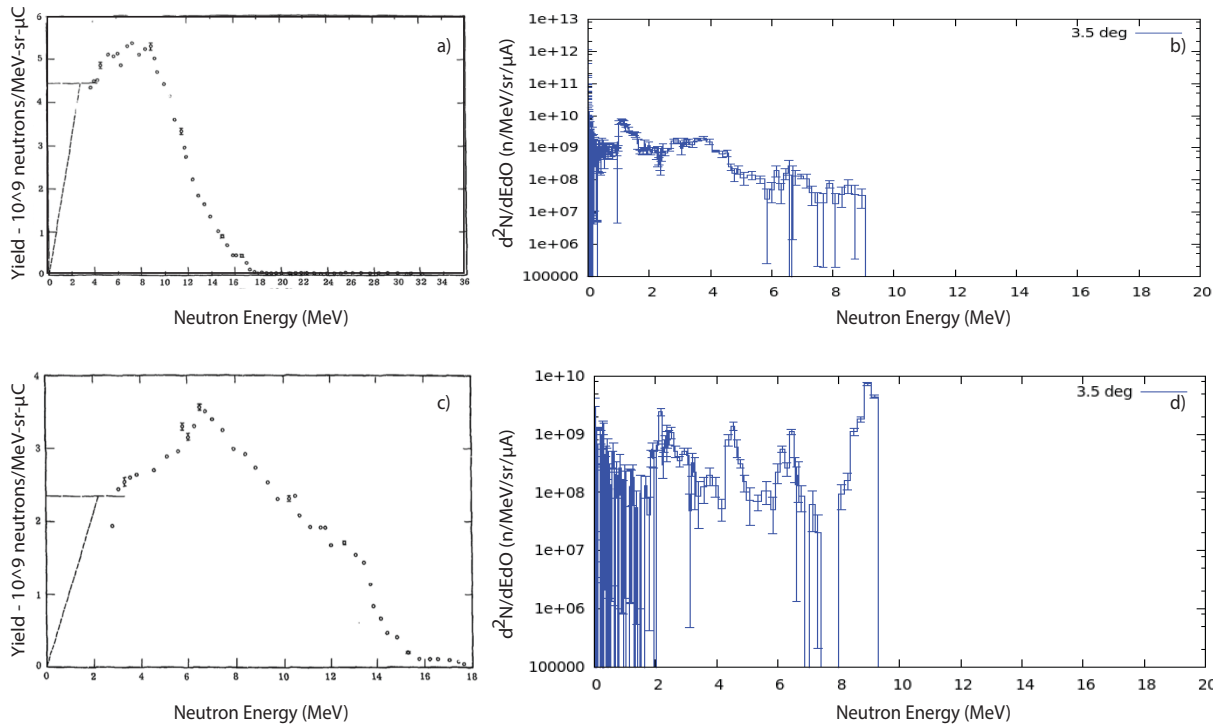


Figure 184: Deuterons on Li & C Targets @ 3.5° (a,c-Data, b,d-FLUKA).

Figure 185 shows the results of the experimental data and FLUKA at different angles.

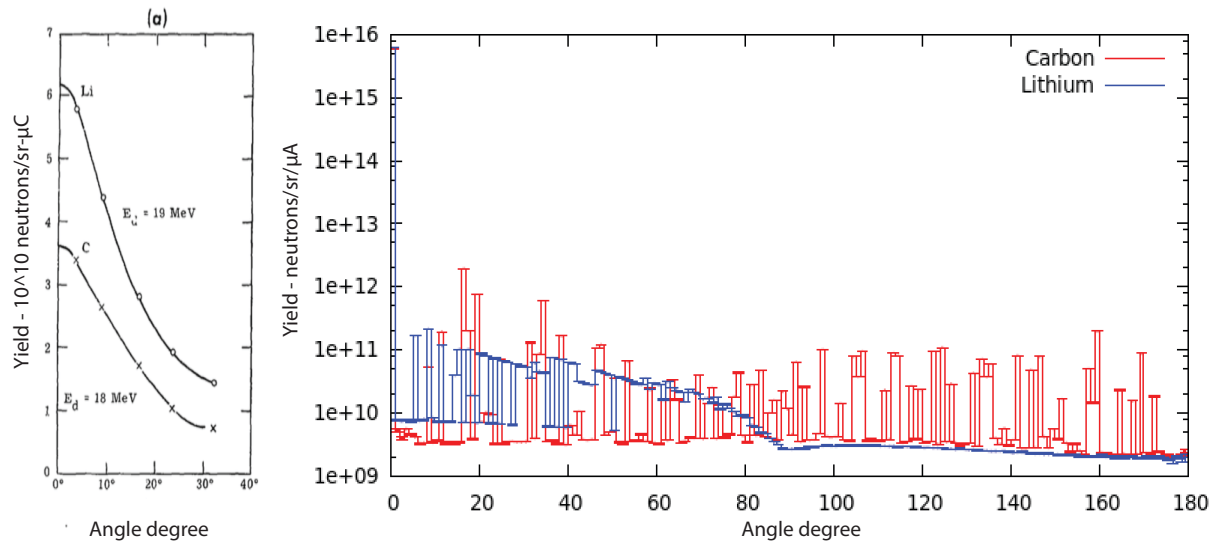


Figure 185: Deuterons on Li & C Targets @ different angles (Experiment & FLUKA).

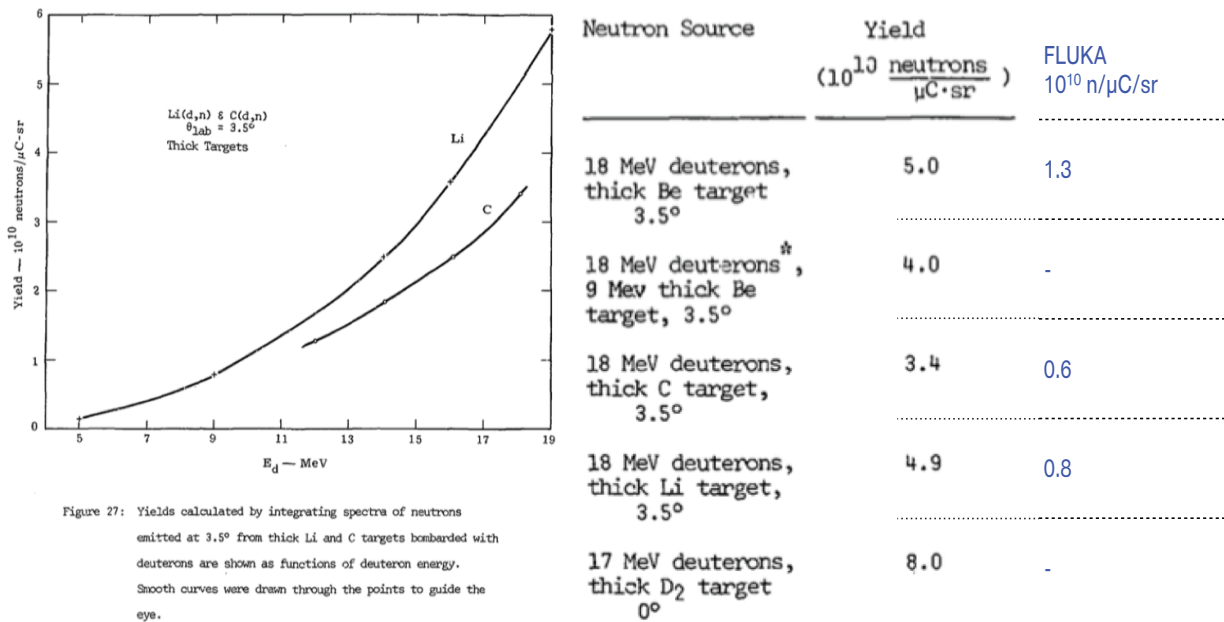


Figure 186: FLUKA deuteron beam simulation.

According to Figure 186 one can conclude as follow:

- FLUKA estimates the neutron yield much higher than the experiment data at 0 deg ($\sim 2.47 \times 10^{16}$ n/ $\mu\text{C}/\text{sr}$ for Be, $\sim 2.55 \times 10^{16}$ n/ $\mu\text{C}/\text{sr}$ for Li, and $\sim 2.41 \times 10^{16}$ n/ $\mu\text{C}/\text{sr}$ for C).
- FLUKA under-estimates the yield at 3.5 degrees and the other angles, except some abnormal yield peaks of existed angles.
- It seems like FLUKA does not work for deuteron beam in our energy region of interest.

Methods and Simulation Model

For the simulation, the geometry of the beryllium target and the detectors are shown in Figure 187. Target has 3 mm thickness and 2 cm diameter. The neutron fluxes were scored at ten laboratory angles between 0° and 90° ; 0° , 5° , 10° , 15° , 20° , 25° , 30° , 40° , 60° and 90° . The score regions are located 1 m from the target.

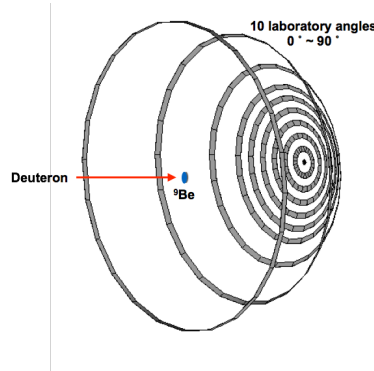


Figure 187: Geometrical model of calculation of neutron yields using GEANT4 and PHITS.

6.3 Deuteron benchmark results

MC calculation was performed with different physics models using GEANT4 and PHITS. These physics models are Binary Light Ion Cascade (“BLIC” on graph) and Liège intra-nuclear cascade model for GEANT4, and INCL with/without DWBA option and JQMD for PHITS. From Figure 188 until Figure 197 are shown the calculation results at from 0° until 90° .

INCL with DWBA option of PHITS is in good agreement with the Aoki’s experimental data. INCL without DWBA also describes the measurement data, but there are some differences around the incident energy.

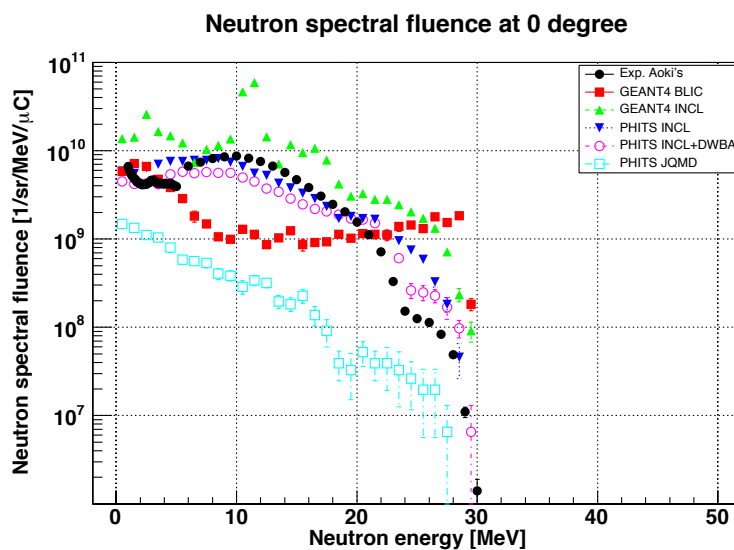


Figure 188: Neutron spectral fluence at 0° of $^9\text{Be}(d, xn)$.

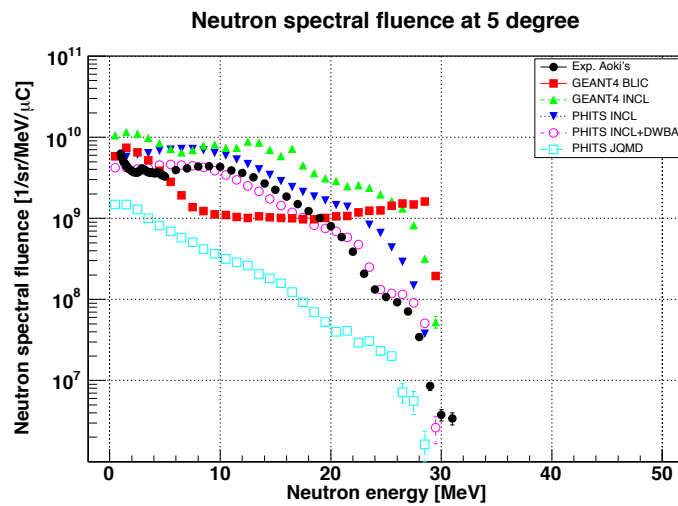


Figure 189: Neutron spectral fluence at 5° of ${}^9\text{Be}(d, xn)$.

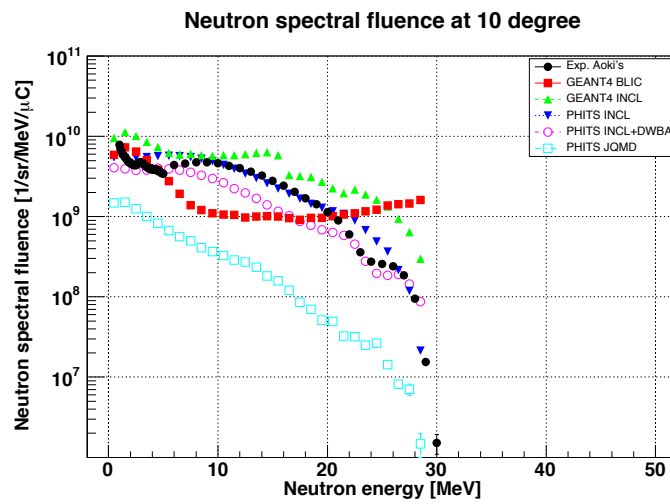


Figure 190: Neutron spectral fluence at 10° of ${}^9\text{Be}(d, xn)$.

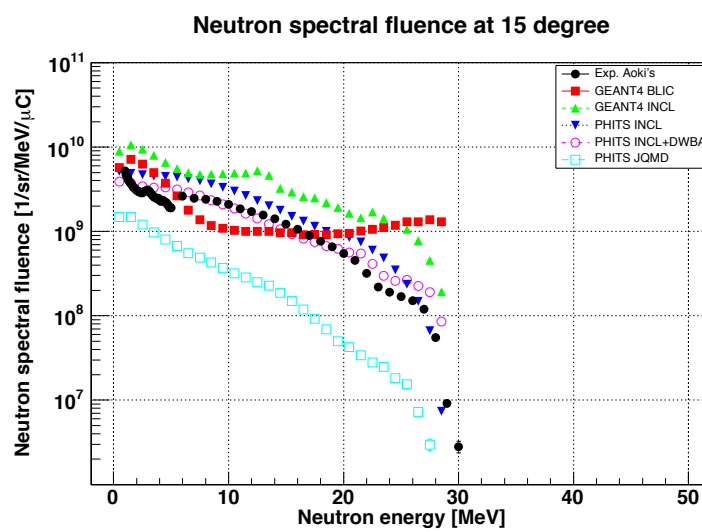


Figure 191: Neutron spectral fluence at 15° of ${}^9\text{Be}(d, xn)$.

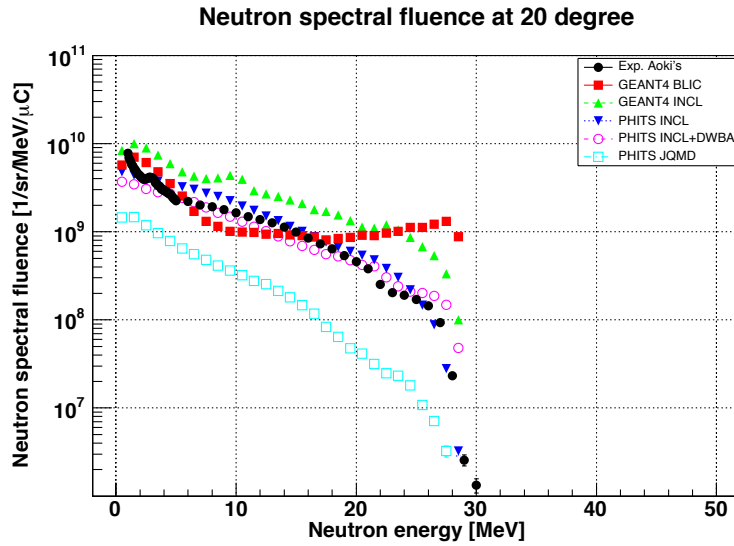


Figure 192: Neutron spectral fluence at 20° of ${}^9\text{Be}(d, xn)$.

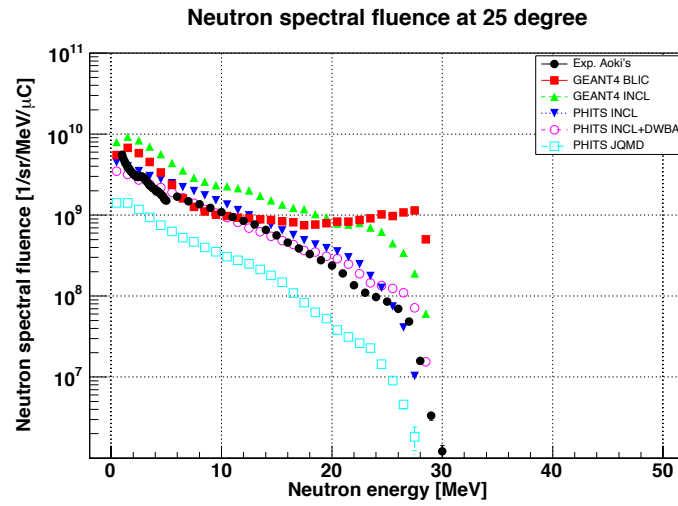


Figure 193: Neutron spectral fluence at 25° of ${}^9\text{Be}(d, xn)$.

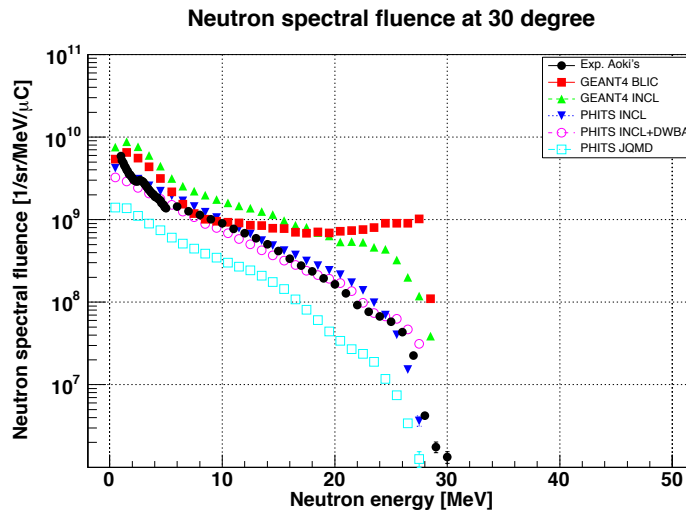


Figure 194: Neutron spectral fluence at 30° of ${}^9\text{Be}(d, xn)$.

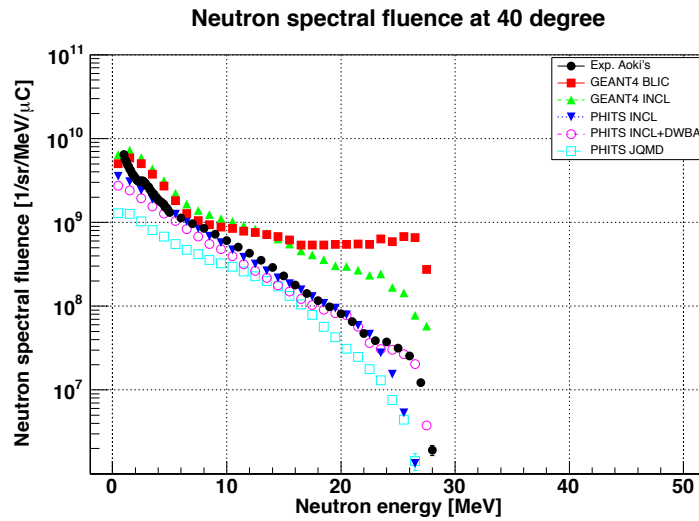


Figure 195: Neutron spectral fluence at 40° of ${}^9\text{Be}(d, xn)$.

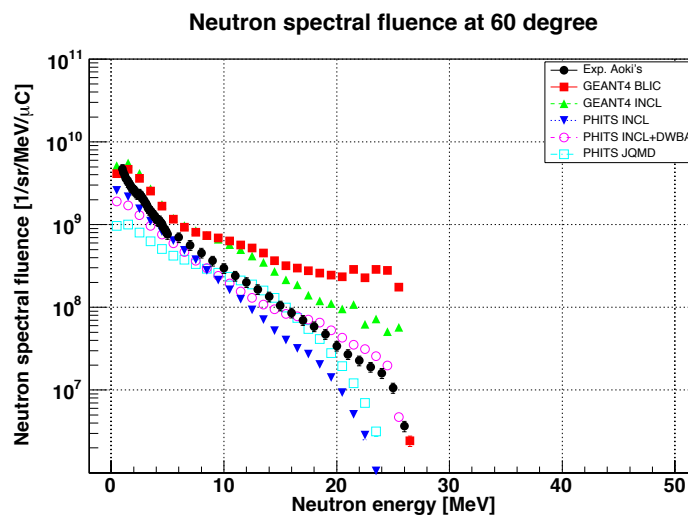


Figure 196: Neutron spectral fluence at 60° of ${}^9\text{Be}(d, xn)$.

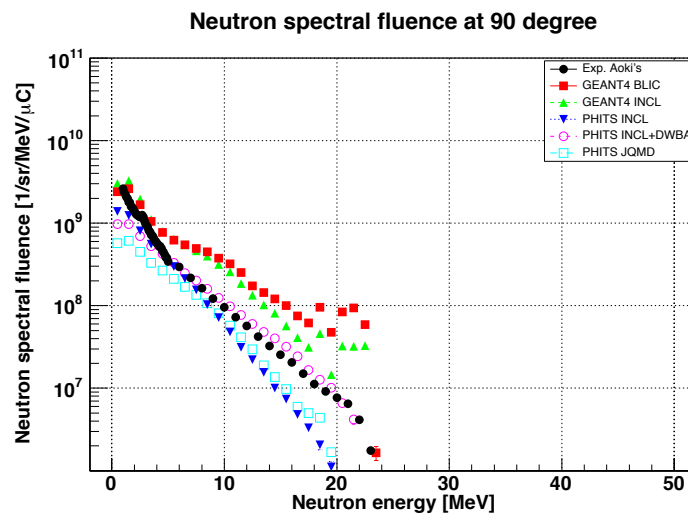


Figure 197: Neutron spectral fluence at 90° of ${}^9\text{Be}(d, xn)$.

7 FESTA benchmark with other Experiments

The main goal of the Activator is the production of ^{99}Mo and other medical isotopes, obtained by neutron capture reactions in the resonance region where the ARC method is applied. The cross section of $^{98}\text{Mo}(n, \gamma)^{99}\text{Mo}$ and other reactions is shown in Figure 198, where we can distinguish the major resonances peaks in the region of few eV. In the case of ^{98}Mo the major resonance peak is observed near to 130 barns, however for thermal neutron reaction is about 0.13 barns. A contribution to neutron capture reactions in the Activator, could also obtained considering the integral in the range of epithermal energies where all the resonances are located (10 eV-100 KeV), which is about 6.5 barns for ^{98}Mo . The energy range covered by FESTA Experiment is well visualized in Figure 199.

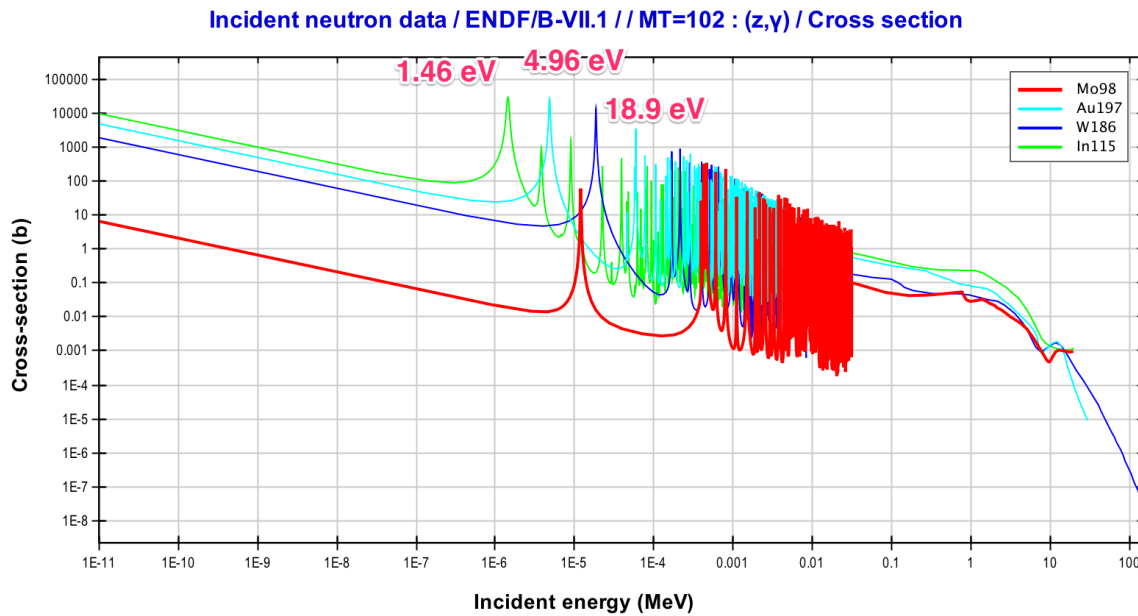


Figure 198: Neutron cross sections of ^{98}Mo , ^{197}Au , ^{186}W & ^{115}In .

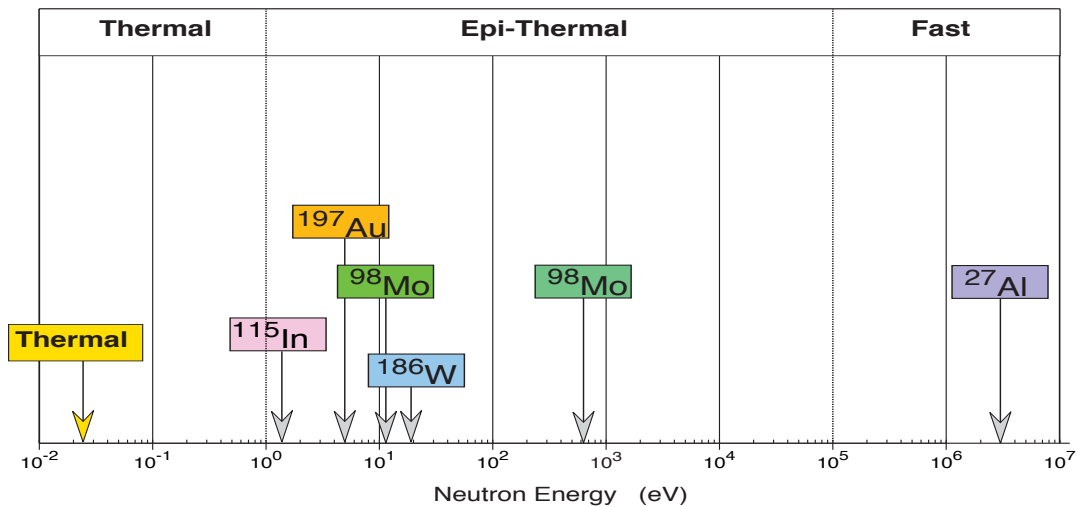


Figure 199: Energy range of FESTA Experiment.

7.1 TARC Experiment

The purpose of the TARC Experiment (Transmutation by Adiabatic Resonance Crossing), was to test directly the concept of ARC method to enhance significantly the neutron capture efficiency for Long lived Fission Fragments (LLFF), an idea of C. Rubbia (C. Rubbia, 1997), using a spallation neutron source driven by the 3.5 GeV proton beam from PS accelerator at CERN and 330 tons of lead with 99.99% purity.

The ARC method to be applied requires mandatory the presence of Lead as a diffuser medium. In relation to this some special behavior of neutrons in the lead can be noticed:

- Small average lethargy $\xi = [1 + \alpha/(1 - \alpha) \times \ln(\alpha)] \approx 9.6 \times 10^{-3}$, where $\alpha = [(m_{Pb} - m_n)/(m_{Pb} + m_n)^2]$, due to the high atomic mass of lead;
- A high and nearly energy independent elastic scattering cross section (Figure 200);
- A long “storage time because, below the capture resonances ($E_n \leq 1$ KeV) and down to epithermal energies, the elastic process is nearly isotropic and the transparency to neutrons is very high (it takes 3 ms, 1800 scatterings and a path in lead of 60 m to thermalize a 1 MeV neutron).

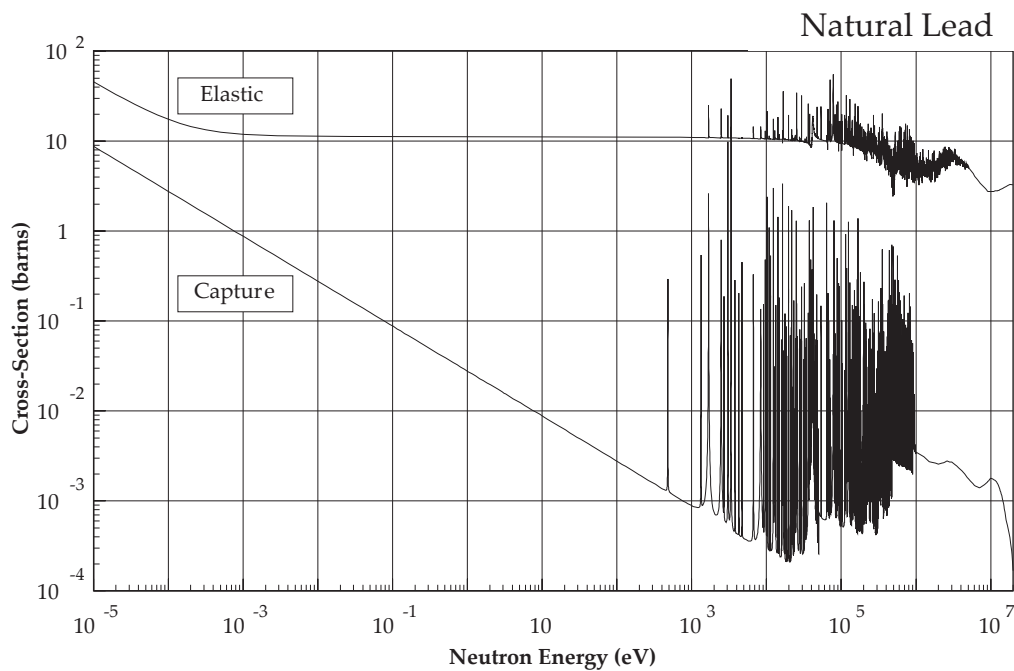


Figure 200: Cross sections of Lead.

Neutrons at relatively high energy ($E_n \approx$ few MeV), after being rapidly moderated by (n, xn), (n, n') reactions down to energies of a few hundred KeV, are slow down quasi adiabatically with small iso-lethargic steps and reach the capture resonance energy of an element to be transmuted or activated where they have a high probability to be captured. The resonance width is usually larger than the average lethargic step.

At low energies, below ~ 10 KeV, the neutron fluence, in good approximation, shows spherical symmetry and behaves approximately in the same way as if produced by a point-like source. This is, of course, precisely what is expected from the small elastic collision length in lead. The $dF/d[\ln(E)]$ distribution decreases very slowly, only one order of magnitude when the neutron energy decreases by four orders of magnitude (from 10 KeV to 1 eV). This is an important element in the demonstration of the efficiency of ARC method. The measurements performed with ^{99}Tc throughout the lead volume have provided self-consistency checks and confirmed the validity of ^{99}Tc cross-section data. From the excellent agreement with the simulation, the ARC effect was well confirmed.

The Activator claimed in the C. Rubbia CERN patent (C. Rubbia, 1997) considered as main parameter the angularly integrated neutron production rate. In fact, the energy spectrum of the initially produced neutrons is relatively unimportant, however the inelastic processes in the Diffuser quickly damp the neutron energy down to about 1 MeV, where the lethargy slow-down of the neutrons is taking over. Therefore, the neutrons capture efficiency for activation and the geometry of the Activator are relatively independent of the details of the realization of the source. So as conclusion, the activation of medical isotopes such as ^{127}I , ^{98}Mo , ^{130}Te and ^{129}Xe can be conducted at their exposure to a neutron production rate of few times 10^{13} neutrons over the full solid angle. According to what is reported in the Esposito report based on the C. Rubbia Patent (Figure 201), for ^{98}Mo requires a neutron source yield of some units in 10^{14} s^{-1} in order to obtain 5-10 mCi/g ($\sim 21\text{-}42 \text{ mCi/g}$ for 100% enrichment).

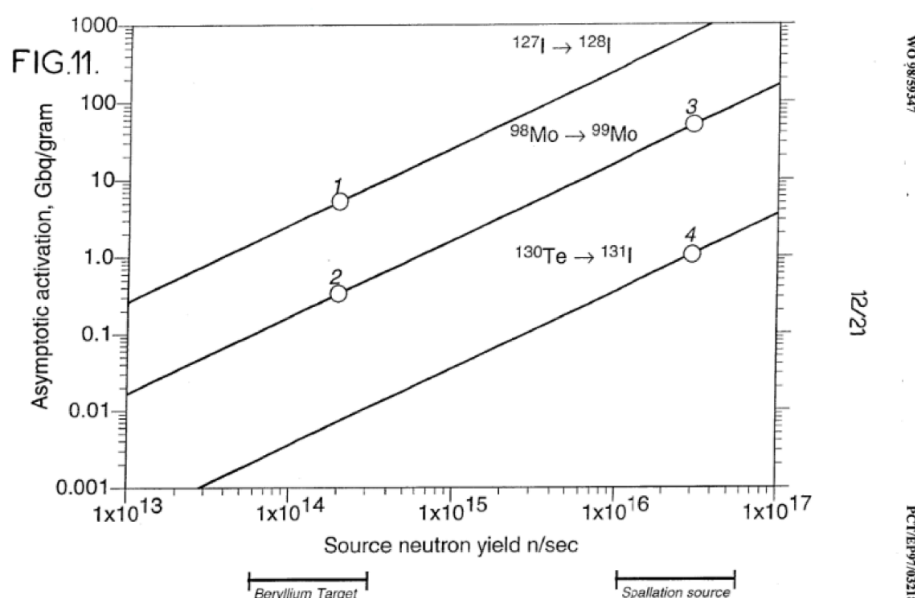


Figure 201: Specific activity at saturation conditions expected for $^{98}\text{Mo}(n, \gamma)^{99}\text{Mo}$ neutron capture reaction.

7.2 Accelerator-driven, neutron activator at Louvain-la-Neuve (LLN)

A lead assembly has been totally designed and constructed at the University of Louvain (UCL- Belgium) and tested; with a 65 or 75 MeV proton beam CYCLONE 110.

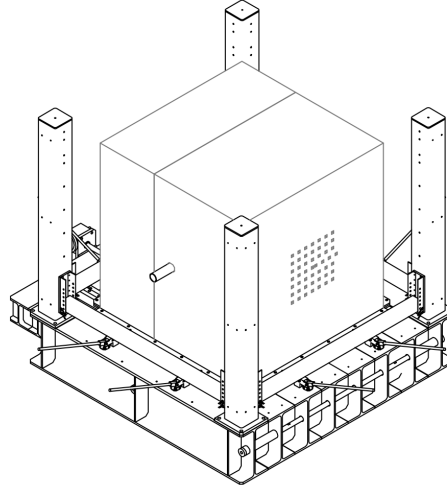


Figure 202: General view of the lead assembly. The holes for the introduction of solid samples and the beam transport are also shown.

The lead assembly weighs 47 tons, has a $1.6 \times 1.6 \text{ m}^2$ basis and a 1.63 m height. The Be target (diameter: 40.3 mm, thickness: 36.7 mm) is placed at the center of the assembly, at the end of the proton beam transport tube.

The experimental activity ($\mu\text{Ci/g}/\mu\text{A}$) for each foil is presented in Table 65.

Table 65: Experimental Activity in the LLN Activator

	$R = 7.9 \text{ cm}$ $\theta = 82.2^\circ$		$R = 11.5 \text{ cm}$ $\theta = 48.6^\circ$		$R = 19.8 \text{ cm}$ $\theta = 39.9^\circ$		$R = 30.0 \text{ cm}$ $\theta = 120.4^\circ$	
	Activity ($\mu\text{Ci/g}/\mu\text{A}$)	Error (%)	Activity ($\mu\text{Ci/g}/\mu\text{A}$)	Error (%)	Activity ($\mu\text{Ci/g}/\mu\text{A}$)	Error (%)	Activity ($\mu\text{Ci/g}/\mu\text{A}$)	Error (%)
¹⁹⁸ Au								
Outer foil 1	1403.9	11.2	1399.6	10.2	1363.5	10.2	1308.8	11.2
Inner foil	1179.8	11.2	1142.4	10.2	1163.4	10.2	1106.6	11.2
Outer foil 2	1426.7	11.2	1385.2	10.2	1380.5	10.2	1343.5	11.2
¹⁸⁷ W								
Outer foil 1	156.6	10.2	146.8	10.1	148.9	10.1	139.8	10.2
Inner foil	127.0	10.3	121.1	10.1	121.5	10.1	123.9	10.3
Outer foil 2	157.4	10.2	147.2	10.1	146.4	10.1	142.6	10.2
¹¹⁶ In								
Outer foil 1	—	—	1826.1	10.2	—	—	1778.0	10.2
Inner foil	—	—	1495.5	10.1	—	—	1452.6	10.2
Outer foil 2	—	—	1890.4	10.1	—	—	1744.9	10.2

For small distances (less than about 10 cm) between the target and the ^{Nat}Mo sample, the effect of the polar angle is strongly marked otherwise the angle effect is less important and the ⁹⁹Mo activity produced in each sample is quite independent on the distance. This activity is of the order of 13mCi/g/mA.

7.3 Accelerator-driven, neutron activator at Joint Research Centre (JRC Ispra, Italy)

A compact, accelerator-driven, neutron activator has been tested at the Scanditronix MC40 Cyclotron of the Joint Research Centre (JRC Ispra, Italy) to explore the feasibility of activation of Re (^{186}gRe , $T_{1/2} = 89.25\text{h}$, ^{188}gRe , $T_{1/2} = 16.98\text{h}$) and Ho (^{166}Ho , $T_{1/2} = 26.80\text{h}$).

The activator prototype has been installed on a dedicated beam line of the Scanditronix MC40 Cyclotron of the Institute for Health and Consumer Protection (IHCP) at JRC Ispra, the nominal beam characteristics of which are given in Table 66.

Table 66: Beam characteristics

Particle	Energy (MeV) (min.-max.)	Maximum Extracted Current (μA)
p	8 – 40	60
d	8 – 40	30
4He^{2+}	8 – 53	30
3He^{2+}	4 – 20	60

Figure 203 shows the neutron activator configuration proposed by JRC Ispra Lab.

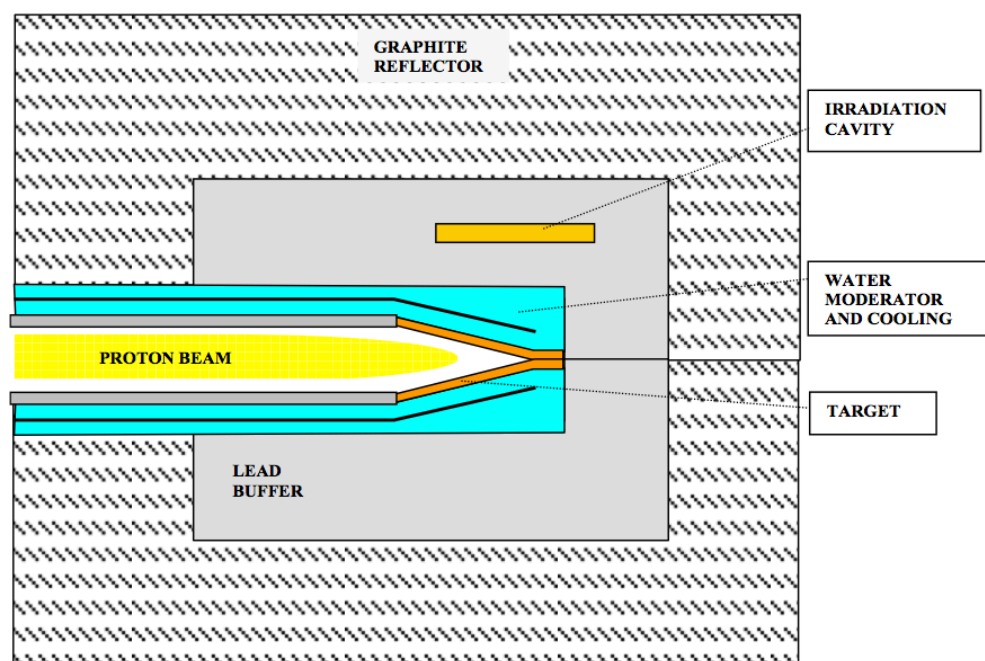


Figure 203: Activator configuration proposed for the JRC Ispra Lab.

The target system (Be target and water cooling sheath) is 40cm long and 83 mm in diameter. It is inserted in a 30 x 30 x 30 cm³ high-purity lead block which is itself in a center of a cube of nuclear-grade graphite, bringing the outer dimensions of the activator to 65x65x65cm³.

A reference simulation of the complete activator with a proton beam of 36 MeV estimates the average neutron flux in the lead volume at $5.07 \times 10^8 \text{ n cm}^{-2}\text{s}^{-1}\mu\text{A}^{-1}$ ($1.77 \times 10^{10} \text{ n cm}^{-2}\text{s}^{-1}$ with 35 μA beam current), whereas the neutron flux in an irradiation channel has been estimated at $1.26 \times 10^9 \text{ n cm}^{-2}\text{s}^{-1}\mu\text{A}^{-1}$ ($4.42 \times 10^{10} \text{ n cm}^{-2}\text{s}^{-1}$ with 35 μA).

The yields for Au, Al, Mo, Ho and Re metallic foils, irradiated in the fully constructed system, are reported in Table 67.

Table 67: Activation yields of Au, Al, Mo, Ho and Re foils (in kBq.g-1. μA -1.h-1).

Reactions	MCNPX yield	Exp. Yield (Ispra)	Exp. yield (LLN)
$^{197}\text{Au}(\text{n},\gamma)^{198}\text{Au}$	554		
$^{197}\text{Au}(\text{n},2\text{n})^{196}\text{Au}$	2		
$^{197}\text{Au}(\text{n},2\text{n})^{196\text{g}}\text{Au}$		1.58	
$^{197}\text{Au}(\text{n},\gamma)^{198\text{g}}\text{Au}$		972	526. 99 (194.46)
$^{98/100}\text{Mo}(\text{n},\gamma/2\text{n})^{99}\text{Mo}$	0.3	7	5.02 (1.85)
$^{165}\text{Ho}(\text{n}, \gamma)^{166}\text{Ho}$	1646		
$^{165}\text{Ho}(\text{n}, \gamma)^{166\text{g}}\text{Ho}$		2520	
$^{185}\text{Re}(\text{n}, \gamma)^{186}\text{Re}$	246		
$^{187}\text{Re}(\text{n}, \gamma)^{188}\text{Re}$	1142		
$^{185}\text{Re}(\text{n}, \gamma)^{186\text{g}}\text{Re}$		385	
$^{187}\text{Re}(\text{n}, \gamma)^{188\text{g}}\text{Re}$		2100	
$^{27}\text{Al}(\text{n},\alpha)^{24\text{g}}\text{Na}$		5.37	

The values in parentheses are the Louvain-la-Neuve yields modified to take into account the difference in proton beam energy

7.4 $^{99}\text{Mo}/^{99\text{m}}\text{Tc}$ production systems at INFN Legnaro Laboratories

At the INFN Legnaro National Laboratories (Padova), two more proton accelerators are expected to be installed and put into operation:

1. A cyclotron of 40-70 MeV and a relatively high beam current up to 500 mA (maximum power 35 kW).
2. A high intensity RFQ (Radio Frequency Quadrupole) type linac, with fixed 5 MeV output energy and beam current ranging from 20 to 50 mA (maximum power 250 kW).

These accelerators have performances for the production of some medical radioisotopes in particular ^{99}Mo ($^{99\text{m}}\text{Tc}$), either by exploiting the direct (p,xn) reaction channel, or by the usual (n, xn) and (n, γ) neutron capture reactions.

Both accelerators are included as main drivers in two research projects:

1. **SPES**, aims at constructing a Radioactive Ion Beam's (RIB's) facility dedicated to the nuclear physics research of unstable nuclei, far from stability valley.
2. **TRASCO-IFMIF** is instead focused on the field of applied nuclear physics of the future fusion, as well as Generation-IV fission reactors.

Clearly then, according to the type of accelerators that will be installed at LNL, that the intention is to achieve a high neutron yield of some units in 10^{14} s^{-1} , using a thick-target neutron yield from Be (p, xn) reaction.

7.4.1 ^{99}Mo production based on TRASCO RFQ-driven neutron activator

The purpose is to provide an epithermal neutron spectrum inside the sample holder volume in order to maximize the $^{98}\text{Mo}(\text{n}, \gamma)$ neutron capture reactions in the resonance energy region by using a quite intense proton beam (30 mA) of low energy (5 MeV) and a Be-based high power neutron converter.

Figure 204 shows the neutron activator configuration proposed based on the TRASCO RFQ accelerator and the beryllium neutron converter.

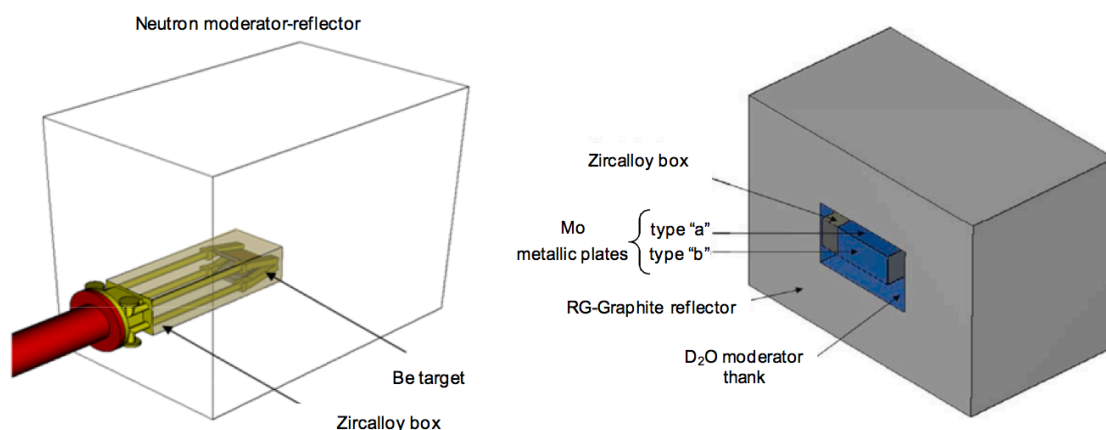


Figure 204: Activator configuration proposed for the TRASCO RFQ accelerator.

Under these conditions, just a small Lead volume is needed to slow neutrons, generated by the beryllium target, down to epithermal neutron source. In such a way neutron captures, as well as losses are minimized during the partial slowing down process.

Moderator

It is very interesting to appreciate that the Be target is located inside a D₂O (could be also Lead) filled tank which acts as main moderator medium (Figure 205). The parameter considered for the moderator volume optimization is the thickness "d" around the target. Possible locations for the ⁹⁸Mo samples to be irradiated are on the Zircalloy box walls around Be target (Pos 1), or above the moderator side wall (Pos 2).

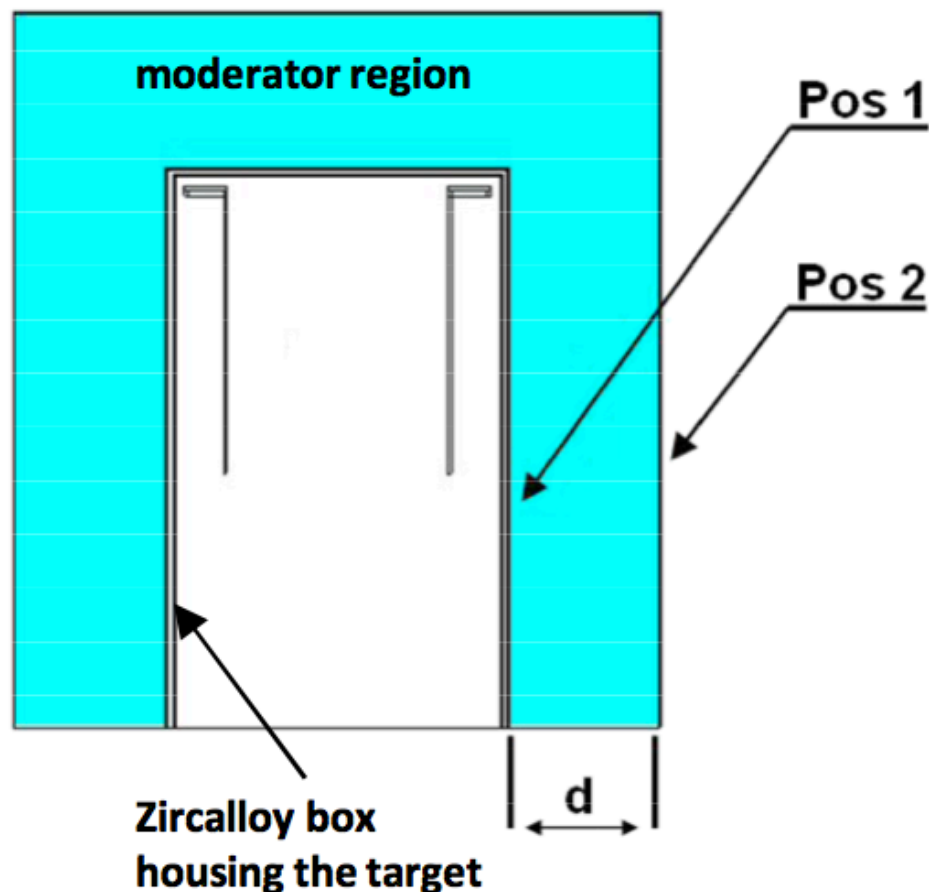


Figure 205: Moderator region around the Be neutron converter.

Different thicknesses have been simulated with MCNPX. Two ⁹⁸Mo samples (<1 mm thickness) positions (1 & 2) have been tested. The maximum epithermal fluence level was reached using D₂O with a thickness of 10 cm in position 1. For Pb the epithermal fluence in position 1 was twice lower and saturates when the thickness was greater than 30 cm. By applying a spectral selectivity in the epithermal energy range the performance has been improved using D₂O material with 8 cm thickness.

Reflector

One of the proper materials having good albedo properties, low cost, and relatively easy procurement, is Reactor Grade (RG)-graphite. The D₂O volume is surrounded by a RG-graphite reflector. The 100% enriched ⁹⁸Mo thin plate samples are supposed to lay down on the Zircalloy box. Those referred to as "type a" (located on the top and bottom surfaces), while those indicated as "type b" (on right-left walls). The results are shown in Figure 206, normalized per unit volume and per source neutron, versus the RG-graphite thickness surrounding the moderator.

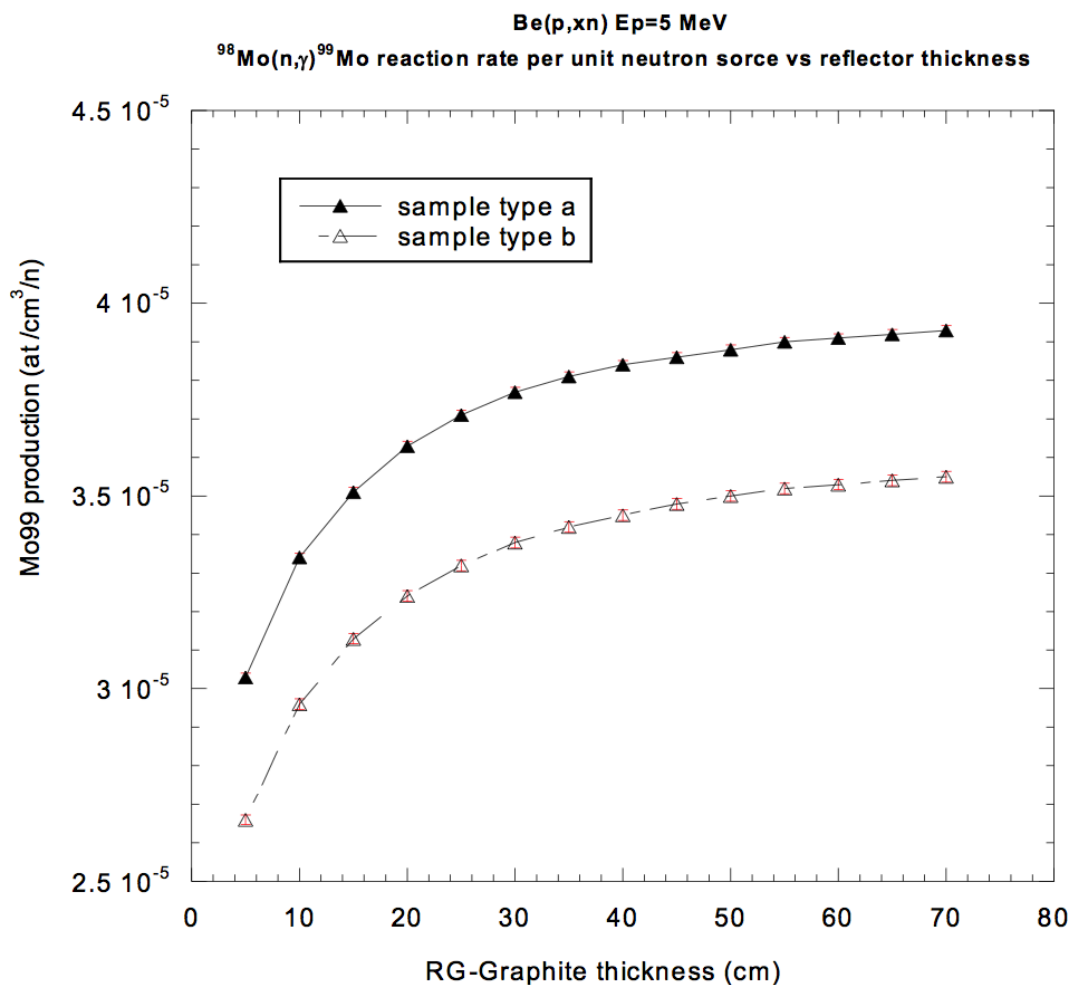


Figure 206: ⁹⁸Mo(n, γ)⁹⁹Mo reaction production.

It is very important to remark the different ⁹⁹Mo production in both cases. Neutrons passing through "type a" ⁹⁸Mo samples, experience a higher probability of being further slowed down from the Be target, thus improving the radioactive capture process. The ⁹⁸Mo "type b" samples are instead mainly crossed by neutrons without being further moderated by the Be target. From the results is concluded that the minimum required RG-graphite thickness is 40 cm.

The calculated normalized neutron fluence is shown in Figure 207. The resulting neutron spectrum in the may considered as the optimal one, which integral is $5.5 \times 10^{-4} \text{ cm}^{-2}$. Considering that the TRASCO RFQ + Be target system is expected to generate, a neutron source intensity around $1.05 \times 10^{14} \text{ s}^{-1}$, an epithermal neutron fluence rate $\Phi_{\text{epi}} \sim 6.0 \times 10^{10} \text{ cm}^{-2}\text{s}^{-1}$ would be available in the aforementioned sample position.

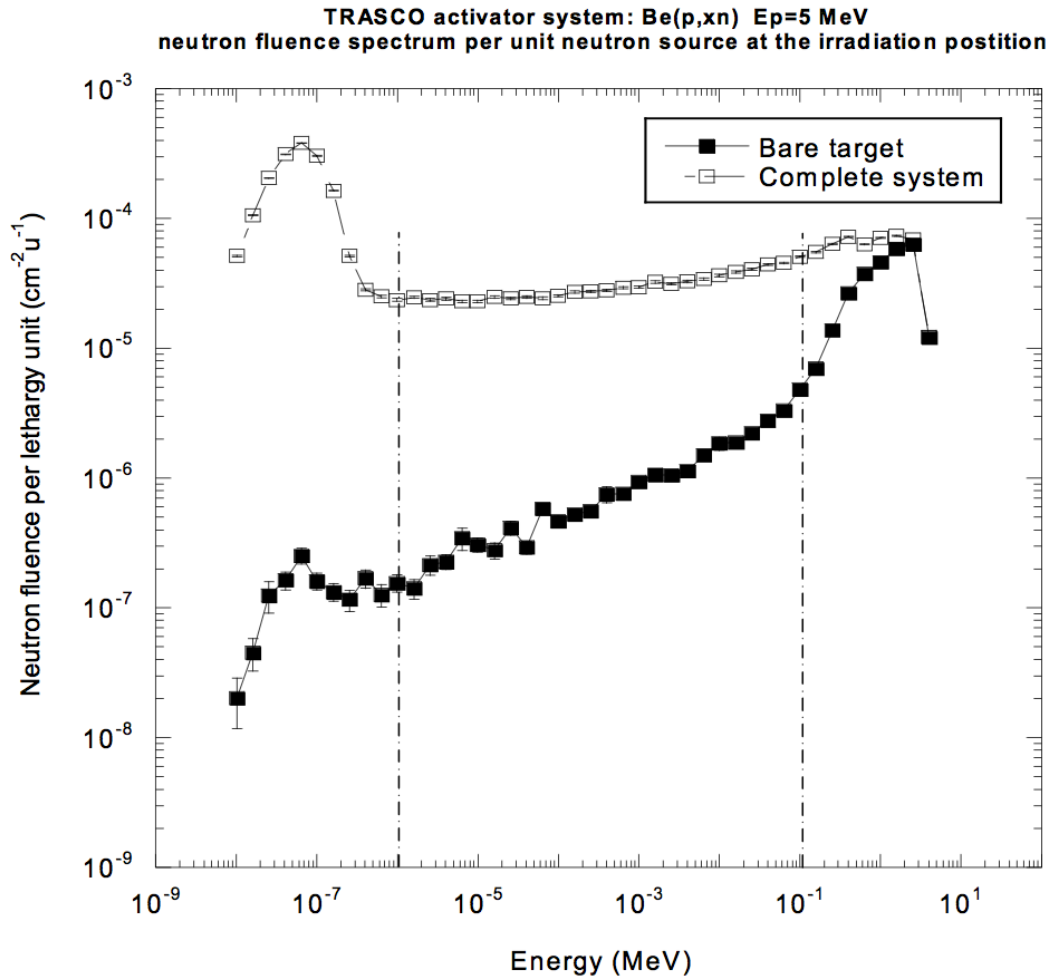


Figure 207: Normalized neutron fluence spectra comparison which would be available at the “type a” irradiation position.

Benchmark with JRC Ispra Lab Activator

In order to assess the proposed activator system based on TRASCO RFQ accelerator, a comparison has been performed with the system developed at the JRC Ispra Lab, based on a cyclotron-driven 40 MeV proton beam. By integrating the spectra plots between the 1 eV - 100 KeV (Figure 208), the resulting normalized neutron fluence level for the JRC activator is better than the proposed one by about a factor 70.

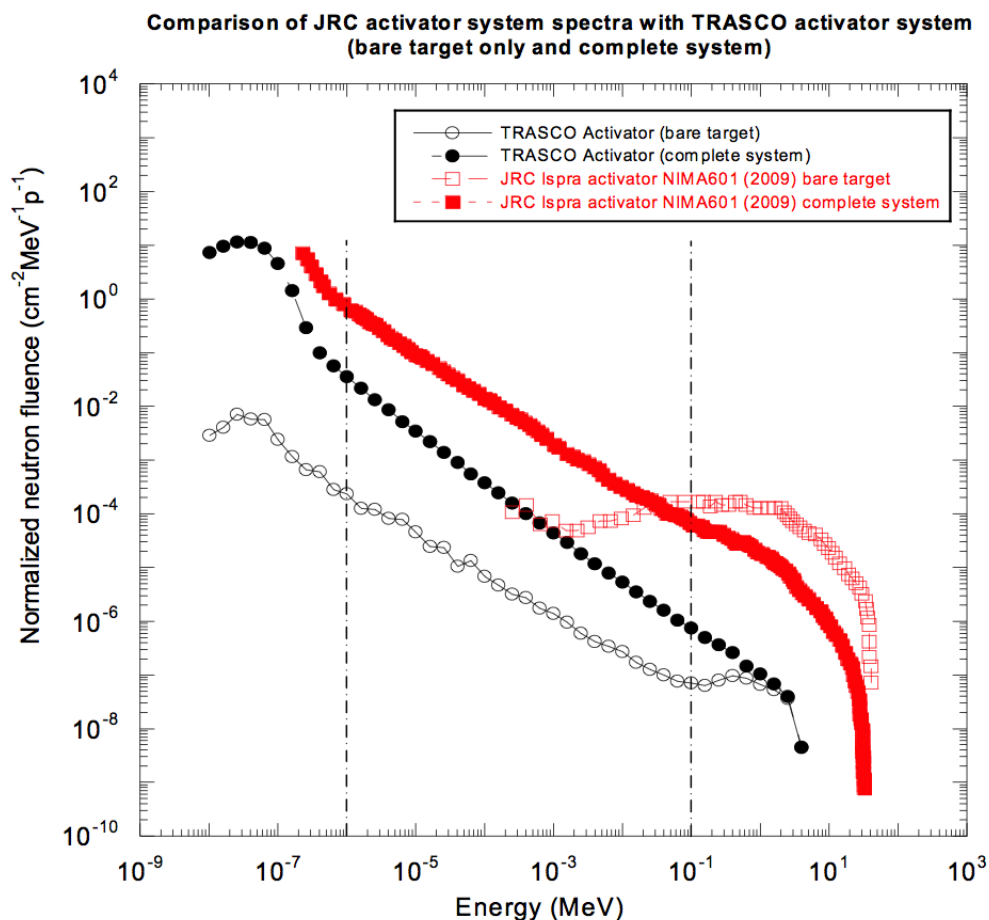


Figure 208: Neutron fluence spectra comparison between the JRC activator system (red plots) and the proposed system based on TRASCO RFQ accelerator (black plots).

Such a difference could be explained as follow: At 40 MeV proton beam the neutron yield is $2.09 \times 10^{11} \text{ s}^{-1}\cdot\mu\text{A}^{-1}$ compared to $3.5 \times 10^9 \text{ s}^{-1}\cdot\mu\text{A}^{-1}$ available with 5 MeV proton beam, which is a factor 60.

TRASCO RFQ-driven neutron activator performance

The main reference parameters and the calculated values aimed at the final estimations of ^{99}Mo specific activity expected both after the first 24 h irradiation and at saturation, are listed in the following table.

Table 68: Activator System based on TRASCO RFQ + Be target

TRASCO RFQ output energy	E_p	5 MeV
Proton and Deuteron beam current	I_p	30 mA
Beam power on the target		150 kW
Neutron converter material		Be
Estimated Total neutron source		$1.05 \times 10^{14} \text{ s}^{-1}$
Epithermal neutron fluence rate (1eV – 100 KeV)	F_{ep}	$5.8 \times 10^{10} \text{ cm}^{-1}\text{s}^{-1}$
Total neutron fluence rate at sample position	Φ_{tot}	$3.05 \times 10^{11} \text{ cm}^{-1}\text{s}^{-1}$
Irradiated sample: ^{98}Mo enrichment	%	100
Production route		$^{98}\text{Mo}(n, \gamma)^{99}\text{Mo}$
Estimated total Reaction Rate (RR) inside sample A5		$4.03 \times 10^9 \text{ cm}^{-1}\text{s}^{-1}$
RR fraction due to epithermal neutrons (1eV-100 KeV)	%	88.5
^{99}Mo Specific Activity at saturation	As	10.6 mCi/g
^{99}Mo Specific activity	As	2.36 mCi/g
^{98}Mo bulk mass requested for 1 Ci ^{99}Mo	m	423 g

7.4.2 ^{99}Mo production based on SPES cyclotron-driven neutron activator

The purpose is the ^{99}Mo production at LNL taking into account an activator system driven by the high performance (40-70 MeV, 500 mA max beam current), proton cyclotron of the future SPES project.

The activator system is basically a scaled-up version of the one already developed at the JRC Ispra and at Louvain-la-Neuve laboratory and is schematically shown in Figure 209.

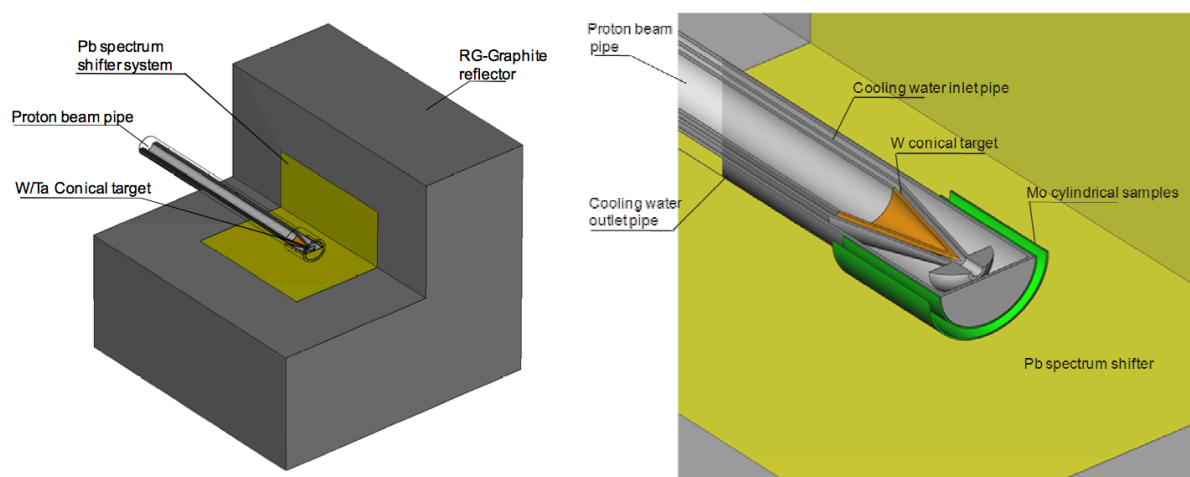


Figure 209: Proposed neutron activator system based on the SPES cyclotron-driven proton accelerator.

In the present study a tungsten neutron converter has been taken into account instead of a beryllium target, because of higher neutron yield efficiency (20-50% better in the 40-70 MeV range) and more favorable lower mean energy spectrum from source neutrons, which are moreover yielded in a roughly isotropic way. The neutron converter modeled, similar to the one developed for the neutron activator system at the JRC Ispra, is of conical shape and light-water cooled. It is located within a Pb- volume acting as neutron buffer.

Moderator

The calculated normalized neutron fluence, as well as the spectral ratio for the epithermal neutrons are plotted against the half-length of Pb cubic buffer, which is representative of the moderator thickness in the radial direction (Figure 210).

The results point out that the spectral ratio approaches the asymptotic value of about 0.26, when the cubic lead buffer reaches about 1.5 m sizes. Such a result is similar to what reported in the paper by Froment et al. (P. Froment, 2002), although the mass required turns out to be about 38 tons. However, since the epithermal spectral ratio improvement is about 8% only (corresponding to $\sim 10\%$ fluence drop) in the last 45 cm outer thickness, the final configuration for the cubic Pb buffer can be limited to 60 cm size, thus greatly reducing the required mass to about 2.5 tons.

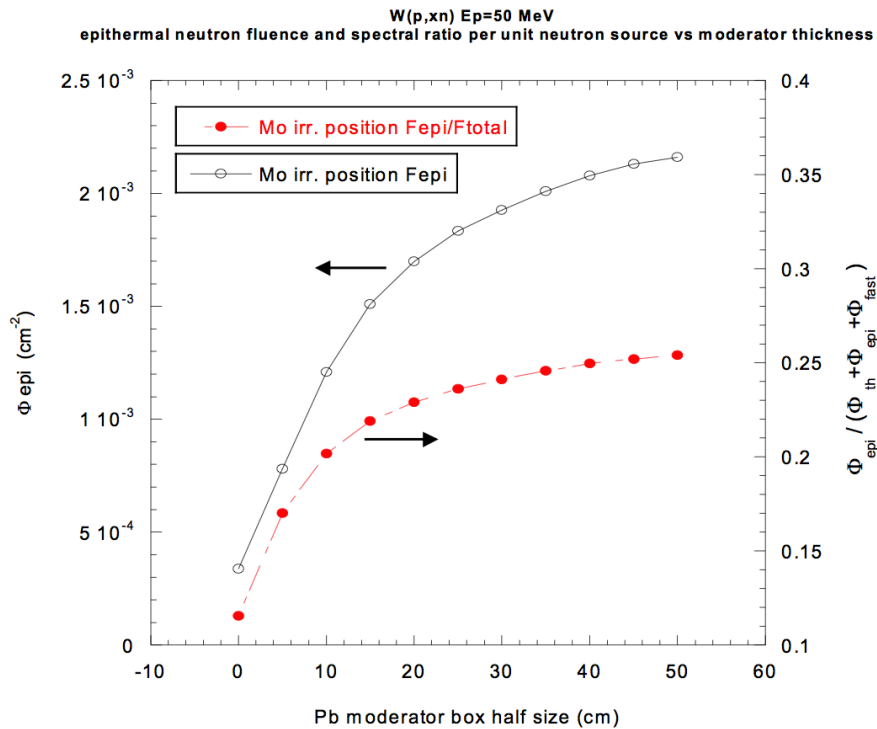


Figure 210: Normalized neutron fluence (void circle black curve) and related spectral ratio (full circle red curve). The x-axis parameter is the half of Pb buffer cubic size around the target.

Reflector

The optimal thickness of RG-graphite reflector was estimated, by calculating the volume-averaged ^{99}Mo production rate, expected at saturation, in a ^{98}Mo -enriched cylindrical sample placed at the irradiation site. The results are plotted in Figure 211 and the minimum RG-graphite size required is at least 20 cm.

The normalized neutron fluence spectra, calculated at the sample irradiation position, are shown in Figure 212. The resulting spectral configuration for such a system is optimized, having a nearly flat trend in the 1 eV-100 KeV energy range, which integral is $2.19 \times 10^{-3} \text{ cm}^{-2}$. Reminding that the neutron source yields expected by the SPES-cyclotron-driven W neutron converter under operating conditions is $2.3 \times 10^{14} \text{ s}^{-1}$, an epithermal neutron flux level $\Phi_{\text{epi}} \sim 5.0 \times 10^{11} \text{ cm}^{-2}\text{s}^{-1}$ would be reached in the sample irradiation position.

To test the effectiveness of the proposed activator system based on the SPES cyclotron, a performance comparison with the aforementioned activator system at JRC Ispra lab was performed as well. The spectral characteristics are shown in Figure 212. By integrating the normalized neutron fluence plots in the 1 eV–100 KeV energy intervals, the configuration proposed turns out to be about a factor of 3 better than the JRC one.

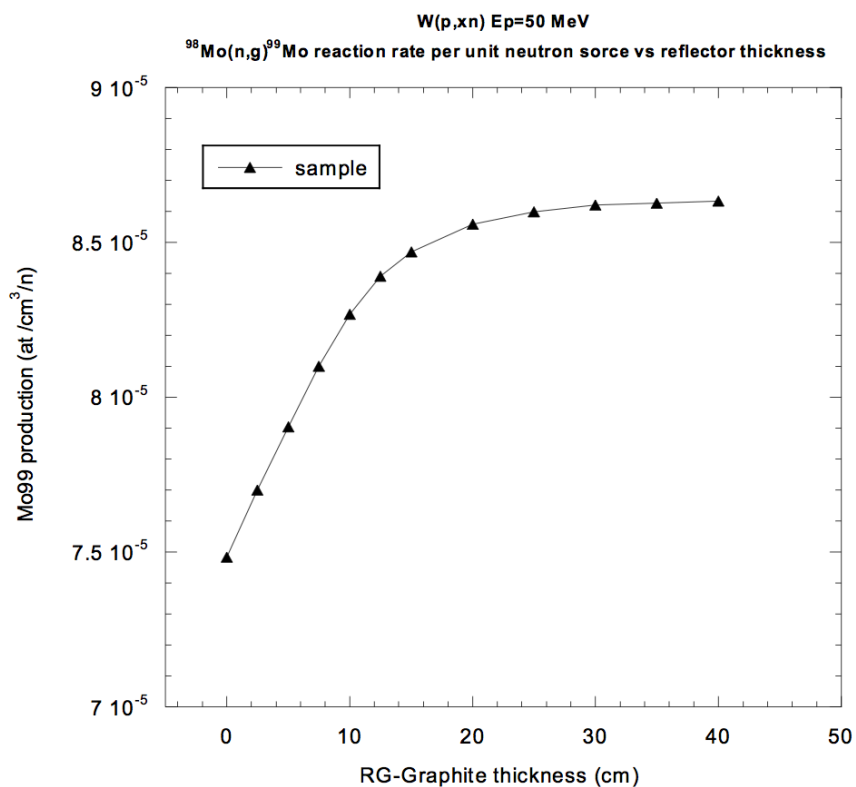


Figure 211: Normalized ⁹⁹Mo production expected by (n, γ) reactions on ⁹⁸Mo (100% enrichment) samples versus the RG-graphite reflector thickness.

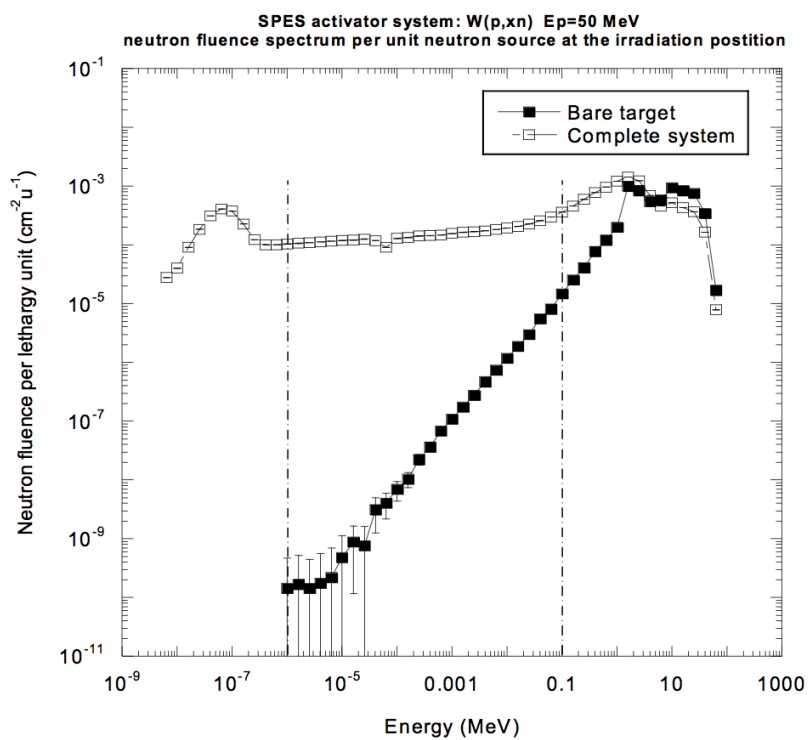


Figure 212: Normalized neutron fluence spectra.

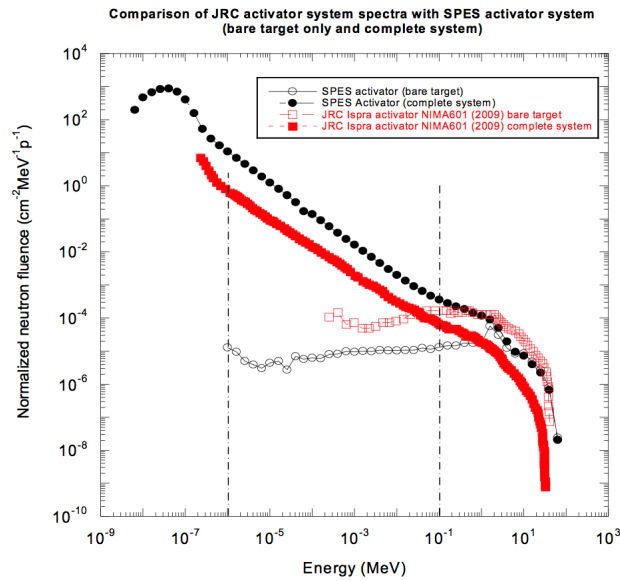


Figure 213: Neutron fluence spectra comparison between the JRC activator system (red square plots) and the proposed system based on the SPES cyclotron (black circle plots).

Such a better performance depends (about 2/3 out of the total contribution) on the improved n/p conversion efficiency, corresponding to 4.51×10^{11} n/s·μA, which can be obtained from W target hit by 50 MeV proton beam. The neutron yield from Be target under 40 MeV proton beam is 2.09×10^{11} n/s·μA. The remaining part (approximately 1/3 out of the total) is instead from the improved system efficiency.

SPES cyclotron-driven neutron activator performance

The main reference parameters and the estimated in-target specific activity calculated either after the first 24 hours of irradiation is listed in the table.

Table 69: Activator System based on SPES cyclotron ($E_p=50$ MeV) + W target

SPES cyclotron output energy	E_p	50 MeV
Proton and Deuteron beam current	I_p	0.5 mA
Beam power on the target		25 kW
Neutron converter material		W
Estimated Total neutron source		$2.3 \times 10^{14} \text{ s}^{-1}$
Epithermal neutron fluence rate (1eV – 100 KeV)	Φ_{ep}	$5.0 \times 10^{10} \text{ cm}^{-2} \text{ s}^{-1}$
Total neutron fluence rate at sample position	Φ_{tot}	$1.9 \times 10^{12} \text{ cm}^{-2} \text{ s}^{-1}$
Irradiated sample: ^{98}Mo enrichment	%	100
Production route		$^{98}\text{Mo}(n, \gamma)^{99}\text{Mo}$
Estimated total Reaction Rate (RR) inside sample A5		$1.93 \times 10^{10} \text{ cm}^{-2} \text{ s}^{-1}$
RR fraction due to epithermal neutrons (1eV-100 keV)	%	81.3
^{99}Mo Specific Activity at saturation	As	50.8 mCi/g
^{99}Mo Specific activity	As	11.3 mCi/g
^{98}Mo bulk mass requested for 1 Ci ^{99}Mo	m	90 g
Production route		$^{100}\text{Mo}(n, 2n)^{99}\text{Mo}$
Estimated total Reaction Rate (RR) inside sample A5		$1.31 \times 10^{10} \text{ cm}^{-2} \text{ s}^{-1}$
RR fraction due to epithermal neutrons (1eV-100 keV)	%	100
^{99}Mo Specific Activity at saturation	As	34.5 mCi/g
^{99}Mo Specific activity	As	7.7 mCi/g
^{98}Mo bulk mass requested for 1 Ci ^{99}Mo	m	130 g

7.5 Benchmark

During the work progress of FESTA experiment it was presented such a comparison table (Table 70) of several experiments with the aim of producing ^{99}Mo with an accelerator-driven neutron source via the $^{98}\text{Mo}(n, \gamma)^{99}\text{Mo}$ nuclear reaction, based on the Activator idea proposed by C. Rubbia. These experiments are describes in the initial part of this section and their results of the Activity measurements are in $\text{Kbq/g}/\mu\text{A/h}$ units.

Table 70: Experimental and MC results from FESTA compared with other Experiments

	MCNPX (Ispra) ²⁾	EXP (Ispra) ²⁾	EXP (Louvain-la-Neuve)#	FESTA		
			R=11.5 cm, Θ=48.6 deg	FLUKA	EXP	
	unit, (kBq/g/μA/h)					
¹⁹⁸ Au	554.00	972.00	523.44	149.22	346.00	B7
⁹⁹ Mo	0.30	7.00	5.09*	2.14	1.86	A3
^{116m} In			49758.74	16279.07	36662.11	B5
¹⁸⁷ W			149.65	70.65	85.71	B5
	36 MeV p beam		65 MeV p	60 MeV p		

a) The values in parentheses are the Louvain-la-Neuve yields modified to take into account the difference in proton beam energy.

* This value is mentioned in the conclusion. (Not same position with Au, In and W)

Louvain results are in $\mu\text{Ci/g}/\mu\text{A}/7T_{1/2}$ on the paper. They normalized the values as 7 time half-lives irradiation as shown in the next equation:

$$\frac{I}{37} \int_0^{7T_{1/2}} x e^{-\lambda t} dt = y \ (\mu\text{Ci/g}/\mu\text{A}/7T_{1/2}),$$

where x is $\text{kBq/g}/\mu\text{A/h}$ and $\frac{I}{37}$ is for converting kBq to μCi

$$x = \frac{37y}{\left[\frac{T_{1/2}}{\ln 2} \left\{ 1 - \left(\frac{I}{2} \right)^7 \right\} \right]} \ (\text{kBq/g}/\mu\text{A/h})$$

Positions from the center of the Be target: B7 (28.2 cm), A3 (23.3 cm), B5 (22.6 cm).

JRC Ispra does not clearly mention its sample position, but should be less than 10 cm from the target. The position of the sample in Louvain-la-Neuve was 11.5 cm from the target. FESTA (^{99}Mo) is 22.1 cm from the target.

8 Discussion

8.1 Introduction

During the FESTA experiment, we have conducted the following studies:

- **Neutron flux measurements** by irradiating with neutrons produced in a beryllium target, different activation foils of high purity (> 99%) such as Al, Au, Cu, Fe, In, Mg, Ni, Sc, Ti, V and Zr.
- *Neutrons capture rate measurements* of a number of isotopes of particular interest in nuclear medicine as ^{98}Mo , $^{\text{Nat}}\text{Lu}$, $^{\text{Nat}}\text{Re}$ and $^{\text{Nat}}\text{Ho}$.
- *Monte Carlo Simulation* to reproduce the experimental data. Originally, only FLUKA and MCNPx were considered, however a problem was detected in the deuteron beam case conducting us to consider other MC codes like PHITS and GEANT4.

The large number of measurements using a proton/deuteron beams (60 and 30 MeV respectively) carried out on samples over the whole volume of the activator, provide self-consistency checks including reproducibility of measurements and systematic control, and checks the validity of the cross section data of the studied samples.

The gamma radiation of these samples was measured with a germanium detector (HPGe). We were able to collect a lot of information by placing the samples in different positions of the activator as a function of the target distance. The activator was composed of 10 holes that allow the positioning of samples where up to three samples would be paced simultaneously for each irradiation. For this reason, technically speaking the activator could contain up to 30 positions simultaneously.

8.2 Activator assembly

The volume of lead diffuser, the target system and the sample holder, are the main components of the Activator. These three components are part of the activator from a viewpoint of mechanical engineering. The Physics of the Activator of course comprises the entire system itself, where the target system is the main source of neutrons released in the system and its performance depends on the power of the beam and the material of the target compound. Once the neutrons are released in the Activator are diffused, reflected and moderated because of different types of nuclear reactions, which takes place in the different regions of the device as a function of time and energy.

The first observations in relation to the construction of the activator can be summarized as follows:

- The positions of the samples make difficult the study the transport of neutrons. It is difficult to place the samples for an optimum radial distribution of the flux as a function of target distance (as it was done in TARC experiment). Would have been more favorable to have the holes in the direction of the particle beam.

- Very difficult access to the target system. This complicates the need to change the target, which could have been useful for an optimal study of the dimensions of the Be target dimensioned for 60 MeV proton beam and not for a 30 MeV deuteron beam.
- The metallic support (table) of the volume diffuser may be a reason for the unexpected and important neutron leakage out of the system.
- The sample holders have effectively served to place the samples in the system, but were not taken safety measures for the handling of samples, once irradiated.
- The degrader unit, it has not been used. The possibility of using degraders to operate with different beam energies was not used due to lack of time.

8.3 Target Assembly

The beryllium target (99.4% purity; $d = 4$ cm & thickness = 2 cm) was placed at the center of the assembly at the end of the proton beam transport tube. It produces a high neutron flux when bombarded with a proton/deuteron beams.

The expected number of neutrons released from the Be target can be estimated as follow:

- Experimentally it was not possible to reproduce the high production number of neutrons from deuteron on Be target as indicated by FLUKA (50 – 80 times between 50 MeV protons and 25 MeV deuterons)
- One should have included in the design of the Activator the possibility of exchanging the target material and geometry, of varying the dimensions of the different active regions (graphite reflector, lead diffuser, polyethylene neutron shield, etc.)
- The use of the degrader unit allowing the possibility to vary the incoming beam energy and to study in details the deuteron break-up process.

8.4 Monte Carlo Simulation

As mentioned above, FLUKA & PHITS were used for the runs with a 60 MeV proton beam and only PHITS for the runs with a 30 MeV deuteron beam. Originally, only FLUKA was considered for the simulation campaign runs with proton and deuteron beams, however because of the inconsistency of FLUKA with the deuteron beam we have considered the use of other PHITS to reproduce the data of the irradiated samples from a 30 MeV deuteron beam. A full report (Luis, Raul) describing the neutron characteristics of FESTA Activator using FLUKA with 15 MeV deuterons and 40 μ A of beam intensity is available in Annex 1.

Deuterons are transported with FLUKA, but the used of SDUM = IONSPLIT option does not follow to estimate the break-up process correctly. It is clear that a reliable nuclear data for the production and transport of neutrons and photons produced by deuterons is required. Up to now, due to the lack of such nuclear data libraries,

Monte Carlo transport codes use built-in semi empirical nuclear models to deal with charged particle interactions. An example facing the same problem can be found in the (P. Sauvan, J. Sanz et al.) Article.

PHITS seems to reproduce correctly this phenomenon and therefore was used to compare against the results obtained in FESTA for both protons and deuterons.

We are interested in the neutron fluence produced by proton (deuteron) beams of a given energy. A very special suggestion is to reproduce, in the same way as was done in TARC experiment (Arnould, 1999), the neutron fluence from the resonance energies of the activation foils as a function of radial distance from the center of the Be target. At this level MC simulation (FLUKA & PHITS) of the neutron fluence and energy-time correlation were conducted in each part of the activator: Be Target, Lead diffuser, Graphite reflector, Polyethylene & Borated Polyethylene Layer, for no appreciable capturing sample. Some of the results of this work (Figure 44) allow us to clearly observe the signature of deuteron break-up through the neutron peak at 15 MeV, which is the mono-energetic peak at more than 10^{13} n/cm²/μC, from the breakup of deuterons.

8.5 Neutron Flux measurements

The flux measurements have to be compared to the prediction of the Monte Carlo codes (FLUKA in case of protons and PHITS in case of deuterons) for neutron transport in FESTA Experiment. This comparison is an important prerequisite step for the understanding the ARC method applied in the Activator. It is therefore crucial to assess precisely the agreement between the data and the prediction. This requires not only a technical evaluation of the Monte Carlo calculations, with FLUKA or PHITS coherently, but also a rigorous assessment of the experimental results.

The dependence of the neutron fluence on energy and space, which is the integral of the flux over a certain period of time, validates the neutron transport and tests the reliability of the neutron-induced cross-sections of lead. Pure lead was chosen to ensure that impurities have a negligible effect on the neutron flux.

We observe that the neutron flux is quasi iso-lethargic in the diffuser and graphite reflector regions, in the energy range between 0.1 eV and 1 KeV, which corresponds to the resonance region of interest for the ARC method as claimed in the Patent PCT/EP2014/069463.

We used Monte Carlo simulations PHITS and FLUKA codes to reproduce the neutron flux spectrum in the Activator with 60 MeV protons and 30 MeV deuterons. The results with FLUKA clearly show the following:

- In the case of lead diffuser, the ratio between fast neutrons (> 0.1 MeV) and epithermal neutrons (1 eV to 100 KeV) is of the order of 10.
- The sharp slowing down/moderation of the fast neutrons after exiting the Be Target (100 times less). This is due namely to the large (n, xn) cross section of lead,

that contributes significantly to the multiplication on the neutrons produced in the target.

- (n, xn) reactions are less in the case of 30 MeV deuterons because the average neutron energy (almost 12 MeV) is less than in the case of 60 MeV protons (nearly to 27 MeV).
- The polyethylene layer efficiently reduces the number of neutrons escaping the system.
- The deuteron break-up with a neutron peak in the neutron flux spectrum at 15 MeV.
- Polyethylene and Borated polyethylene neutron flux levels are 20 times higher, compared with the case for 30 MeV deuterons.

The results with PHITS clearly show the following:

- No more break-up peak at 15 MeV (properly production of secondary particles induced by light ions nuclear reactions)
- 60 MeV protons produce 2-4 times more neutrons than in the case of 30 MeV deuterons.

One should have considered the use of special neutron detectors such the gas detectors, ionization chambers or track detectors, in order to measure directly the neutron flux spectrum. The use of activation foils, as in TARC is acceptable but requires extensive post-processing work in order to deconvolute the neutron flux.

8.6 Activation foils measurements

It is very important to have an idea of the results of the activity obtained from the measured samples and MC simulation calculations. The table shows the average values of the ratio of the experimental data results for all samples used in FESTA with the MC simulation results.

Table 71: Data/MC average values of the Activity for activation foils.

Beam type	Sample	Radionuclides	EXP/FLUKA	EXP/PHITS
p	natIn	In-116m	1.52	1.9
p	natIn	In-115m	0.58	0.45
p	natAu	Au-198g	2.24	2.94
p	natW	W-187	0.92	1.85
p	natAl	Na-24	1.71	1.35
p	natZr	Zr-97	1.29	1.48
p	natSc	Sc-46	1.75	
p	natCu	Cu-64	1.35	1.88
d	natIn	In-116m	0.03	1.67
d	natIn	In-115m	0.02	0.52
d	natAu	Au-198g	0.04	2.8
d	natW	W-187	0.02	1.56
d	natAl	Na-24	0.01	1.19

^{nat}In irradiated at different positions in the Activator with a proton beam and the MC simulation codes are showing almost the same results either for ^{116m}In & ^{115m}In comparing with the experimental data.

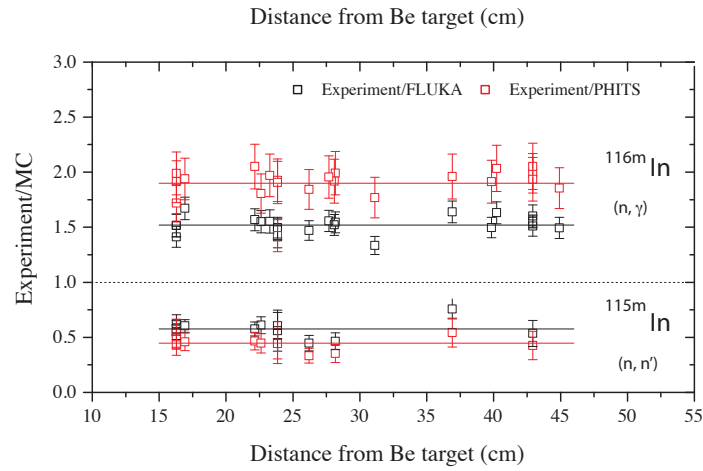


Figure 214: Ratio of the Data/MC activity for ^{115m}In & ^{116m}In (60 MeV p).

This ratio does not reproduce well high-energy interactions (n, n') because X-section is not so well known.

In the case of deuteron beam, FLUKA overestimates the experimental values in a factor 30 for ^{116m}In in a factor 50 for ^{115m}In , while in the case of PHITS we can remark that for ^{116m}In the data is almost 70% higher and for ^{115m}In is less in a 50% comparing with PHITS calculations.

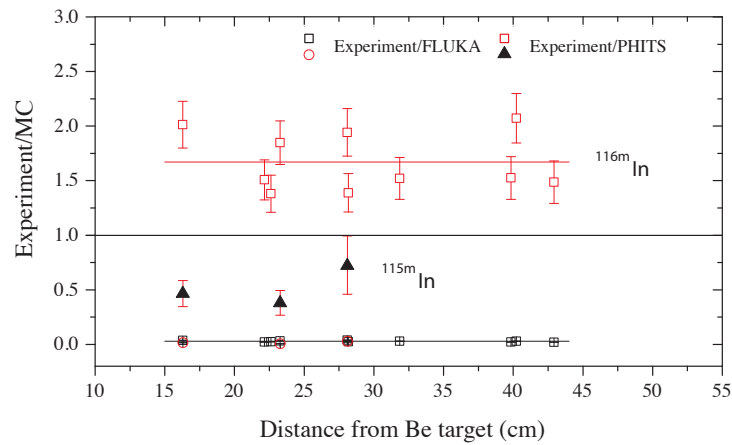


Figure 215: Ratio of the Data/MC activity for ^{115m}In & ^{116m}In (30 MeV d).

Next figures show both reactions (n, γ) & (n, n') that behaves differently. In the first case for ^{116m}In (Figure 216) dominated by thermal neutrons, thermal neutrons peaks (1.46 eV @ ~29000 barns) caused by graphite and polyethylene moderator/reflectors, one can observe almost a flat behaviour with the distance from the Be target while in the second case for the (n, n') reactions originated by fast neutrons, the activity decays exponentially almost following the law $1/r^2$ (Figure 217).

The cross section curve with a threshold nearly to 0.6 MeV and its value of $\sigma = (325 \pm 5)$ mb in the range of energies until 10 MeV are well known and inelastic processes are sensitive to the scattered neutrons.

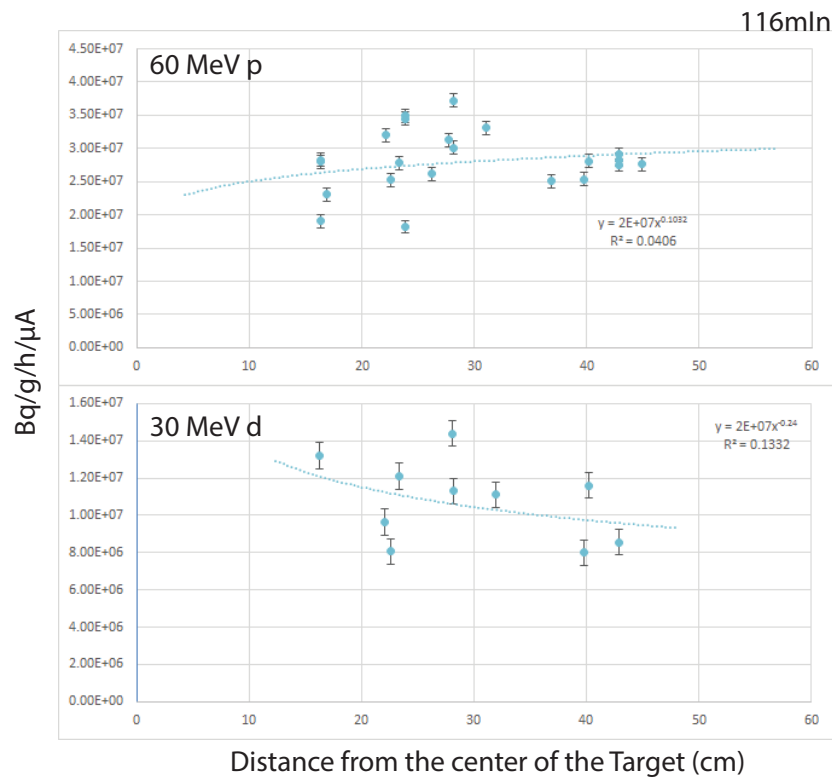


Figure 216: ^{116m}In curve fitting - (n, γ) reaction.

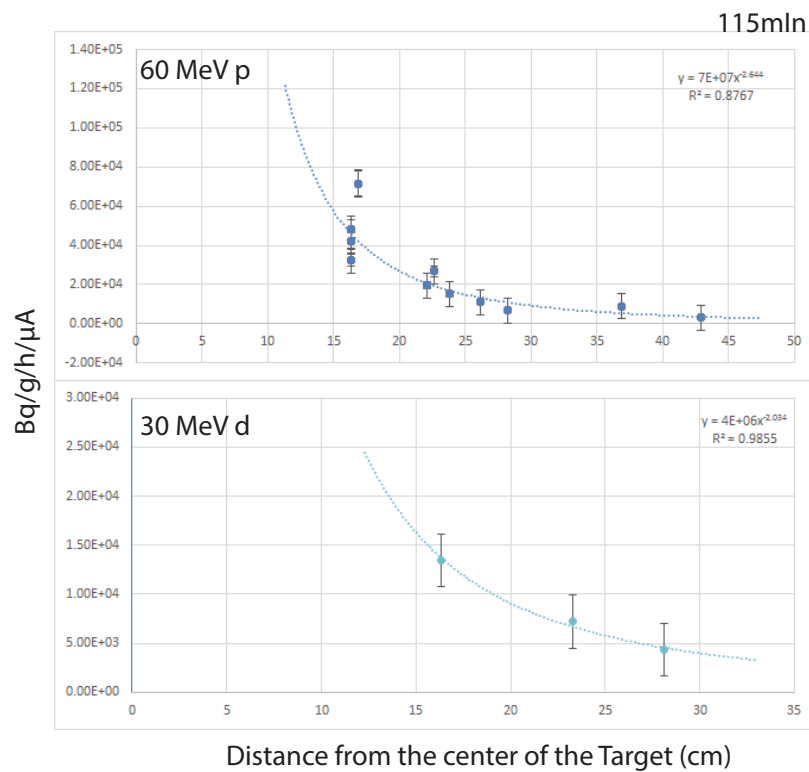


Figure 217: ^{115m}In curve fitting - (n, n') reaction.

^{Nat}W deduced activities of different samples with a proton beam in average are showing a good agreement with the FLUKA calculations and a difference in a factor 2 comparing with PHITS calculations.

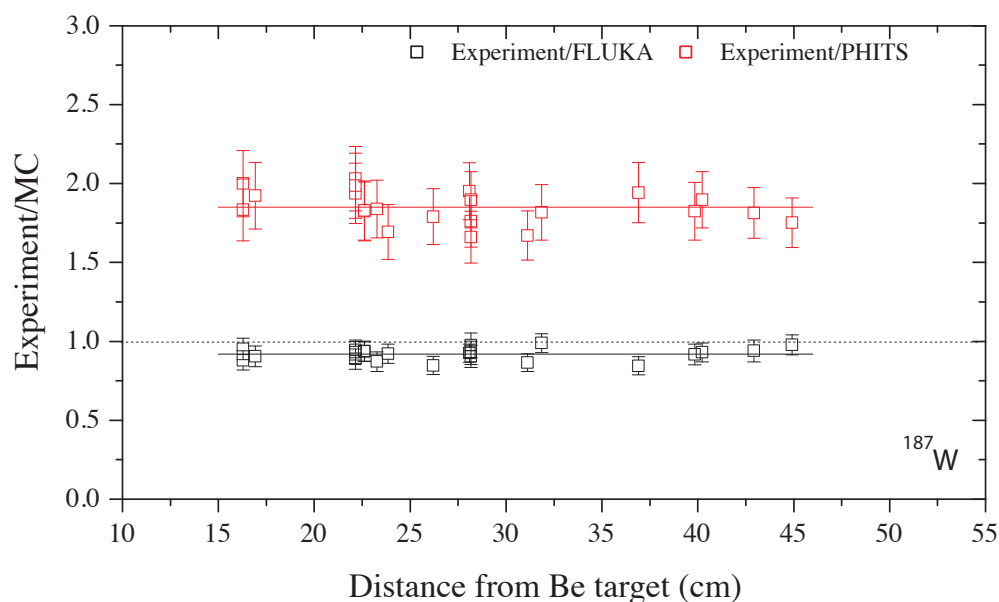


Figure 218: Ratio of the Data/MC activity for ^{187}W (60 MeV p).

Using a deuteron beam, again FLUKA calculations are overestimating the experimental data in a factor 50, while PHITS reproduce the almost 70% in average the experimental data.

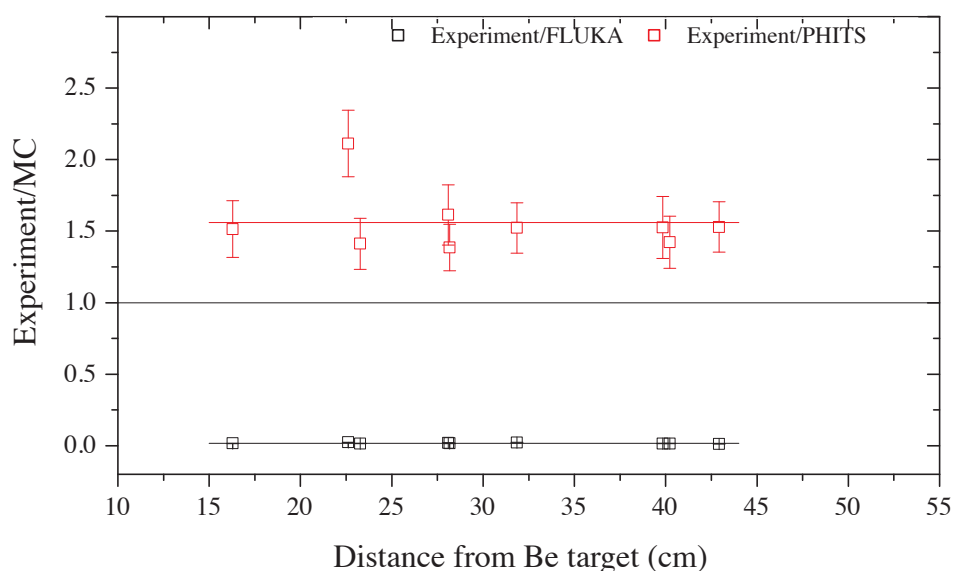


Figure 219: Ratio of the Data/MC activity for ^{187}W (30 MeV d).

Tungsten activation foils with natural abundance has ^{186}W (28.4%) and we are interested in the (n, γ) reactions with a resonance peak at energy of 18.8 eV with at $\sigma = \sim 14000$ barns. Figure 220 shows the induced measured activities in both cases for p & d where we can distinguish almost the same behaviour, almost a flat dependence along the target distance, dominated by thermal neutrons peaks caused by graphite and polyethylene moderator/reflectors. The most important difference between the two cases is the average energy of neutrons in the device (27 MeV and 12 MeV), and this is observed in the induced activity, which is less in the case of 30 MeV deuterons compared with 60 MeV protons.

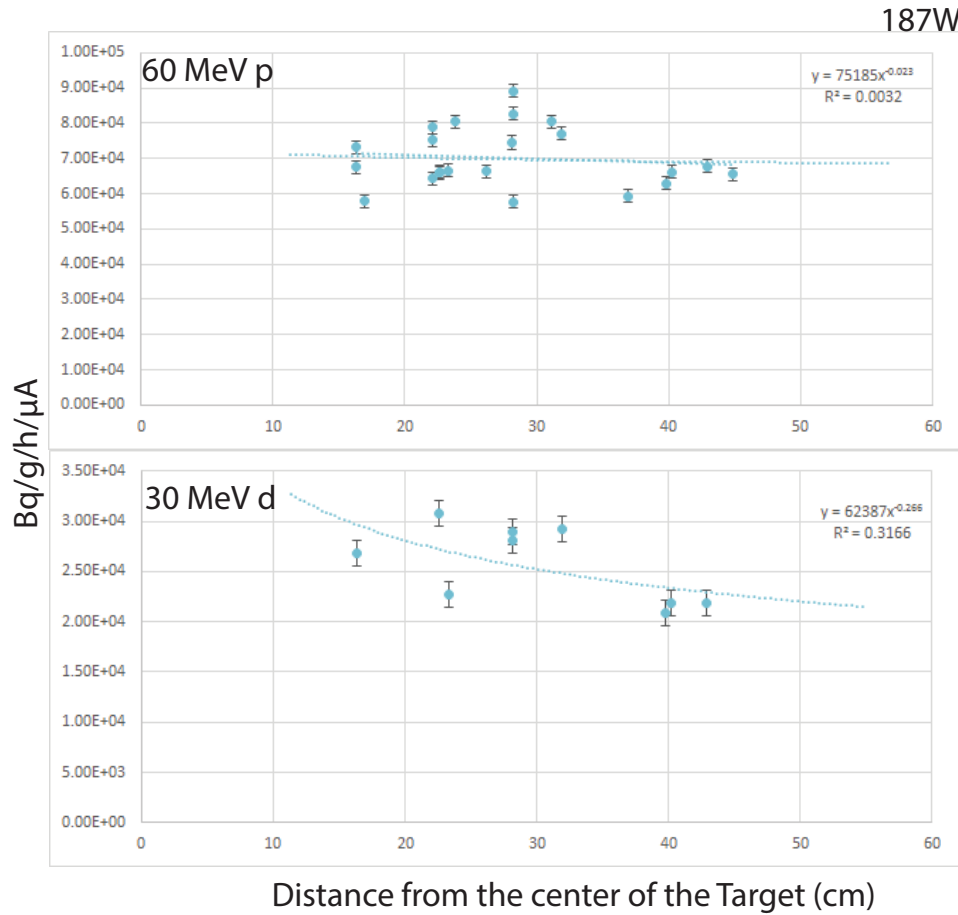


Figure 220: ^{187}W curve fitting.

^{Nat}Au

In the case of natural gold with a proton beam, the difference between the experimental data and MC simulation varies in a factor from 2-3.

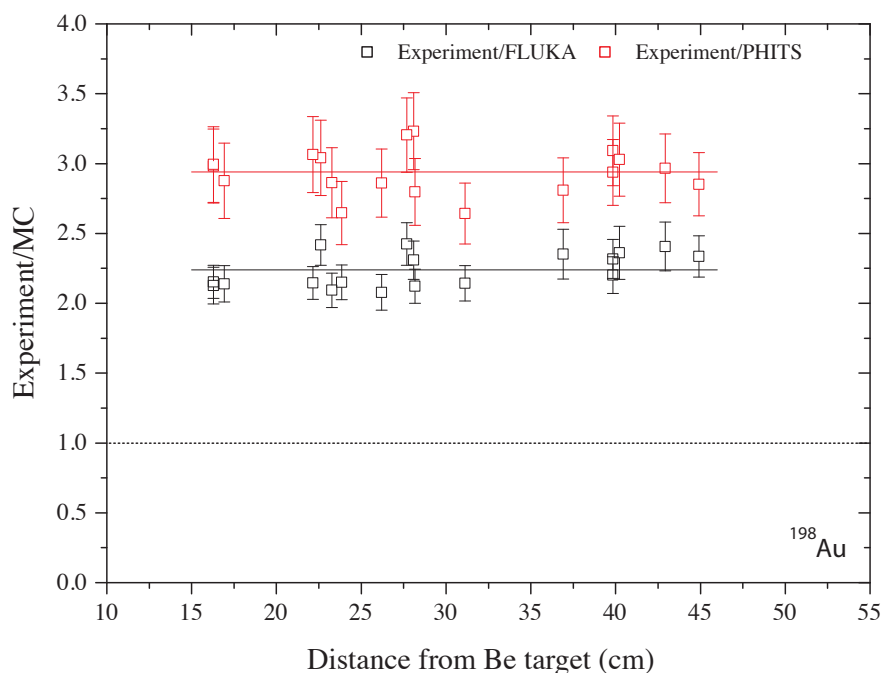


Figure 221: Ratio of the Data/MC activity for ^{198}Au (60 MeV p).

FLUKA with a deuteron beam overestimates the experimental data in a factor 25. Using PHITS the experimental data is bigger in a factor 3.

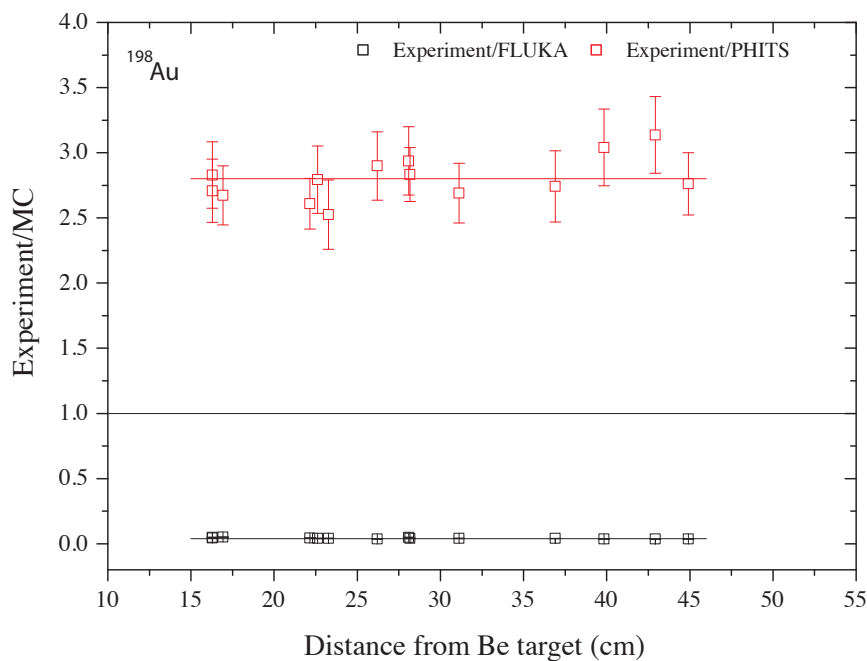


Figure 222: Ratio of the Data/MC activity for ^{198}Au (30 MeV d).

Gold activation foils with natural abundance has ^{197}Au (100%) and we are interested in the (n, γ) reactions with a resonance peak at energy of 4.9 eV with at σ of the order of ~ 27000 barns. Figure 223 shows the induced measured activities in both cases for p & d where we can distinguish almost a flat dependence along the target distance, dominated by thermal neutrons peaks caused by graphite and polyethylene moderator/reflectors.

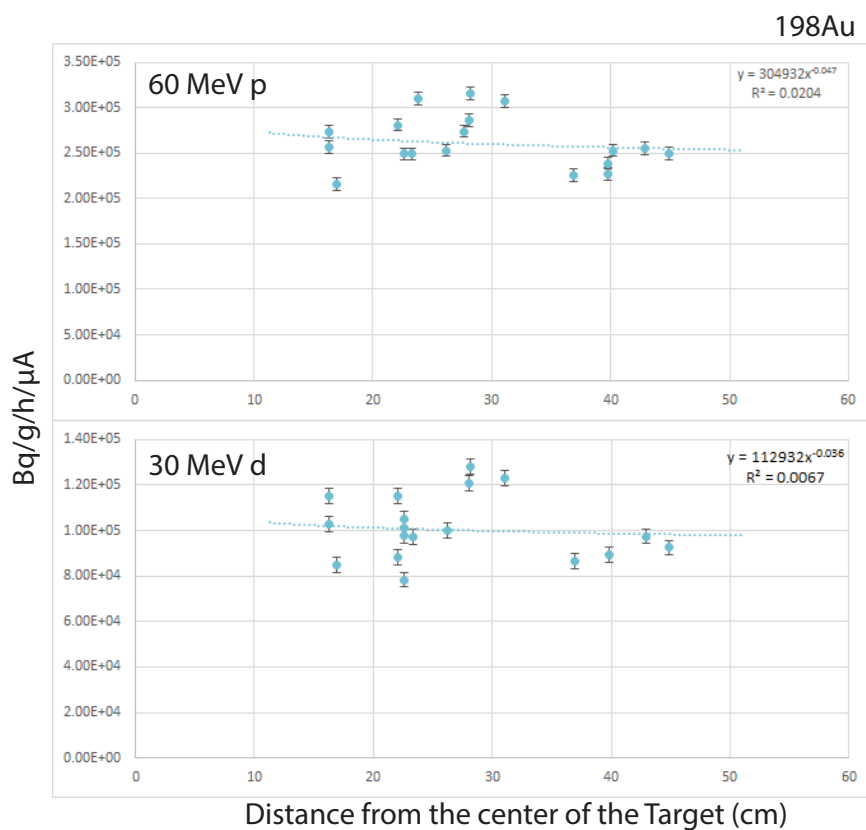


Figure 223: ^{198}Au curve fitting.

For Natural Aluminum with a proton beam, the difference between the experimental data and FLUKA/PHITS is 1.7 and 1.4 larger respectively.

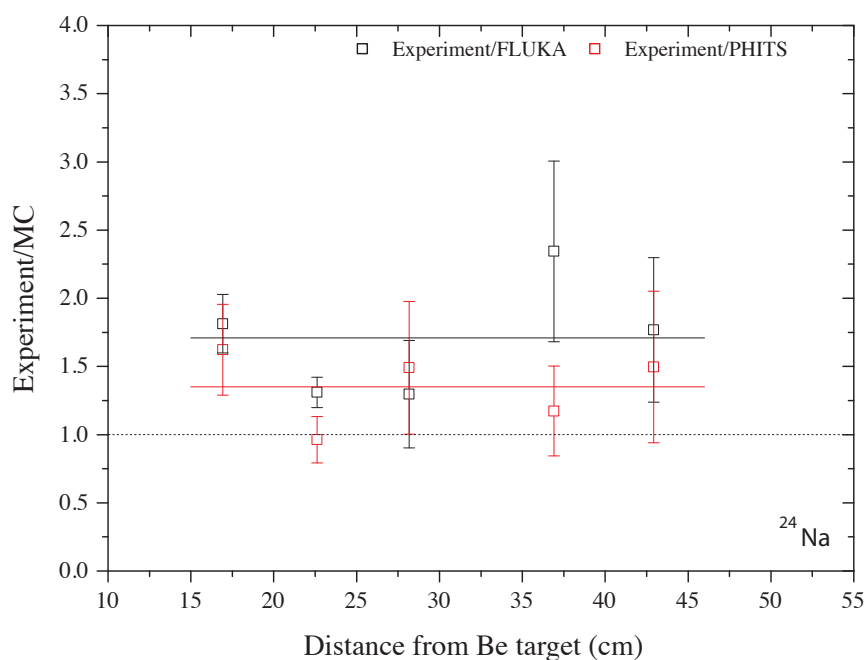


Figure 224: Ratio of the Data/MC activity for ^{24}Na (60 MeV p).

With a deuteron beam, we can see again that FLUKA overestimates the experimental data in a factor 100 while the data is larger by a factor 1.2 comparing with PHITS.

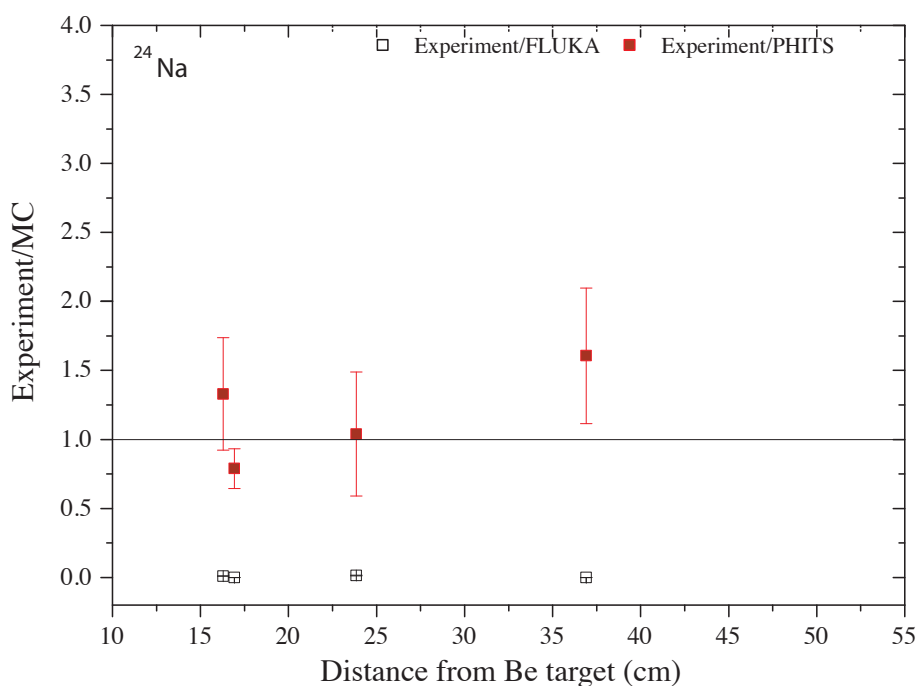


Figure 225: Ratio of the Data/MC activity for ^{24}Na (30 MeV d).

Aluminium activation foils with natural abundance has ^{27}Al (100%) and we are interested in the (n, α) reactions. The neutron capture threshold energy is another important factor. In the $^{27}\text{Al}(n, \alpha) ^{24}\text{Na}$ reactions thermal or low energy neutrons do not activate the sample, where the cross section is 121.3 mbarns at 14.1 MeV. It means we have mainly the presence of fast neutrons.

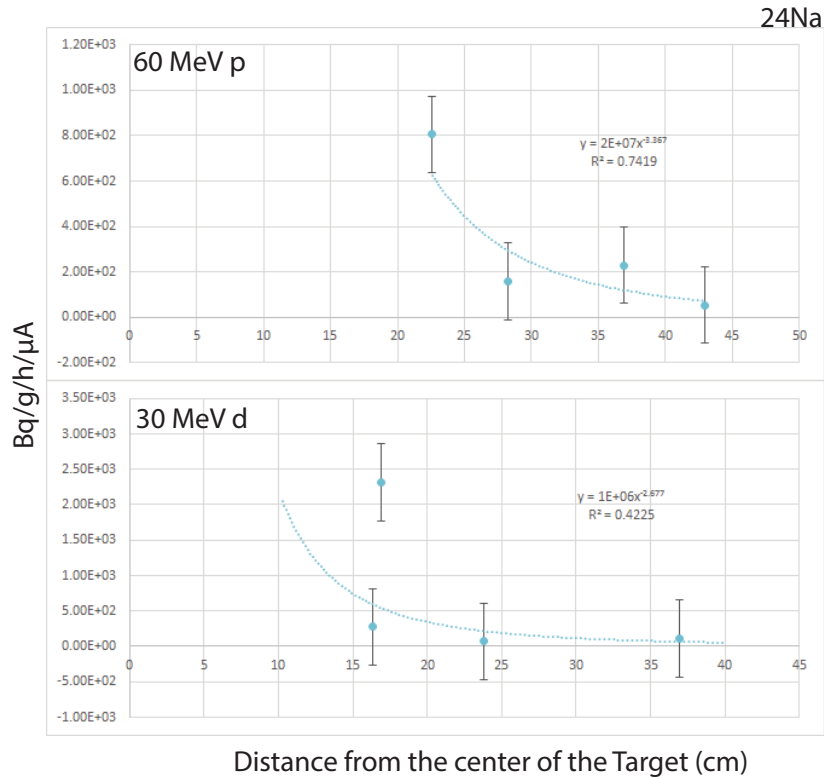


Figure 226: ^{24}Na curve fitting.

One can notice the behaviour of the induced activity in both case and they are following the law $1/r^x$. In case of protons $x = 3.4$ and which means a rapid decrease of the Activity along the distance from the Be target, than in the case of deuterons, where $x = 2.7$.

For Natural Zirconium with a proton beam, the difference between the experimental data and FLUKA/PHITS are 50% and a factor 1.5 respectively.

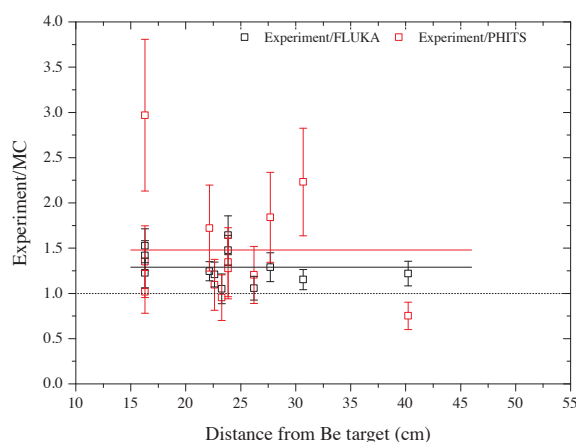


Figure 227: Ratio of the Data/MC activity for ^{97}Zr (60 MeV p).

Zr activation foils with natural abundance has ^{90}Zr (51.5%) and ^{96}Zr (2.8%). We are interested in the (n, 2n) & (n, γ) reactions respectively. The neutron capture threshold energy is another important factor. In the $^{90}\text{Zr}(\text{n}, 2\text{n})^{89}\text{Zr}$ reactions thermal or low energy neutrons do not activate the sample, where the cross section is 9 barns at 22 MeV. It means we have mainly the presence of fast neutrons. In the $^{96}\text{Zr}(\text{n}, \gamma)^{97}\text{Zr}$ reactions epi-thermal neutrons are captured at the energy resonance cross section of 1000 barns at 300 eV. In Figure 228 again we can remark that the behaviour of the induced activity for the (n, γ) reactions decrease slightly & for the (n, 2n) reactions the activity decreases obeying the $1/r^x$ law, where $x = 2.23$.

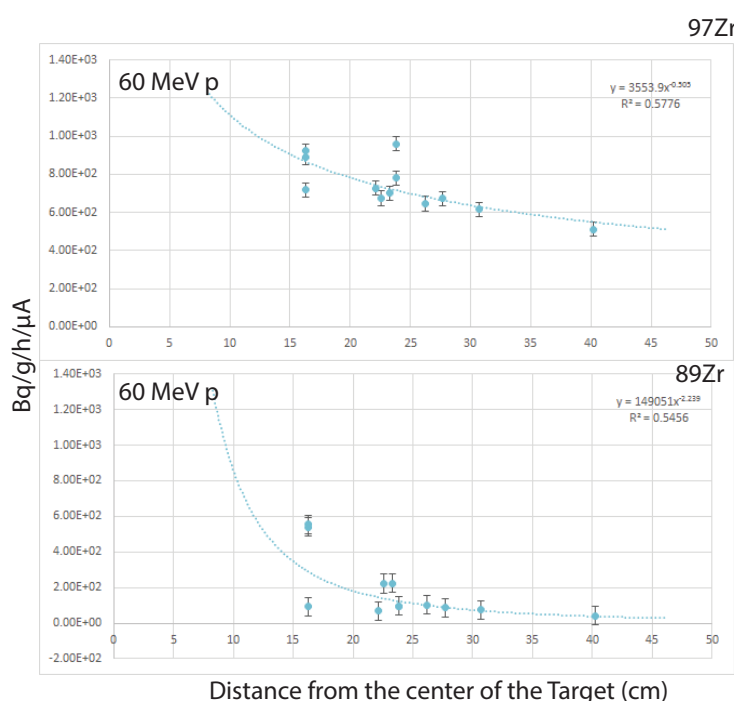


Figure 228: ^{97}Zr & ^{89}Zr curve fitting.

8.7 Medical Isotopes Data/MC Ratio

The table shows the average values of the ratio of the experimental data results for all samples used in FESTA with the MC simulation results.

Table 72: Data/MC average values of the Activity for all medical isotopes.

Beam type	Sample	Radionuclides	EXP/FLUKA	EXP/PHITS
p	natMo	Mo-99	0.8	1.11
p	98Mo	Mo-99	0.82	1.53
p	natRe	Re-188	1.16	1.72
p	natLu	Lu-177	2.59	3.2
p	natHo	Ho-166		3.03
d	natMo	Mo-99	0.01	0.89
d	98Mo	Mo-99	0.02	1.59
d	natRe	Re-188	0.02	1.75
d	natLu	Lu-177	0.05	3.12
d	natHo	Ho-166		2.91

With a ratio Data/MC varying from 1.5 to 3.0, it is very difficult to estimate the total number of doses that will be produced as a function of beam energy and/or beam current. Contrary to the Activation foils there is a rather large uncertainty of the (n, γ) cross sections for these isotopes.

^{Nat}Mo

In the case of ^{Nat}Mo irradiated at different positions in the Activator with a proton beam we can see that data is 0.8 times less than the FLUKA results and 1.1 larger than the results provided by PHITS.

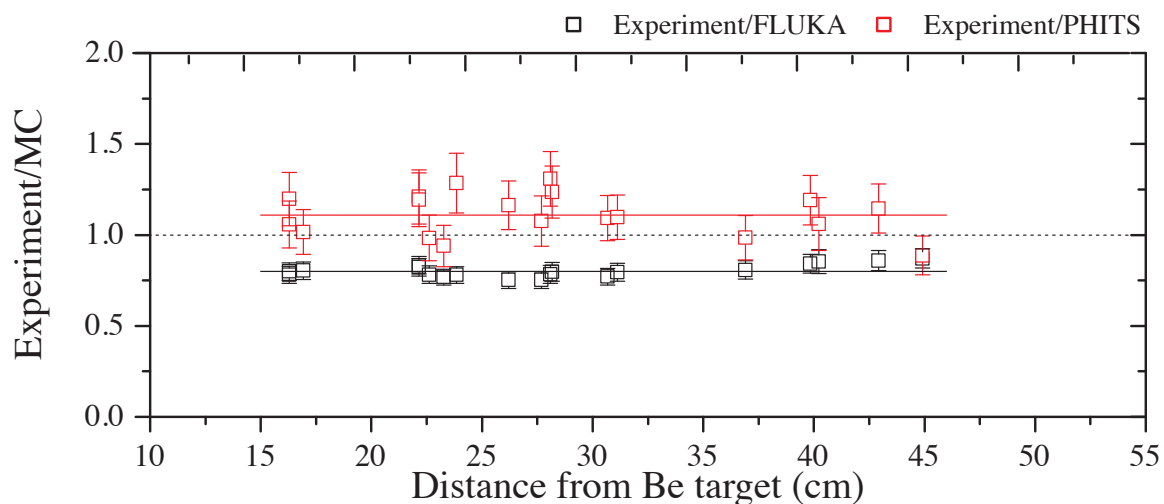


Figure 229: Ratio Data/MC of the activity of ⁹⁹Mo from ^{Nat}Mo (60 MeV p).

Using a deuteron beam FLUKA is overestimating the experimental results in a factor 100 and the data is 0.9 times less than the results provided by PHITS code.

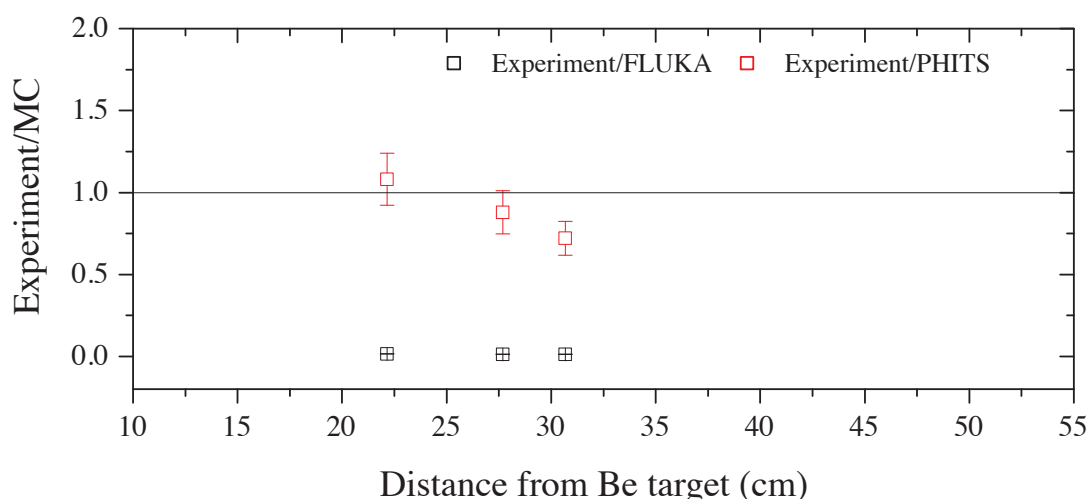


Figure 230: Ratio Data/MC of the activity of ^{99}Mo from ^{Nat}Mo (30 MeV d).

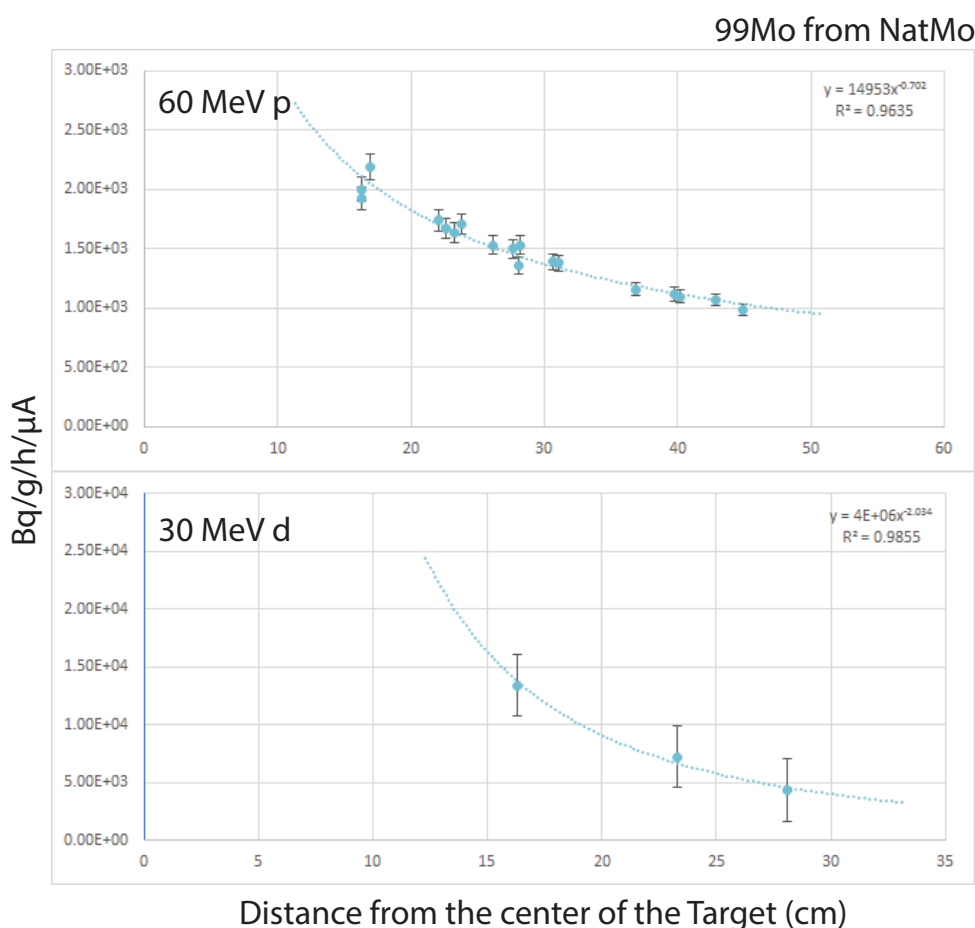


Figure 231: ^{99}Mo from ^{Nat}Mo curve fitting.

Natural molybdenum has seven stable isotopes ^{92}Mo (14.84%), ^{94}Mo (9.25%), ^{95}Mo (15.92%), ^{96}Mo (16.68%), ^{97}Mo (9.55%), ^{98}Mo (24.13%), and ^{100}Mo (9.63%). ^{99}Mo comes from $^{98}\text{Mo}(n, \gamma)^{99}\text{Mo}$ neutron capture reaction with a cross section of 60.1 barns at 12.1 eV. These reactions for p & d have almost the same behaviour following the $1/r^x$ law, where $x(p) = 0.7$ & $x(d) = 2.04$.

For the enriched ^{98}Mo , the Data/MC results shows that the data in average is 0.82 times bigger than the FLUKA calculations and 1.5 bigger than the values obtained with PHITS code. Using the deuteron beam FLUKA again goes up, and underestimates the data results in a factor 50. In the case of PHITS, the data is larger by a factor 1.6.

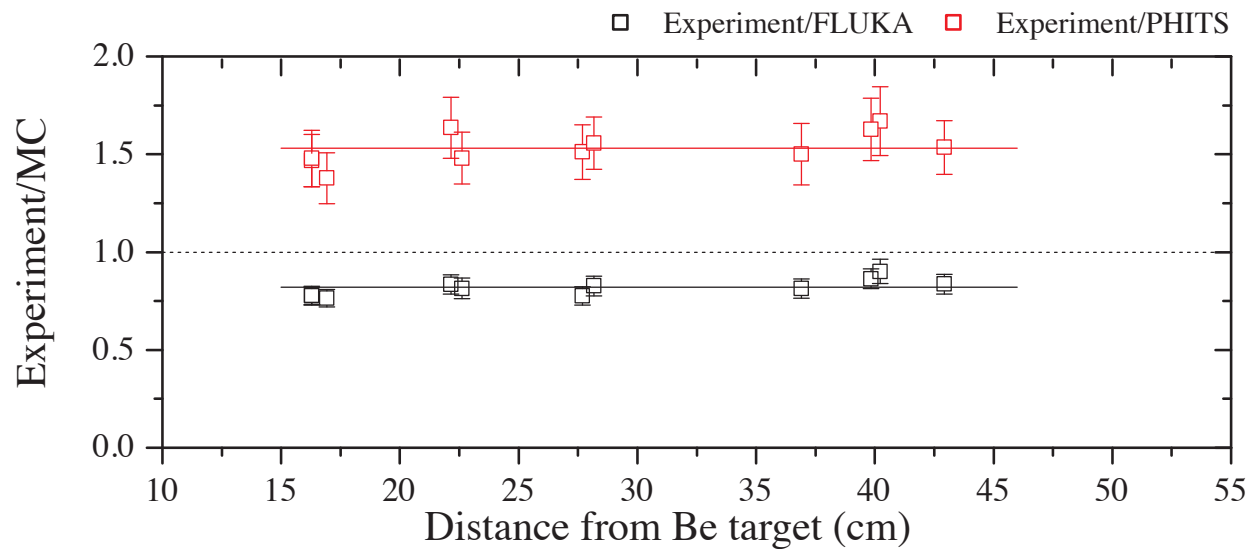


Figure 232: Ratio Data/MC of the activity of ^{99}Mo from enriched ^{98}Mo (60 MeV p).

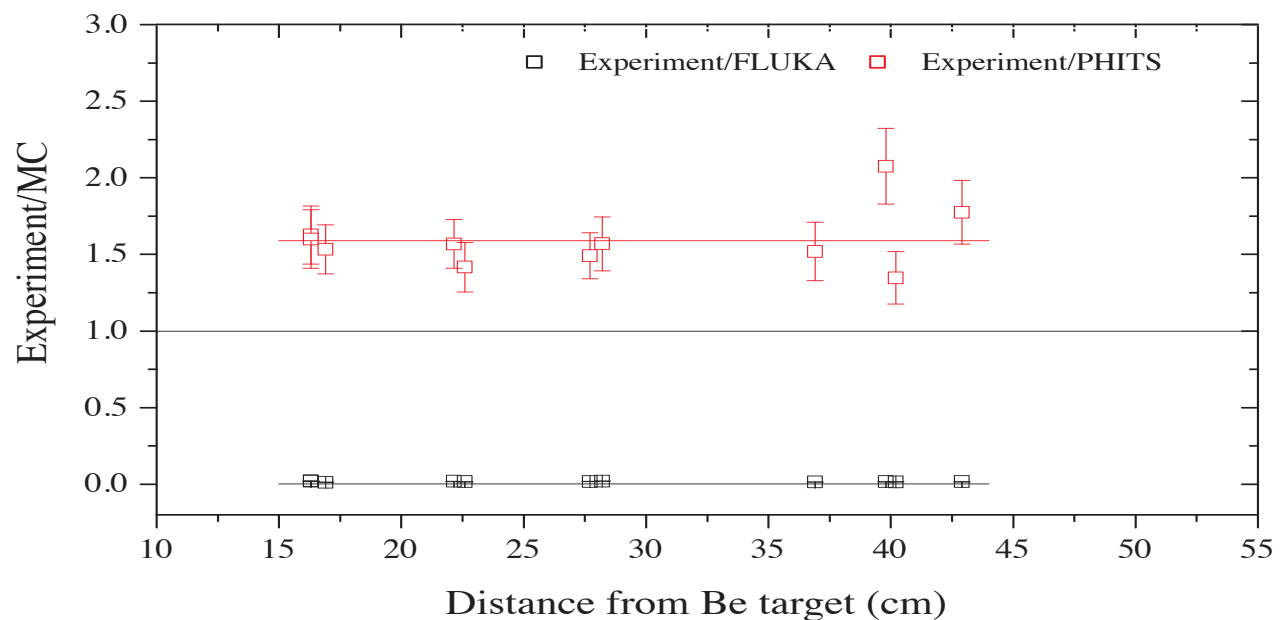


Figure 233: Ratio Data/MC of the activity of ^{99}Mo from enriched ^{98}Mo (30 MeV d).

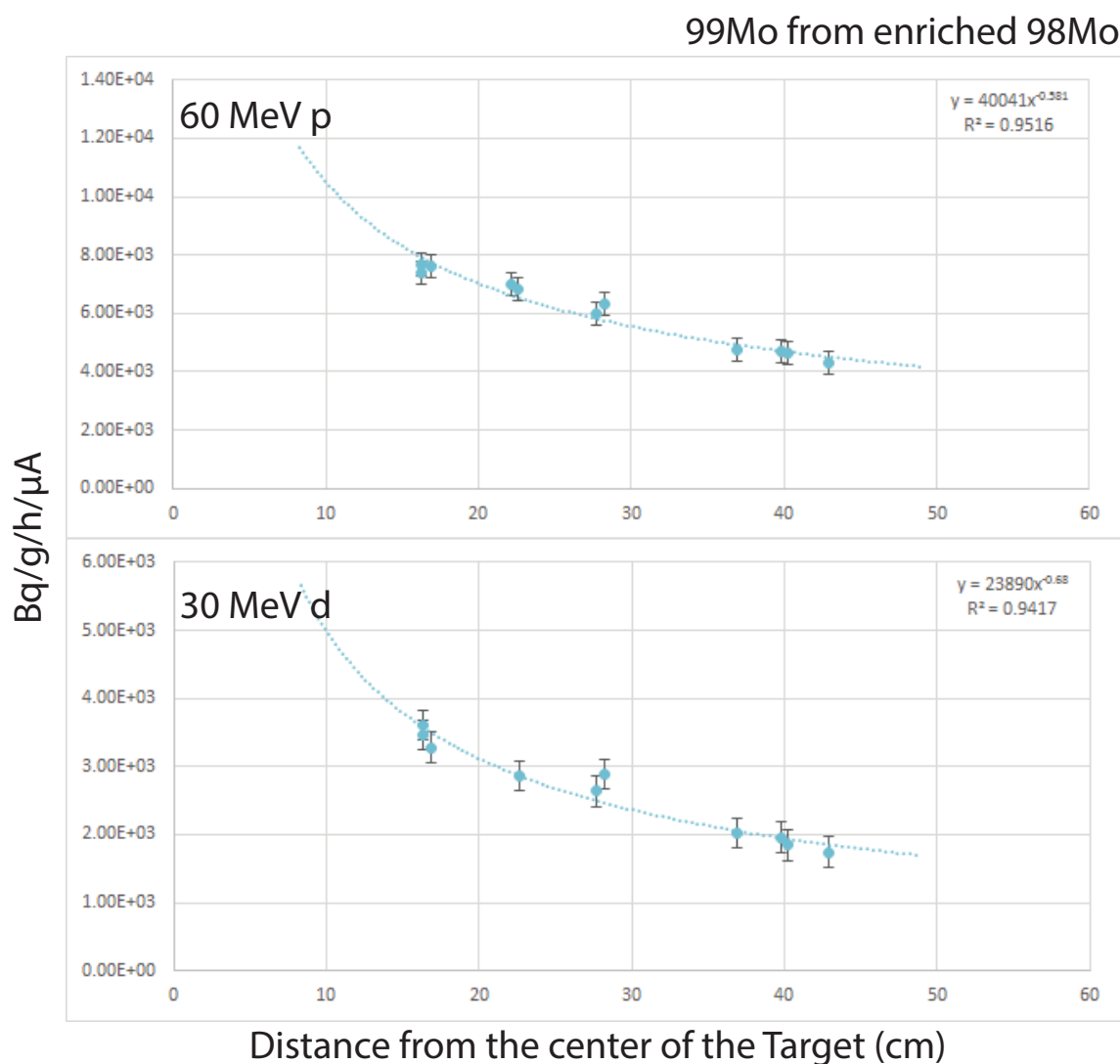


Figure 234: ⁹⁹Mo from enriched ⁹⁸Mo curve fitting.

⁹⁹Mo comes from enriched ⁹⁸Mo(n, γ)⁹⁹Mo neutron capture reaction with a cross section of 60.1 barns at 12.1 eV. These reactions for p & d have almost the same behaviour following the 1/r^x law, where x(p) = 0.6 & x(d) = 0.7.

For ^{188}Re with a proton beam, the Data/MC results shows that the data in average is 1.16 times bigger than the FLUKA calculations and 1.7 bigger than the values obtained with PHITS code. Using the deuteron beam FLUKA again underestimates the data results in a factor 50. In the case of PHITS, the data is larger by a factor 1.8.

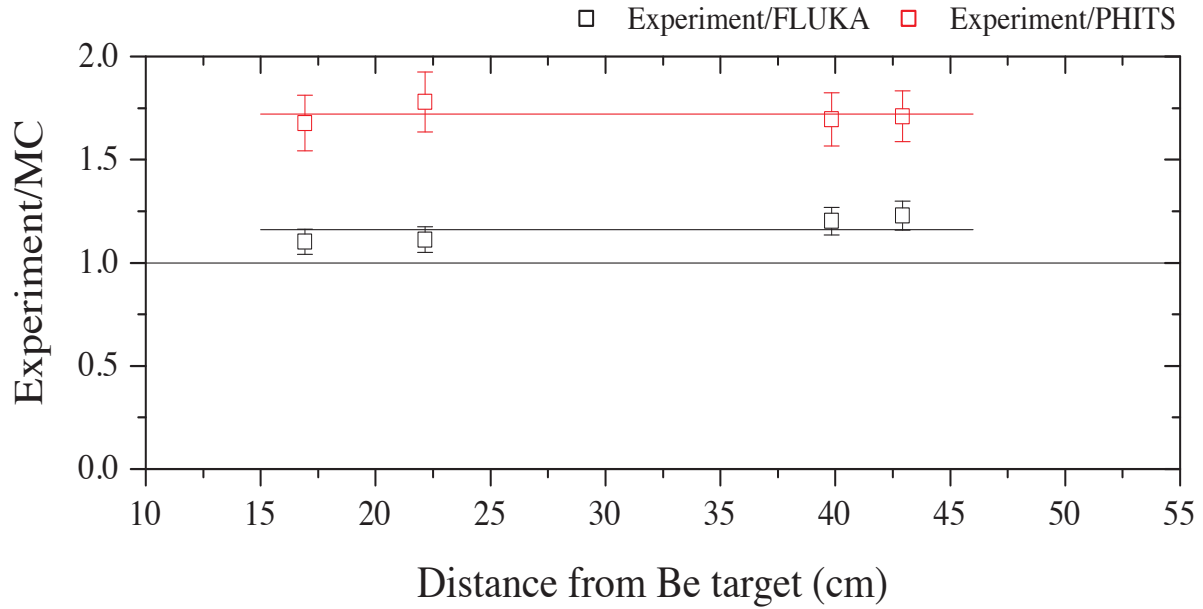


Figure 235: Ratio Data/MC of the activity of ^{188}Re (60 MeV p).

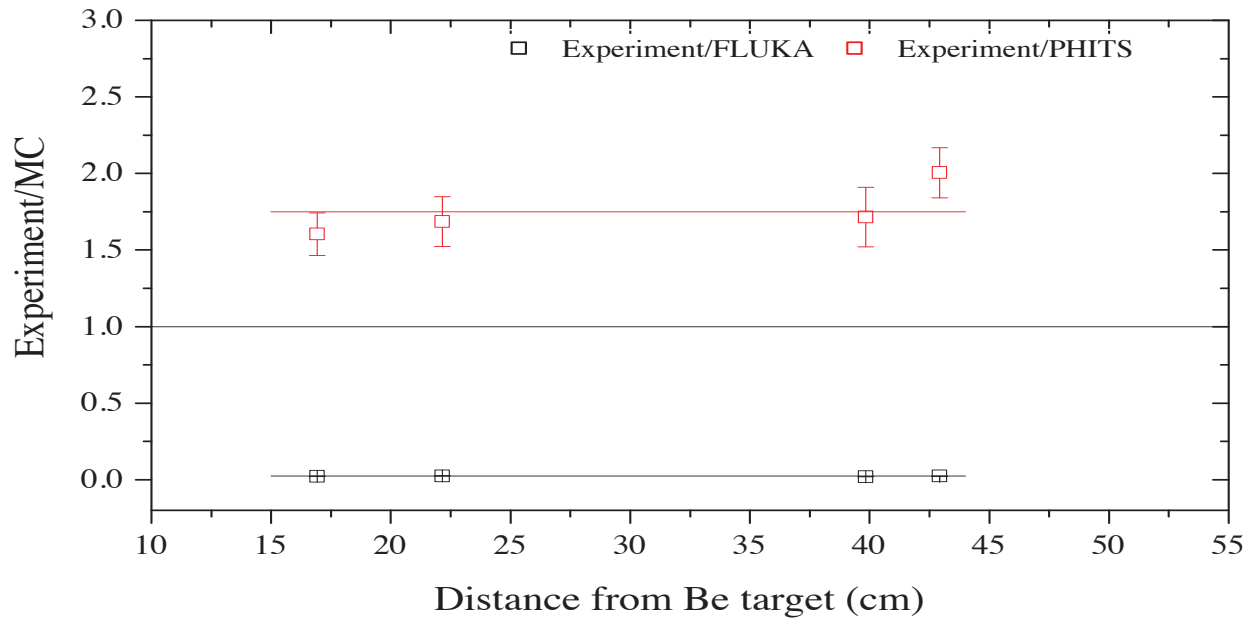


Figure 236: Ratio Data/MC of the activity of ^{188}Re (30 MeV d).

$^{\text{Nat}}\text{Re}$ has two stable isotopes ^{185}Re (37.4%) and ^{187}Re (62.6%). ^{188}Re comes from $^{187}\text{Re}(n, \gamma)^{188}\text{Re}$ reaction with a cross section near to 1000 barns at 8 eV. In both cases for p & d one can observe a slightly decrease of the induced activity.

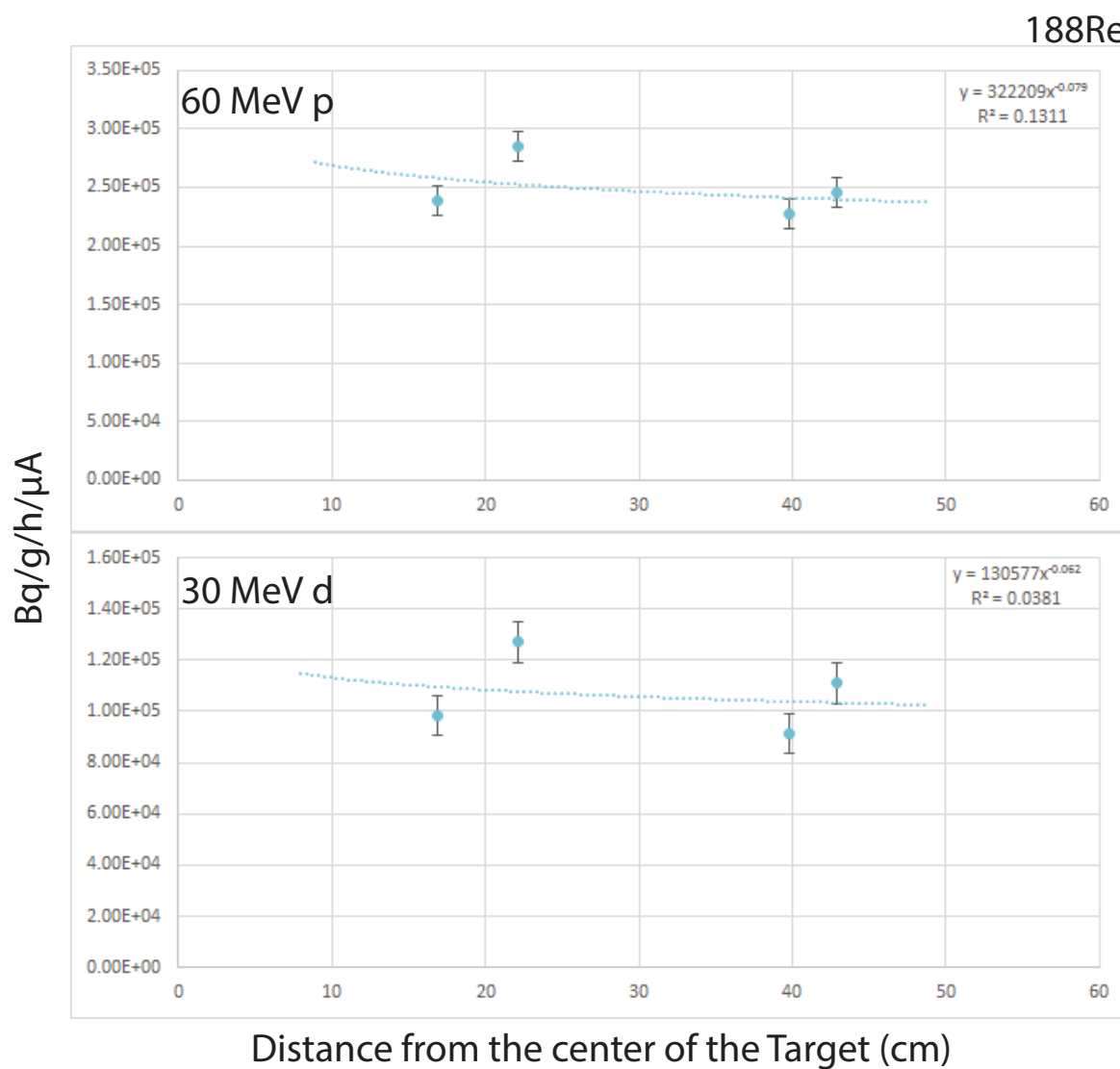


Figure 237: ^{188}Re curve fitting.

For ^{177}Lu with a proton beam, the Data/MC results shows that the data in average is 2.6 times larger than the FLUKA calculations and 3.2 larger than the values obtained with PHITS code.

Using the deuteron beam FLUKA again underestimates the data results in a factor 20. In the case of PHITS, the data is larger by a factor 3.1.

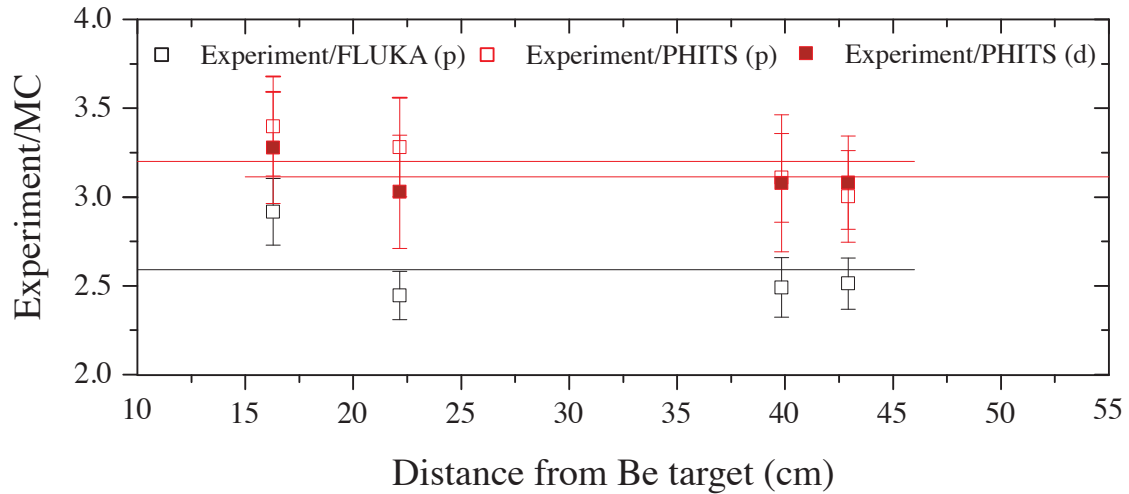


Figure 238: Ratio Data/MC of the activity of ^{177}Lu (60 MeV p & 30 MeV d).

^{177}Lu has two stable isotopes ^{175}Lu (97.41%) and ^{176}Lu (2.59%). ^{177}Lu comes from $^{176}\text{Lu}(n, \gamma)^{177}\text{Lu}$ reaction with a cross section of 8000 barns at 2 eV. Unfortunately, we have not enough data to see the expected behaviour, which has to follow the law $1/r^\alpha$.

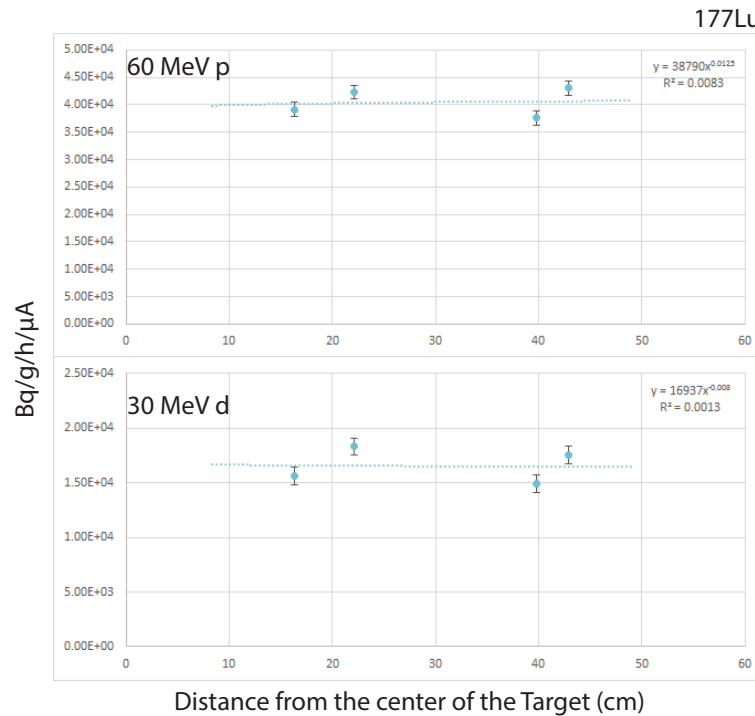


Figure 239: ^{177}Lu curve fitting.

^{166}Ho

For ^{166}Ho with a proton beam, the Data/MC results shows that the data in average is 3 times larger than the PHITS calculations. There are no values with FLUKA because the capture cross-section was not available.

Using the deuteron beam, data result is larger in average by a factor 3 that the PHITS results.

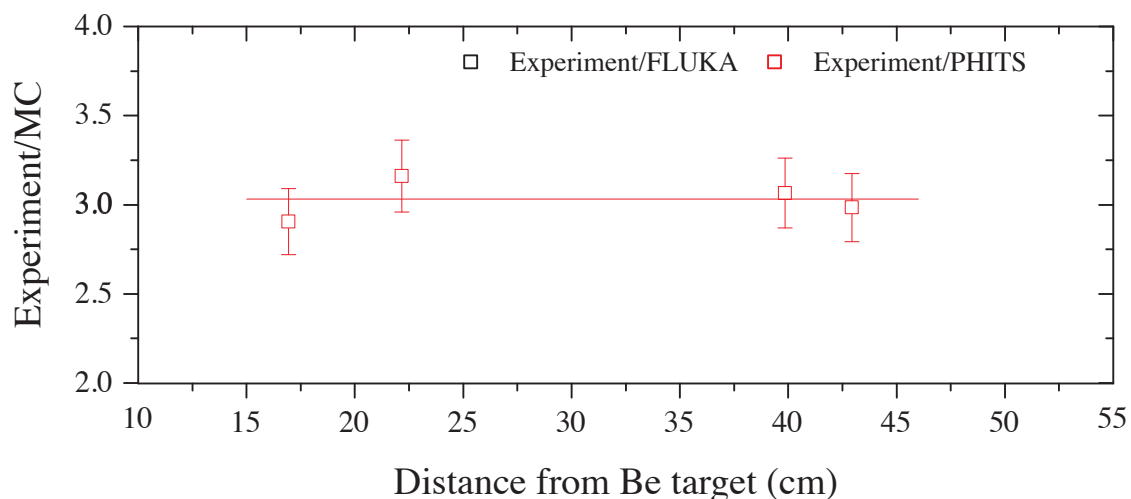


Figure 240: Ratio Data/MC of the activity of ^{166}Ho (60 MeV p).

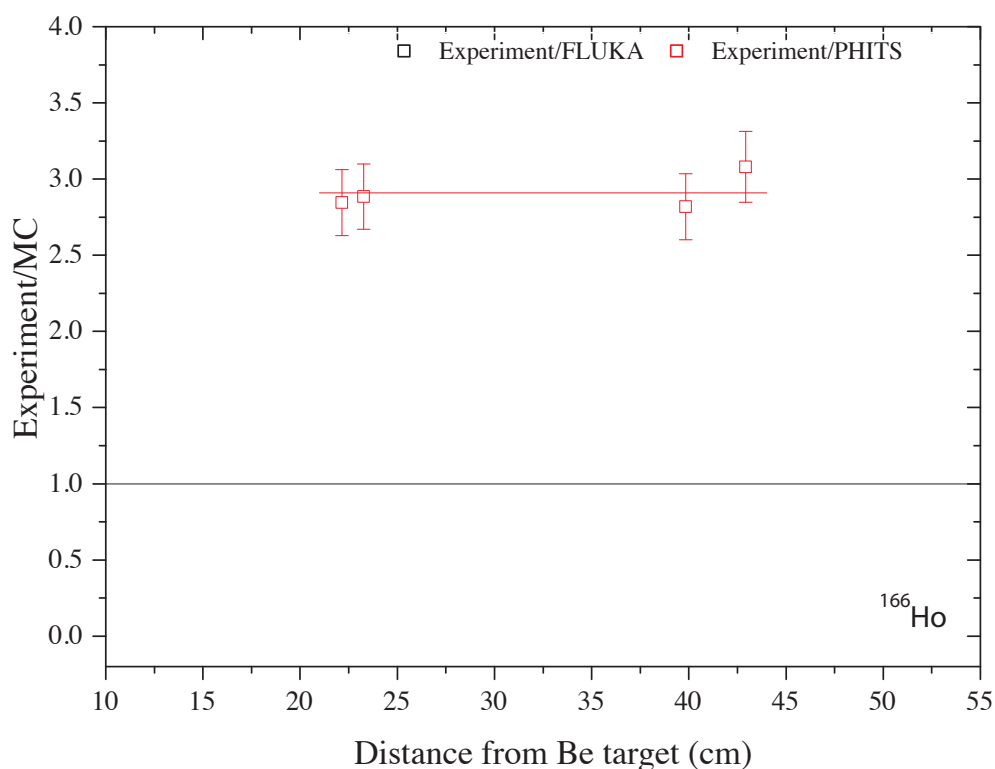


Figure 241: Ratio Data/MC of the activity of ^{166}Ho (30 MeV d).

^{166}Ho has only one stable isotope ^{165}Ho . ^{166}Ho is produced by $^{165}\text{Ho}(n, \gamma)^{166}\text{Ho}$ reaction with a cross section of 9000 barns at 8 eV. The decrease of the induced activity is observed in both cases as it is expected.

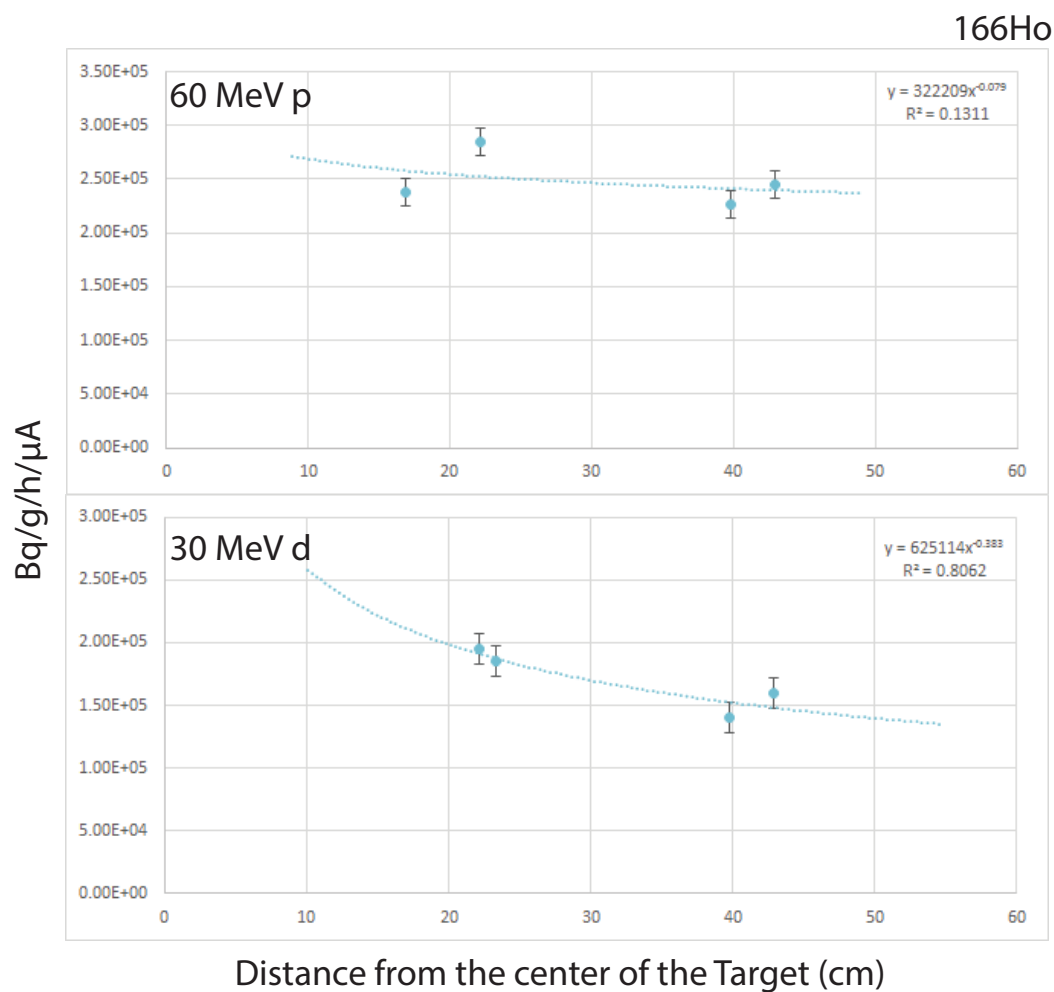


Figure 242: ^{166}Ho curve fitting.

8.8 Captures and resonances

¹⁸⁸Re

According to FESTA results, we are producing 31.2 times more ¹⁸⁸Re than ⁹⁹Mo.

$$\frac{{}^{188}\text{Re Activity @ A5 position}}{{}^{99}\text{Mo Activity @ A5 position}} = \frac{2.38 \times 10^5}{7.64 \times 10^3} = 31.2$$

If we use ^{Nat}Re, then ¹⁸⁷Re is about 62.6%. The ratio is:

$$\frac{31.2}{0.626} = 49.8$$

Now we check the ratio of the resonance integral of ¹⁸⁸Re & ⁹⁹Mo:

$$\frac{{}^{187}\text{Re RI}}{{}^{98}\text{Mo RI}} = \frac{323 \text{ barns}}{7 \text{ barns}} = 49.1$$

¹⁷⁷Lu

According to FESTA results, we are producing 5.1 times more ¹⁷⁷Lu than ⁹⁹Mo.

$$\frac{{}^{177}\text{Lu Activity @ A8 position}}{{}^{99}\text{Mo Activity @ A8 position}} = \frac{3.91 \times 10^4}{7.69 \times 10^3} = 5.1$$

If we use ^{Nat}Lu, then ¹⁷⁶Lu is about 2.59%. The ratio is:

$$\frac{5.1}{0.0259} = 197$$

Now we check the ratio of the resonance integral of ¹⁷⁶Lu & ⁹⁸Mo:

$$\frac{{}^{176}\text{Lu RI}}{{}^{98}\text{Mo RI}} = \frac{1100 \text{ barns}}{7 \text{ barns}} = 157$$

¹⁶⁶Ho

According to FESTA results, we are producing 50 times more ¹⁶⁶Ho than ⁹⁹Mo.

$$\frac{{}^{166}\text{Ho Activity @ A5 position}}{{}^{99}\text{Mo Activity @ A5 position}} = \frac{3.77 \times 10^5}{7.64 \times 10^3} = 49.35$$

If we use ^{Nat}Ho, then ¹⁶⁵Ho is 100%. The ratio is:

$$\frac{49.35}{1} = 49.35$$

Now we check the ratio of the resonance integral of ¹⁶⁶Lu & ⁹⁸Mo:

$$\frac{{}^{165}\text{Ho RI}}{{}^{98}\text{Mo RI}} = \frac{667 \text{ barns}}{7 \text{ barns}} = 95.3$$

We can remark that this is a confirmation that the captures are taking place all over the resonance range, which extends up to 50 KeV in the case of ⁹⁸Mo and not in the main resonance which is at $E_n \approx 12 \text{ eV}$.

8.9 Sensitivity study

8.9.1 Thickness Effect

Thickness effect studies at 60 MeV proton beam, were conducted in FESTA experiment with ^{nat}W , ^{nat}Mo and ^{98}Mo metallic powder. One can observe that with increasing the sample thickness, the induced activity of the sample decreases slightly and this study could allow defining the optimal thickness of the sample as we can see in Figure 243.

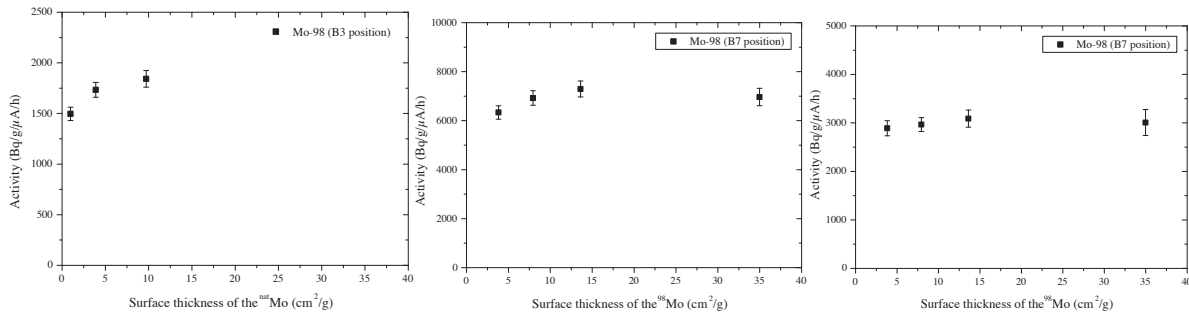


Figure 243: Induced activity for different surface thicknesses (^{98}Mo).

The self-shielding effect could modify the local flux inside the sample volume: This because it is important to consider the flux perturbation due to the detector itself in the Monte Carlo simulation. In particular, the complex structure of cross sections in the resonance region requires careful treatment to describe correctly self-shielding effects.

8.9.2 Effect of the Diffuser support

This refers to the study to verify the asymmetry of neutron capture on the top and bottom region of the activator with respect to the center of the target. Three activation foils were used: ^{nat}Zr , ^{nat}In and ^{nat}W . There is apparently a clear evidence of significant loss of neutrons in the bottom part of the Activator, caused by the presence of the iron table, which supports the lead diffuser. It was estimated that a possible optimization of the Activator to correct this problem might improve the neutron capture.

In order to have a conclusion to this effect it is necessary to compare fluxes and not just the induced measured activities.

8.9.3 Neutron leakage

Confirms that overall dimensions at the activator are sufficiently to limit the neutrons leaking outside, then there is no need to make a larger Activator. Maybe worthwhile to see if one can make the activator smaller, by reducing the polyethylene layer for example.

8.9.4 Moderator/Reflector Effect

A test of graphite reflection and moderation effects was conducted. For the graphite reflection test, one can notice that there is a slight increase of the activity in the case of ^{nat}W when the samples are placed in the graphite region, because of the contribution of captures conducted by the slowing down escape of fast neutrons (W has major resonance cross section at 18.9 eV). In the case of ^{nat}Mo the measured activity in the carbon region decreases because of the absence of epithermal neutrons, which are easily captured by ^{98}Mo . For the graphite moderation test was done by replacing lead bricks by small graphite bricks inside of the hole. The results indicate that the samples are not affected by this modification.

For both cases, a more reliable explanation can be obtained with the analysis of the neutron fluence in the activator. On the other hand, simulations carried out in Annex 1, on the multi-reflector layer, indicate that one can increase the yield by 3-4 times using a 30 MeV deuteron beam. It is suggested to perform the same simulations with a 60 MeV protons.

8.9.5 Symmetry in B & -B layers

The study of the symmetry effect in the activator shows almost same results of the induced activity of the samples placed at the same distance from the target in both layers (B & -B).

8.9.6 Neutron source discussion

Four samples (^{nat}In , ^{nat}Au , ^{nat}W , and ^{nat}Al) were used to check the neutron leakage and where placed close to an open hole entrance in the Activator. The measured induced activity for the samples in the hole was about 10^2 times smaller than the samples inside the activator.

A previous induced activity of ^{116m}In measured in the same position is slightly larger in the order of 5% comparing with the one measured in this test. That could be explained by the neutron loses (escaping) across the open hole.

9 Conclusions

A compact, accelerator-driven, neutron activator based on the Adiabatic Resonance Crossing (ARC) method has been developed in the framework of a Collaboration agreement between CAL, AIMA AD & IBEL SA. It has been constructed by IBEL SA, assembled and tested at CAL using the MEDICYC (60 MeV p + Be & 30 MeV d + Be) Cyclotron, with the aim of efficiently deliver ion-beam generated neutrons for the activation of different activation foils and medical isotopes mostly used in nuclear medicine.

Monte Carlo simulations were carried out to develop the conceptual design of the activator, where a quasi iso-lethargic neutron flux in the diffuser and graphite reflector regions, in the energy range between 0.1 eV and 1 KeV, corresponds to the resonance region of interest for the ARC method (Patent PCT/EP2014/069463) and constitutes an important step toward for the production of radio-pharmaceuticals for nuclear medicine.

We have used computational fluid dynamics methods for thermal hydraulic design of the Be Target, in order to ensure an efficient water-cooling to the possibility of using high-current beam particles.

In FESTA Experiment we obtained the induced activity of different activation foils (^{Nat}In , ^{Nat}Au , ^{Nat}W , ^{Nat}Al , ^{Nat}Sc & ^{Nat}Zr) in order to investigate the neutron flux in the Activator; and of the different medical isotopes (^{Nat}Mo , 10 gr of enriched ^{98}Mo , ^{Nat}Ho , ^{Nat}Lu and ^{Nat}Re) with the purpose of a industrial production of these radioisotopes for diagnosis and therapy radiopharmaceutical markets.

Based on the preliminary results obtained in FESTA we can extrapolate the production yields derive from operating with MEDICYC at full power with the current geometry of the activator. In Table 66, are presented the production yields obtained with the current Activator.

Table 73: Production yields based on the current Activator

60 MeV protons (1/g/μA/h)	Mo-99	Lu-177 (nat-Lu)	Lu-177 (pure-Lu176)	Re-188	Ho-166
Experimental Results FESTA					
Activity (Bq/g/μA/h @ best location)	7585.61	42700.00	1648648.65	141000.00	476000.00
Conversion factor from Bq to Ci	2.7027E-11	2.7027E-11	2.7027E-11	2.7027E-11	2.7027E-11
Activity(MBq/g/μA/h)	7.59E-03	4.27E-02	1.65E+00	1.41E-01	4.76E-01
Activity (Ci/g/μA/h)	2.05E-07	1.15E-06	4.46E-05	3.81E-06	1.29E-05
CAL					
Current Intensity (μA)	65	65	65	65	65
M (gr)	100	100	100	100	100
Time Irradiation(h)	10	10	10	10	10
New Calculated Activity (MBq)	4.93E+02	2.78E+03	1.07E+05	9.17E+03	3.09E+04
1 Dose (MBq)	5.55E+02	5.55E+03	5.55E+03	5.55E+03	5.55E+03
New Calculated Activity (Ci)	1.33E-02	7.50E-02	2.90E+00	2.48E-01	8.36E-01
1 Dose (Ci)	0.015	0.15	0.15	0.15	0.15
Final Result (# doses)	0.89	0.50	19.31	1.65	5.57
Amplification factor needed to produce 1	75	13	0	4	1
Number of Daily Doses	67	38	1449	124	418

The results of FESTA are compared with the results of other experiments reported previously, such as:

1. Accelerator-driven, neutron activator at Louvain-la-Neuve (LLN, Belgium),
2. Accelerator-driven, neutron activator at Joint Research Centre (JRC Ispra, Italy)
3. Accelerator-driven, neutron activator at INFN Legnaro Laboratories, Italy.

With the aim of increasing the production of secondary particles induced by light ions nuclear reactions, one should have included in the design of the Activator the possibility of changing the energy and/or the beam intensity of the cyclotron; the exchange of the target material and geometry; and the possibility of varying the dimensions of the different active regions in the Activator (graphite reflector, lead diffuser, polyethylene neutron shield, etc.). The implementation in the activator of the degrader unit system give the possibility to vary the incoming beam energy and also to study in details the deuteron break-up process.

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Annex 1

CAL Activator - Neutronics Report

Raul Luís

February 2014

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1 First Configuration

1.1 Geometry Implementation in FLUKA

The first implementation of the target system in FLUKA is shown in Figure 1. A 15 MeV deuteron beam with a Gaussian shape (FWHM=0.8) in the x and y directions ($x=0$, $y=80$) travels in the positive z direction and hits the Beryllium target, shown in detail in Figure 2. The target is slightly displaced from the center of the assembly ($z=-5.25$ cm) so that the resulting dose rates are centered in the center of the system.

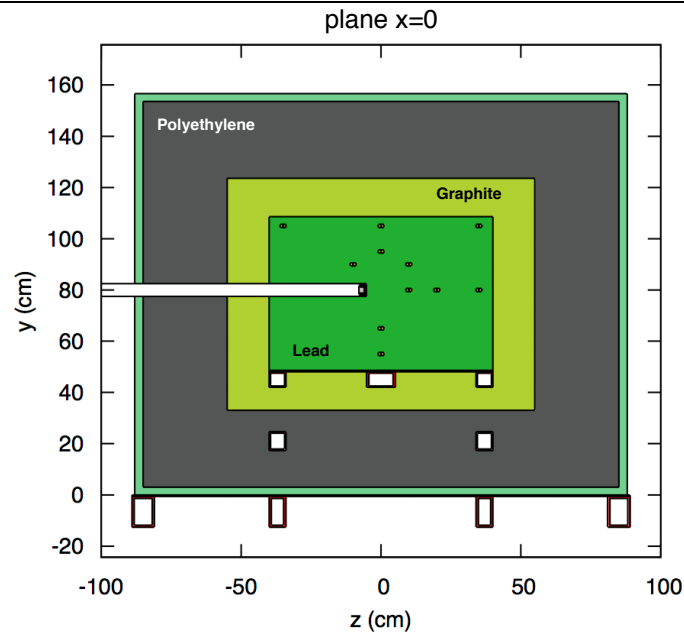


Figure 1 - Geometry implementation in FLUKA, plane $x=0$.

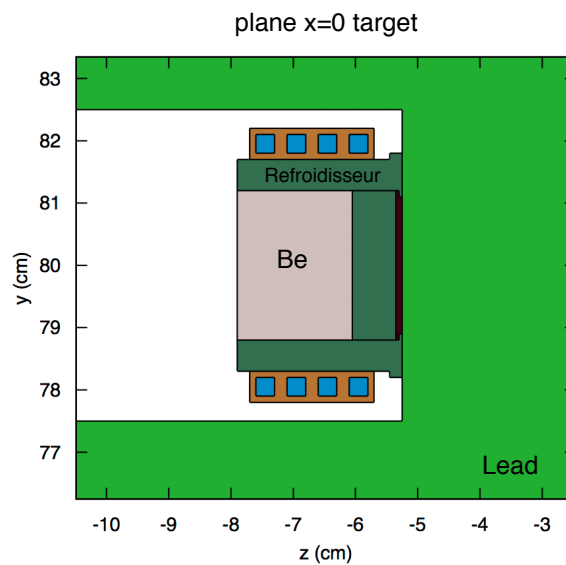


Figure 2 - Target geometry in FLUKA, plane $x=0$.

The produced neutrons are diffused in the lead that surrounds the target. Inside the lead, thirty-three targets made of 98-Mo surrounded by graphite were placed in three separate rows ($x=-20$, $x=0$, and $x=15$), as shown in Figure 3. The neutrons produced in the target are captured in the 98-Mo targets to produce 99-Mo.

The lead is surrounded by 15 cm of graphite ($\rho=1.7 \text{ g/cm}^3$), to reflect the neutrons back to the Molybdenum targets, 30 cm of polyethylene shielding (C_2H_4 , $\rho=0.93 \text{ g/cm}^3$) and 3 cm of borated polyethylene, with 10% of 10-B ($\rho=1.05 \text{ g/cm}^3$). The table that supports the system was also described in detail, as Figure 4 shows.

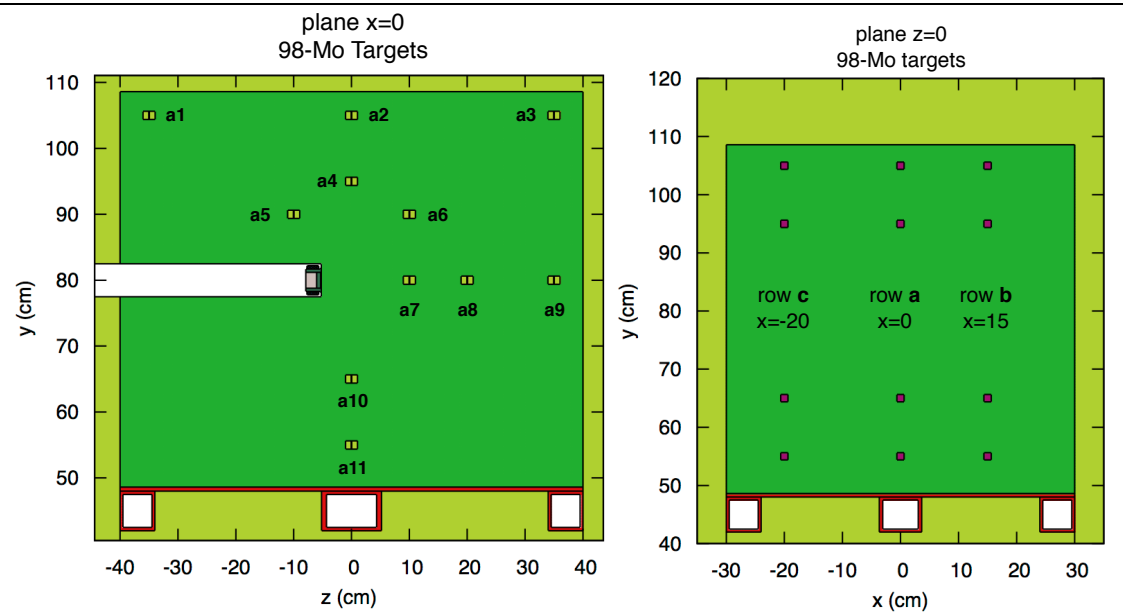


Figure 3 - Positions of the 98-Mo targets, planes $x=0$ (left) and $z=0$ (right).

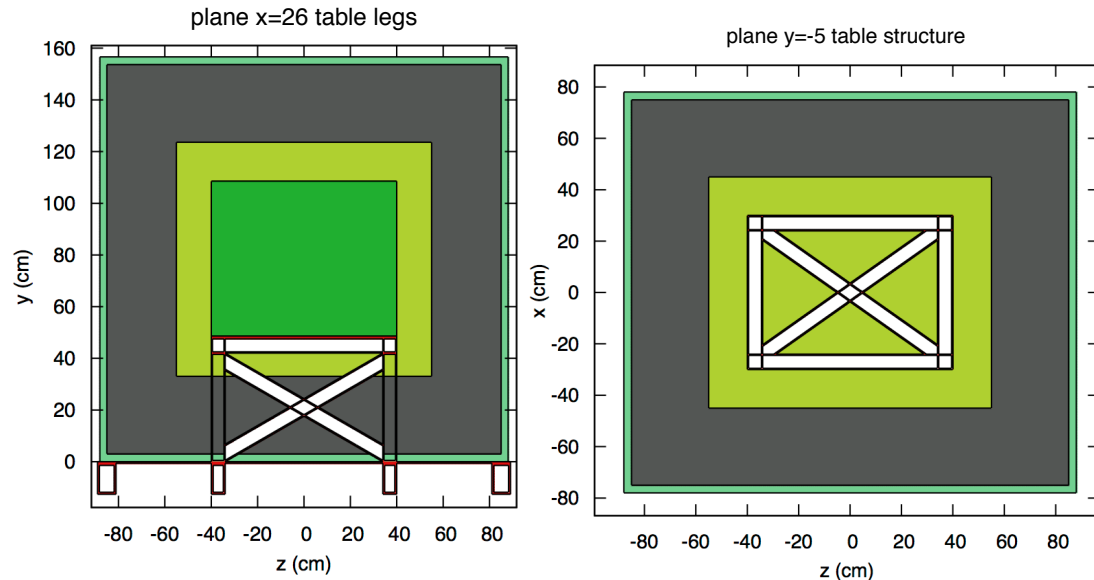


Figure 4 - Table legs and table structure, implemented in FLUKA.

1.2 Fluxes and Power Deposition in the Targets and Shielding

For all the calculations presented in this report, a 40 μA deuteron beam was assumed. Figure 5 and Figure 6 show the average photon and neutron fluxes in the assembly for two different thicknesses in the x direction: 200 cm and 80 cm, respectively. The neutron fluxes go up to 10^{14} neutrons/ cm^2/s in the target and decrease to approximately 10^6 neutrons/ cm^2/s after the shielding, an acceptable value which shows that the materials and the dimensions of the shielding are appropriate for this system. The photon fluxes range from 10^{13} photons/ cm^2/s in the target to approximately 10^9 photons/ cm^2/s outside the shielding. In this case, the lead diffuser is shielding the photons that come from the target effectively, but new photons are being produced in neutron capture reactions in polyethylene, and this secondary source of photons is responsible for the high fluxes outside the shielding.

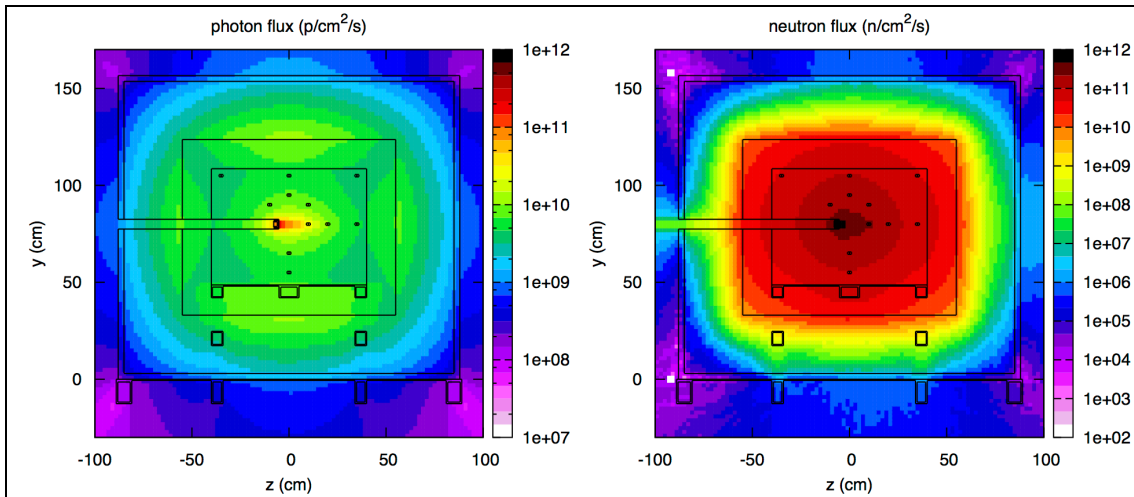


Figure 5 - Photon (left) and neutron (right) fluxes in the target and shielding (average values for a 200 cm thickness in the x direction).

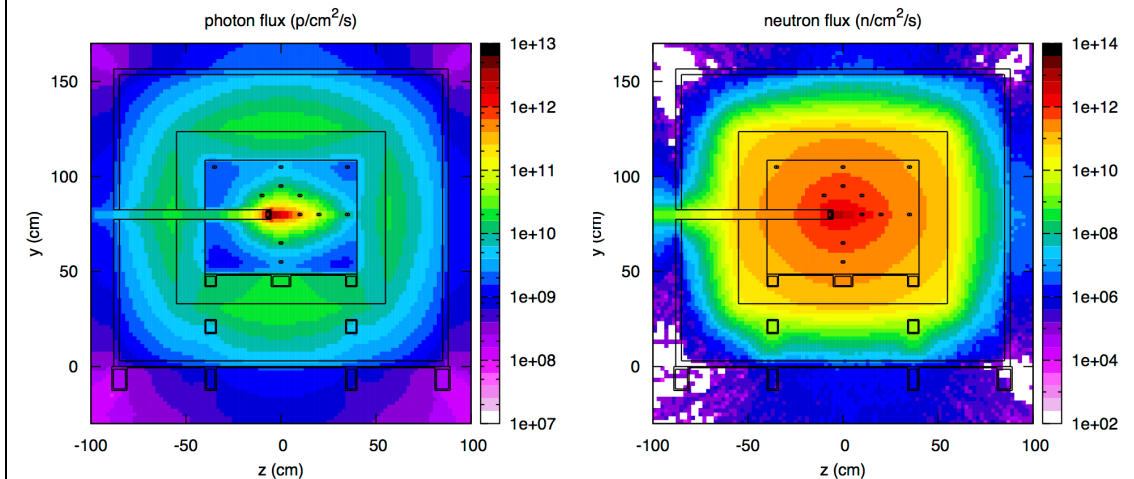


Figure 6 - Photon (left) and neutron (right) fluxes in the target and shielding (average values for an 8 cm thickness in the x direction).

Figure 7 shows the power deposition in the whole system and Figure 8 the power deposition in the target. It is important to highlight that most of the beam power is deposited in the first millimeter of the target, where up to 10 kW/cm³ are deposited.

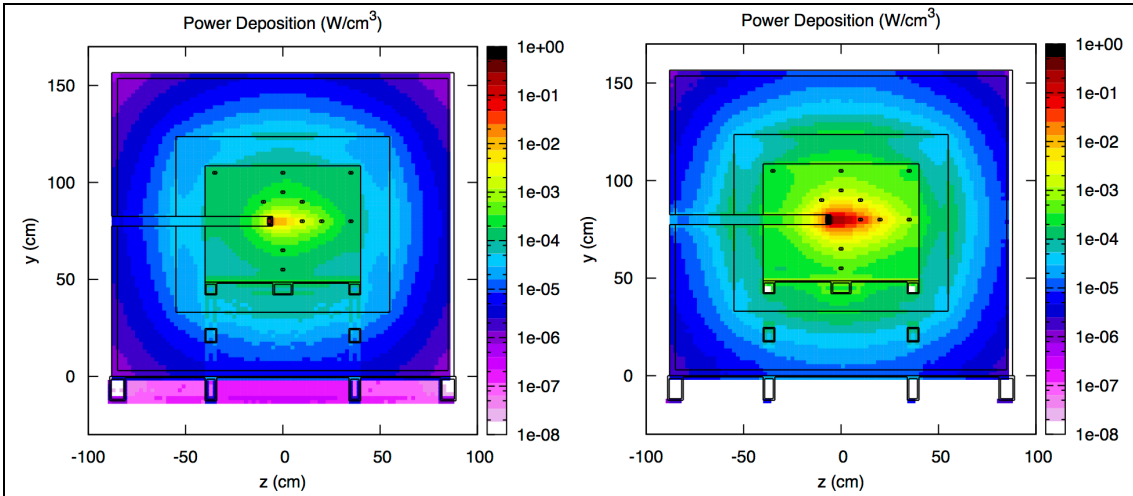


Figure 7 - Power deposition in the target and shielding. Left - average values for a 200 cm thickness in the x direction. Right - average values for an 8 cm thickness in the x direction.

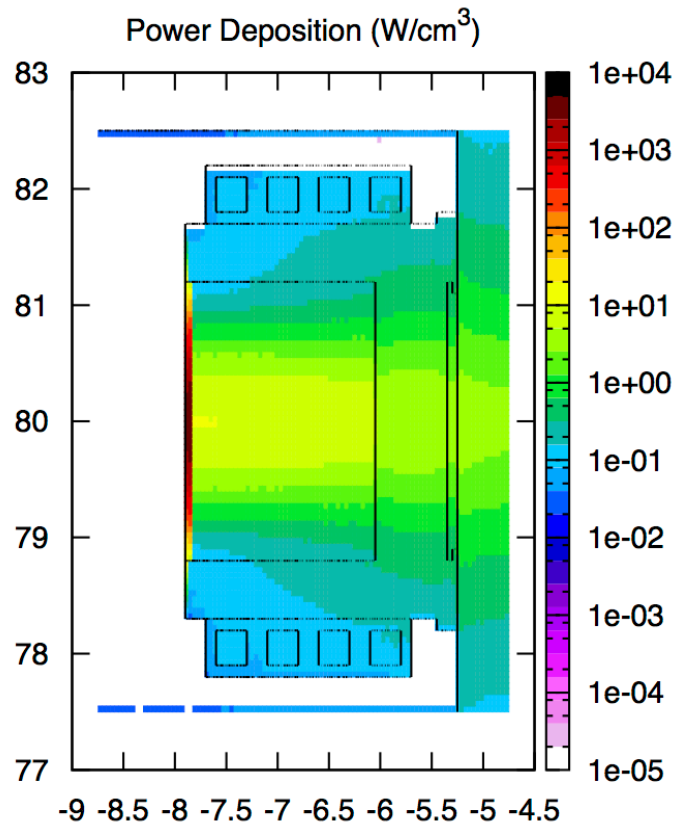


Figure 8 - Power deposition in the target (average values for a 1 cm thickness in the x direction).

The neutron fluxes in the target, lead diffuser, and after the shielding are shown in Figure 9, Figure 10 and Figure 11, respectively. In the target, there is a peak centered at 7.5 MeV. This peak is still seen in the lead and outside the shielding, although it is reduced from 2×10^{15} neutrons/cm²/s in the target to less than 10^6 neutrons/cm²/s

outside. The effects of neutron moderation can be seen inside the lead (Figure 10), with an already pronounced reduction in the flux of 7.5 MeV neutrons, and two "shoulders" centered at 0.05 eV and 500 keV, approximately.

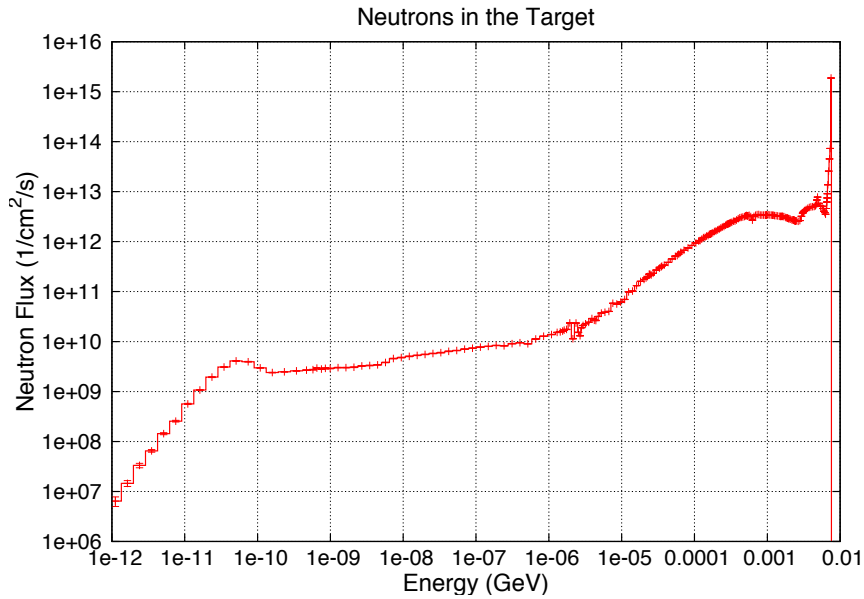


Figure 9 - Neutron flux spectrum in the target.

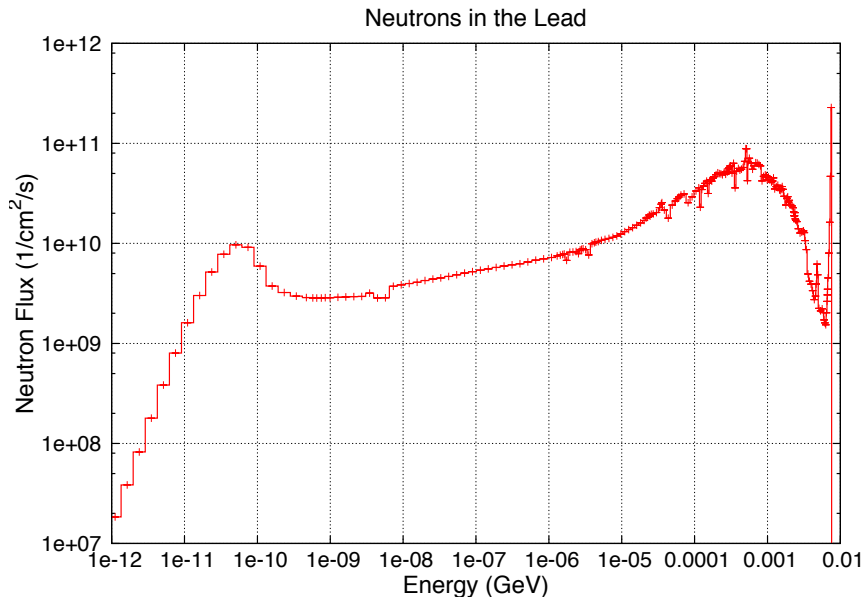
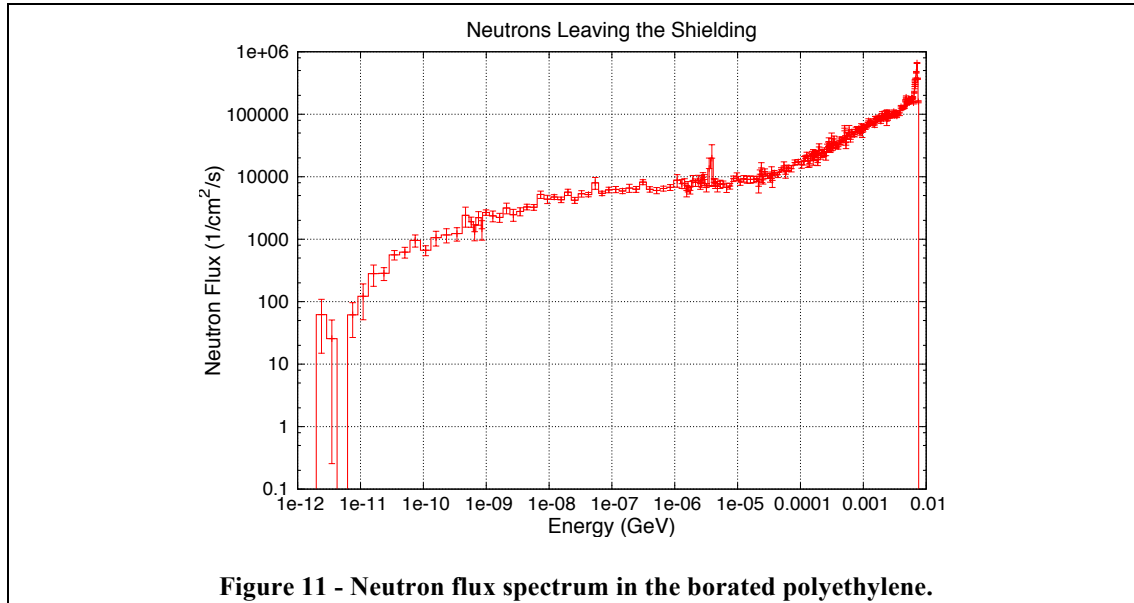


Figure 10 - Neutron flux spectrum in the lead.

Outside the shielding, the benefit of using polyethylene with an outer layer of polyethylene is evident (Figure 11): the "shoulder" of low-energy neutrons disappeared completely, due to neutron absorption, and the fluxes are in general much lower when compared to the neutron fluxes in the lead. This effect was also seen in Figure 5 and Figure 6.



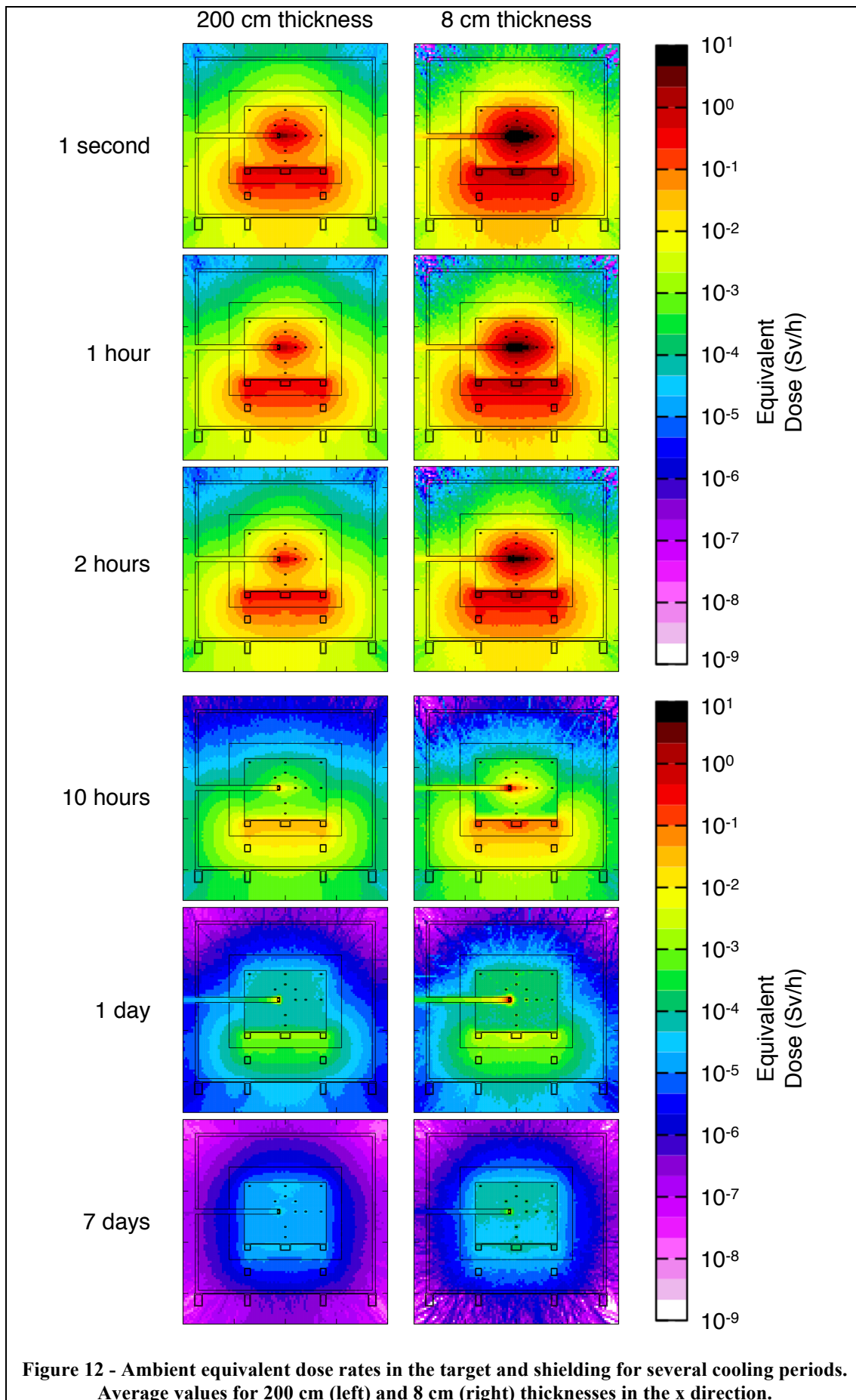
1.3 Residual Dose Equivalent Rates

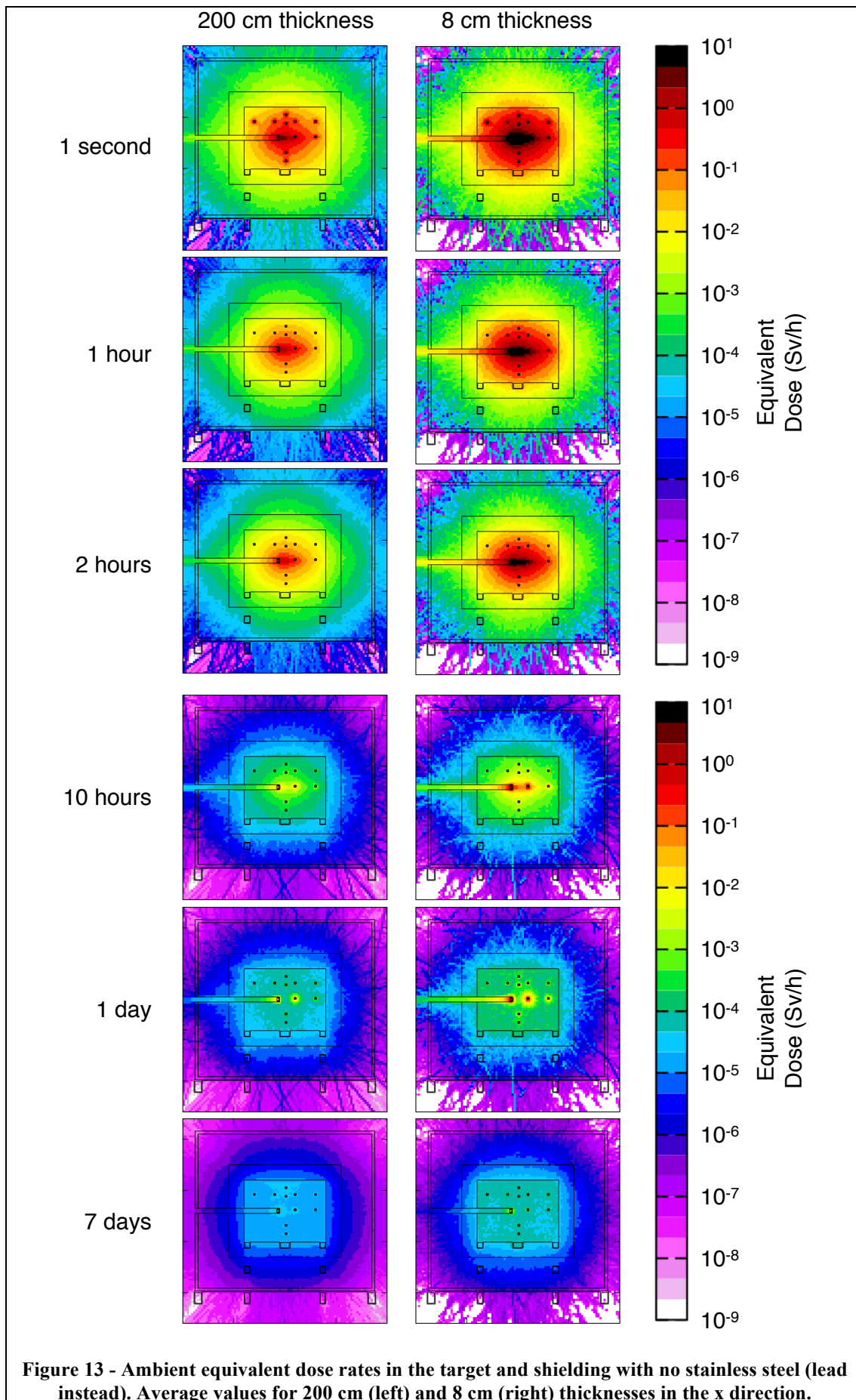
The residual equivalent dose rates (Sv/h) in the assembly are presented in Figure 12, for six cooling periods after the stoppage of the deuteron beam: 1 second, 1 hour, 2 hours, 10 hours, 1 day and seven days. For the irradiation profile, it was assumed that the system operates continuously for one hour at 40 μ A. Once again, the results are presented twice, with average values for 200 cm (left side of Figure 12) and 8 cm (right side of Figure 12) thicknesses in the x direction. The dose rates outside the shielding range from approximately 10-100 mSv/h one second after the beam stops to approximately 1 μ Sv/h ten days after. The highest residual dose rates are below the assembly, due to the activation of stainless steel. The composition of the stainless steel used in the simulations (S235) is presented in Table 1.

Table 1 - Stainless steel (S235) composition.

C	Mn	P	S	N	Cu	Fe
0.17%	1.4%	0.04%	0.04%	0.012%	0.55%	97.788%

In order to confirm that these high residual dose rates are mostly due to the activation of stainless steel, the simulations were repeated for with lead replacing the stainless steel in the structural components. The results, presented in Figure 13, confirm that the stainless steel has indeed a sizable contribution to the residual dose rates outside the shielding. Without this contribution, a dose rate of 1 μ Sv/h outside the shielding could be reached within one day after the stoppage of the beam, instead of seven days. Furthermore, the distribution of the dose rates is more "isotropic", centered on the target. An alternative to the change of material would be to increase shielding below the target, by increasing the height of the table.





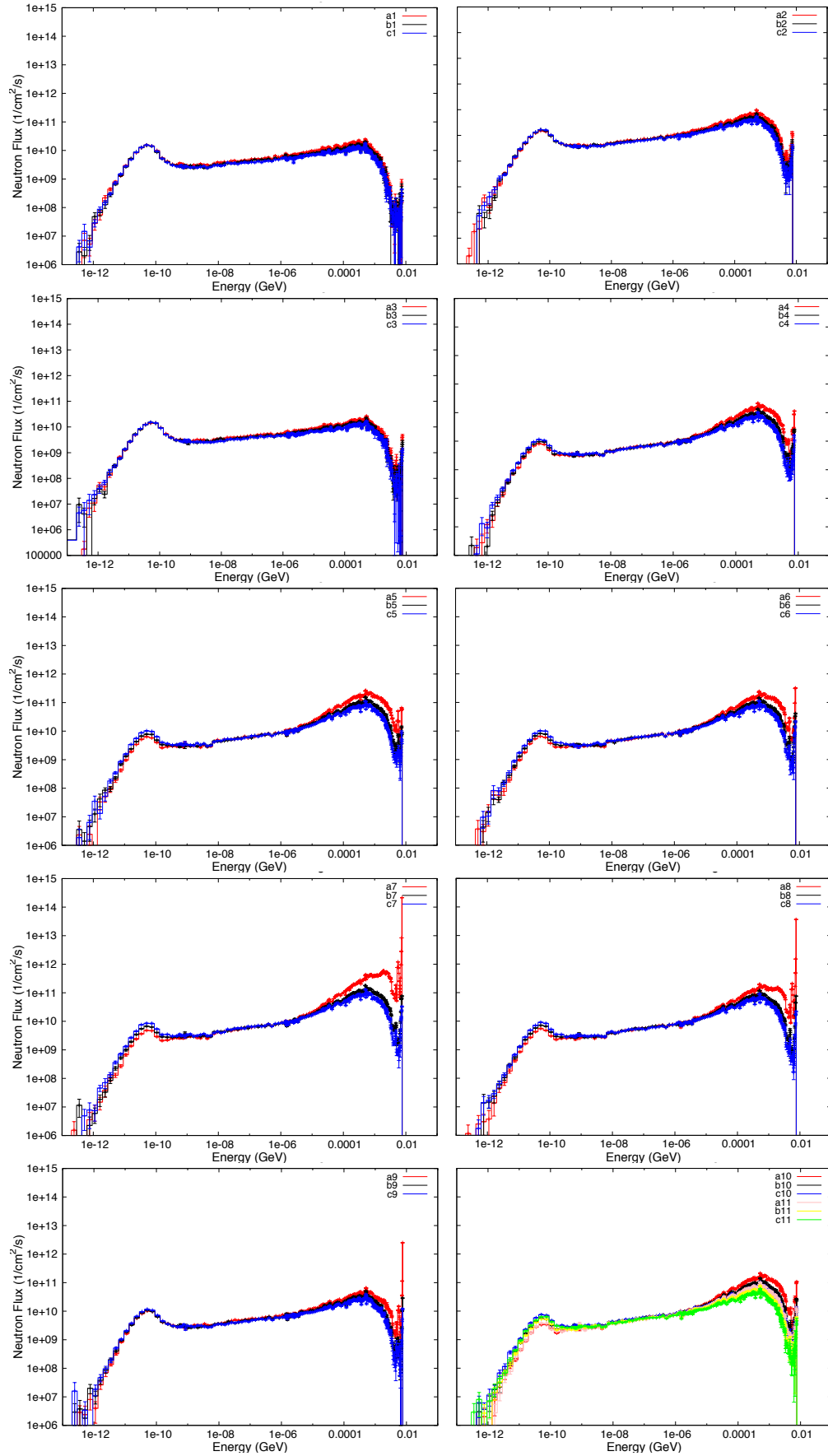


Figure 14 - Neutron fluxes (1/cm²/s) in the thirty-three 98-Mo targets.

1.4 Neutron Fluxes in the Samples

For reference, the neutron fluxes in all thirty-three samples are shown in Figure 14. The highest fluxes are in the samples "a7", "a8" and "a9", which are located immediately behind the target and receive the high-energy neutrons emitted in the direction of the beam. The ones located farther from the target ("a1", "b1", "c1", "a3", "b3", "c3", etc.) receive the lowest fluxes.

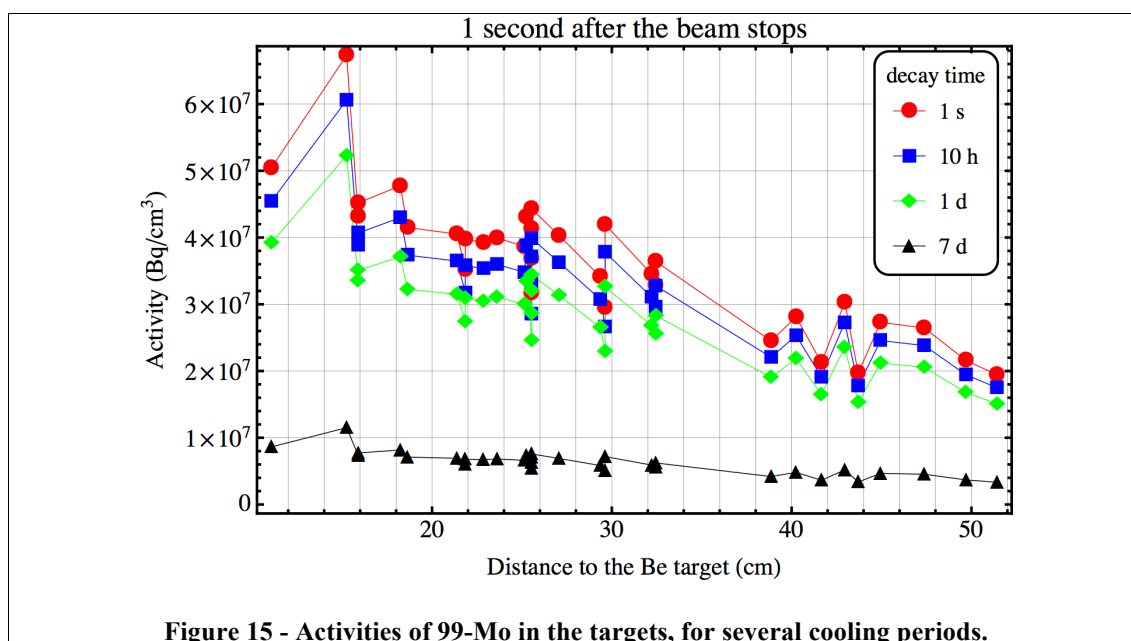
1.5 99-Mo Production

Table 2 - Activities of 99-Mo in the targets, for several cooling periods.

	d	Prompt	1s	1h	2h	10h	1d	7d	Error
	cm	1/cm ³ /s	Bq/cm ³						%
a1	38.9	2.35E9	2.45E7	2.42E7	2.40E7	2.21E7	1.90E7	4.19E6	3.95
a2	25.5	4.24E9	4.43E7	4.38E7	4.34E7	3.99E7	3.44E7	7.57E6	4.41
a3	47.4	2.53E9	2.65E7	2.62E7	2.59E7	2.38E7	2.06E7	4.52E6	5.47
a4	15.9	4.33E9	4.52E7	4.47E7	4.42E7	4.07E7	3.51E7	7.73E6	3.71
a5	11.1	4.83E9	5.05E7	4.99E7	4.94E7	4.54E7	3.92E7	8.63E6	3.85
a6	18.2	4.57E9	4.78E7	4.73E7	4.68E7	4.30E7	3.71E7	8.17E6	3.14
a7	15.3	6.45E9	6.73E7	6.66E7	6.59E7	6.06E7	5.23E7	1.15E7	3.33
a8	25.3	4.13E9	4.31E7	4.26E7	4.22E7	3.88E7	3.35E7	7.37E6	3.42
a9	40.3	2.69E9	2.81E7	2.78E7	2.75E7	2.53E7	2.19E7	4.81E6	5.55
a10	15.9	4.13E9	4.32E7	4.27E7	4.23E7	3.89E7	3.35E7	7.38E6	3.88
a11	25.5	3.03E9	3.17E7	3.13E7	3.10E7	2.85E7	2.46E7	5.42E6	5.58
b1	41.7	2.03E9	2.12E7	2.10E7	2.08E7	1.91E7	1.65E7	3.63E6	4.46
b2	29.6	4.02E9	4.20E7	4.15E7	4.11E7	3.78E7	3.26E7	7.18E6	4.38
b3	49.7	2.07E9	2.16E7	2.14E7	2.11E7	1.94E7	1.68E7	3.69E6	6.64
b4	21.9	3.81E9	3.98E7	3.93E7	3.89E7	3.58E7	3.09E7	6.80E6	3.28
b5	18.6	3.97E9	4.15E7	4.11E7	4.06E7	3.74E7	3.22E7	7.10E6	3.14
b6	23.6	3.83E9	4.00E7	3.95E7	3.91E7	3.60E7	3.11E7	6.83E6	4.05
b7	21.4	3.88E9	4.05E7	4.01E7	3.97E7	3.65E7	3.15E7	6.93E6	4.64
b8	29.4	3.27E9	3.41E7	3.38E7	3.34E7	3.07E7	2.65E7	5.83E6	4.06
b9	43.0	2.90E9	3.03E7	3.00E7	2.97E7	2.73E7	2.35E7	5.18E6	5.49
b10	21.9	3.37E9	3.52E7	3.49E7	3.45E7	3.17E7	2.74E7	6.02E6	4.81
b11	29.6	2.83E9	2.95E7	2.92E7	2.89E7	2.66E7	2.29E7	5.05E6	5.02
c1	43.7	1.89E9	1.97E7	1.95E7	1.93E7	1.77E7	1.53E7	3.37E6	6.33
c2	32.4	3.49E9	3.64E7	3.60E7	3.57E7	3.28E7	2.83E7	6.23E6	5.24
c3	51.4	1.86E9	1.94E7	1.92E7	1.90E7	1.75E7	1.51E7	3.32E6	5.07
c4	25.5	3.95E9	4.13E7	4.09E7	4.04E7	3.72E7	3.21E7	7.06E6	3.99
c5	22.9	3.76E9	3.93E7	3.89E7	3.84E7	3.53E7	3.05E7	6.71E6	4.54
c6	27.1	3.86E9	4.03E7	3.99E7	3.95E7	3.63E7	3.13E7	6.89E6	4.35
c7	25.2	3.70E9	3.86E7	3.82E7	3.78E7	3.48E7	3.00E7	6.61E6	4.44
c8	32.2	3.31E9	3.45E7	3.42E7	3.38E7	3.11E7	2.68E7	5.91E6	3.45
c9	44.9	2.61E9	2.73E7	2.70E7	2.67E7	2.46E7	2.12E7	4.67E6	4.58
c10	25.5	3.53E9	3.69E7	3.65E7	3.61E7	3.32E7	2.87E7	6.31E6	4.89
c11	32.4	3.15E9	3.29E7	3.25E7	3.22E7	2.96E7	2.55E7	5.62E6	3.46

Finally, the production of 99-Mo in each sample is presented in Table 2, during operation and after the already-mentioned cooling periods. One second after the stoppage of the beam, the activities of 99-Mo in the sample range from 1.97×10^7

Bq/cm³ in target "c1" to 6.73x10⁷ Bq/cm³ in target "a7". These results are also show in Figure 15, where the activities are plotted against the distance of the 99-Mo target to the Beryllium target. Even though the target "a5" is closer than "a7", the neutron flux and resulting activities are lower, since "a7" is behind the Beryllium target (in the direction of incidence of the deuteron beam) and receives a higher neutron flux.



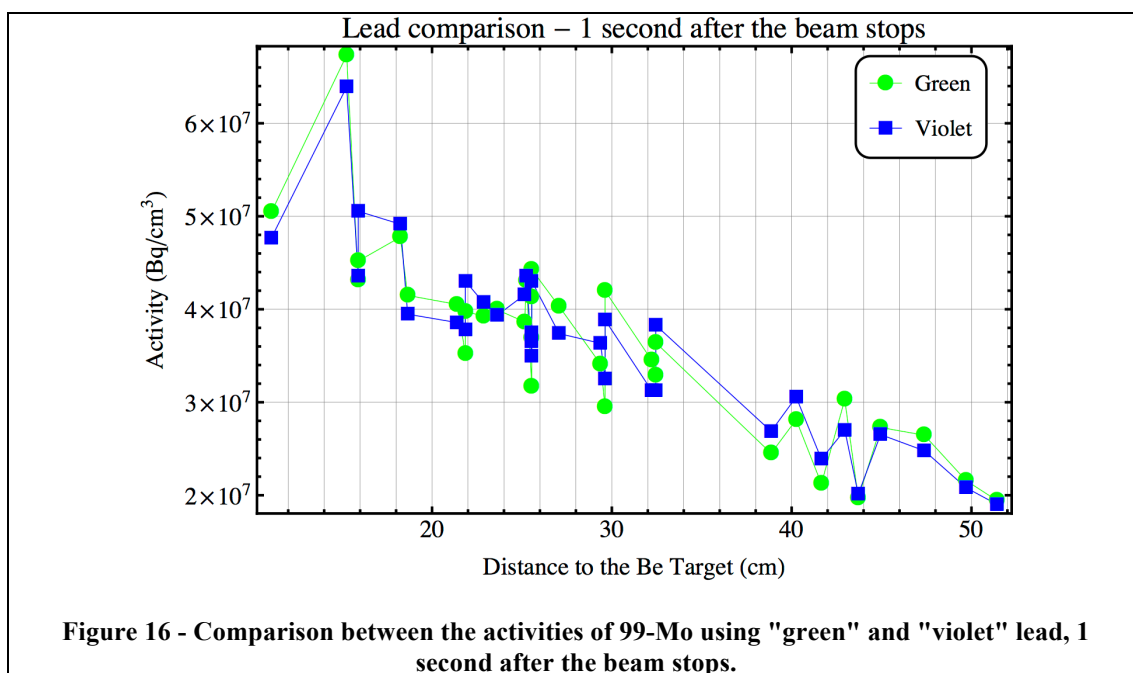
1.6 Lead Analysis

The next step consisted in comparing two different types of lead, to check how the purity of the lead affects the activities of the 99-Mo targets. The composition of the two materials, referred to as "Green" lead and "Violet" lead, are presented in Table 3. Their purities are of 99.97% and 99.99%, respectively.

Table 3 - "Green" and "Violet" lead compositions.

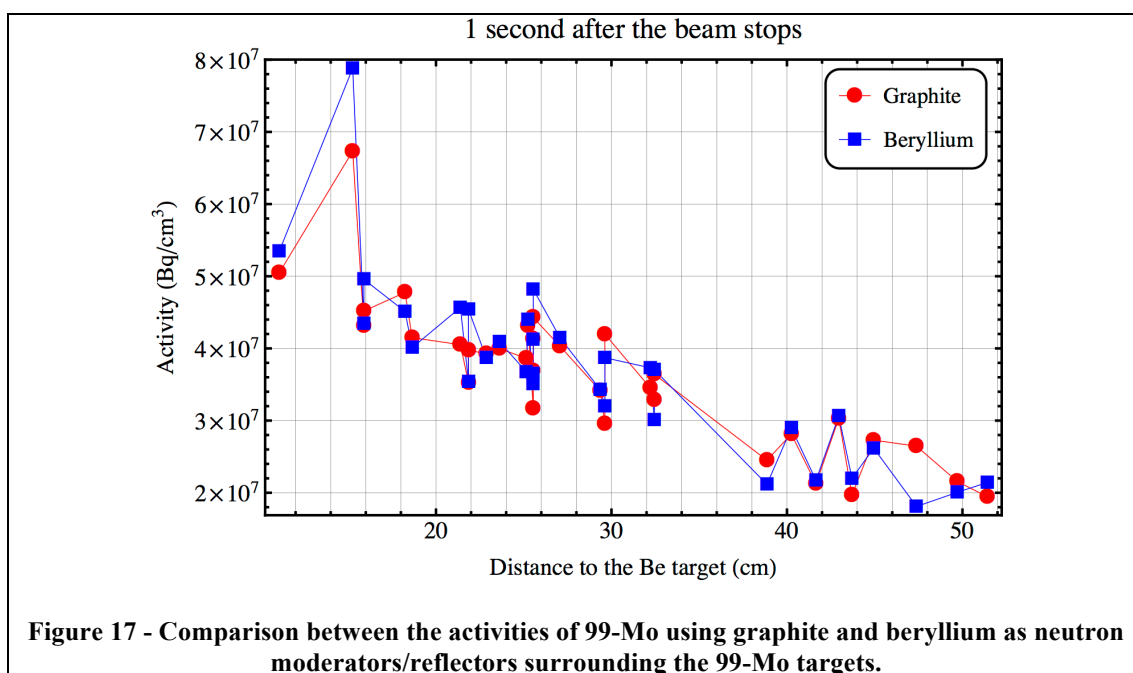
Lead	Sb (%)	Sn (%)	As (%)	Bi (%)	Cu (%)	Ag (%)	Cd (%)	Ni (%)	Zn (%)
Green	0.001	0.001	0.001	0.03	0.003	0.005	0.001	0.001	0.0005
Violet	0.0005	0.0005	0.0005	0.01	0.0005	0.0015	0.0002	0.0002	0.0002

The activities obtained with the two types of lead are shown in Figure 15, for a cooling period of one second. The deviations between the two configurations are very small, within the statistical uncertainties of the simulations. The "Green" lead should be preferred in this case, since it is less expensive and exhibits a similar performance, from the neutronics point of view.



1.7 Graphite vs. Beryllium as Reflectors for the 99-Mo Targets

Another analysis made with this configuration was the comparison of graphite and beryllium as neutron moderators/reflectors surrounding the 99-Mo targets. The results (Figure 17) show that there is no difference between the two. Once again, the preference goes for the least expensive: graphite.



2 Second Configuration

2.1 Main Changes in the Geometry

At this stage, the geometry model used in FLUKA was updated, to describe the target system with more detail. The major changes were made to the 98-Mo targets, which have now a new configuration and a different distribution in the lead. The new distribution is shown in Figure 18, for planes $x=0$ and $z=1$. There are thirty targets now, down from the previous thirty-three, and they are inserted inside aluminum tubes, each tube carrying three targets and numbered from 1 to 10.

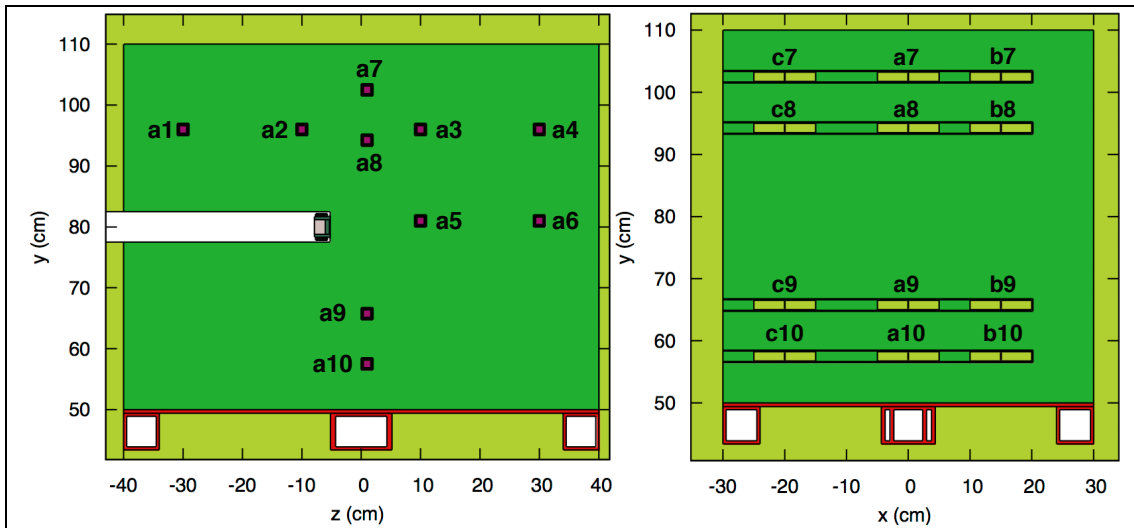


Figure 18 - New distribution of the 98-Mo targets, planes $x=0$ (left) and $z=1$ (right).

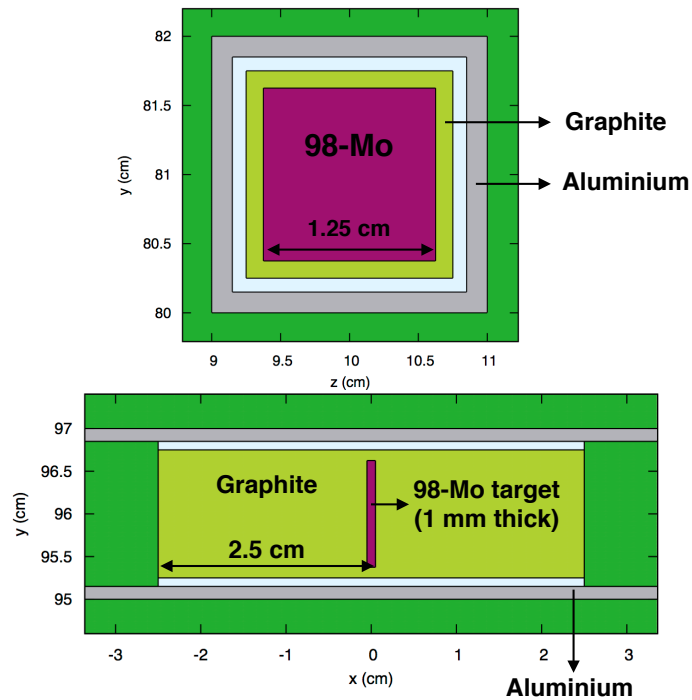


Figure 19 - New configuration of the 98-Mo targets, planes $x=0$ (top) and $z=1$ (bottom).

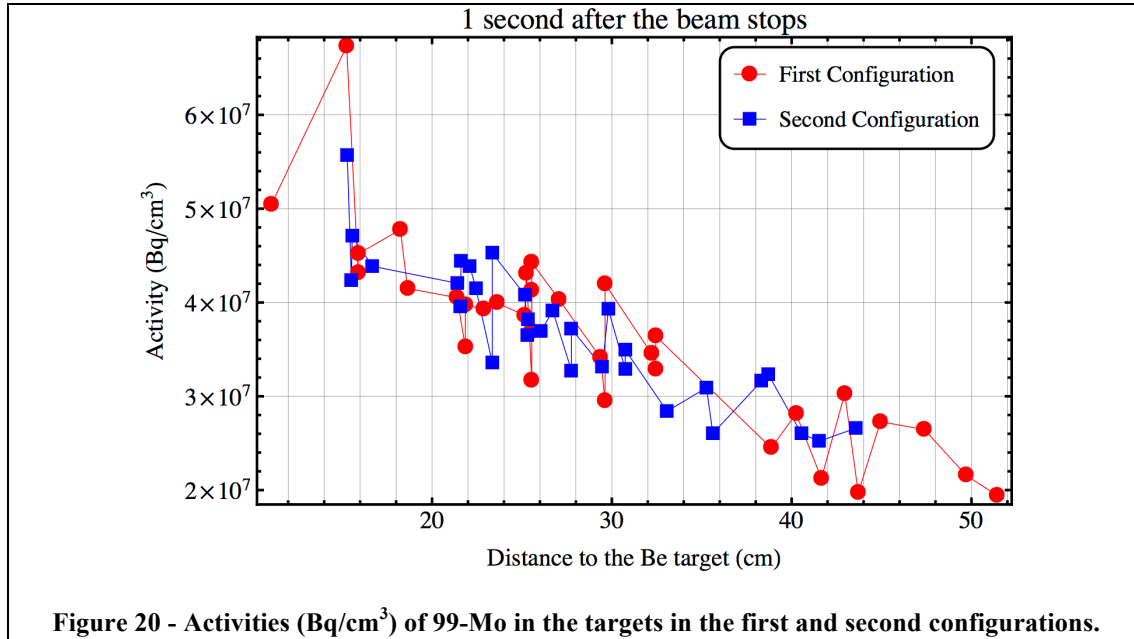


Table 4 - Activities of 99-Mo in the targets, for the second configuration.

	d	Prompt	1s	1h	2h	10h	1d	7d	Error
	cm	1/cm ³ /s	Bq/cm ³						%
a1	29.5	3.17E9	3.31E7	3.28E7	3.24E7	2.98E7	2.57E7	5.67E6	3.49
a2	16.7	4.20E9	4.39E7	4.34E7	4.29E7	3.95E7	3.41E7	7.50E6	5.54
a3	22.1	4.20E9	4.39E7	4.34E7	4.29E7	3.95E7	3.41E7	7.50E6	2.35
a4	38.7	3.09E9	3.23E7	3.20E7	3.16E7	2.91E7	2.51E7	5.52E6	10.70
a5	15.3	5.33E9	5.57E7	5.51E7	5.45E7	5.01E7	4.33E7	9.52E6	5.46
a6	35.3	2.96E9	3.09E7	3.06E7	3.03E7	2.78E7	2.40E7	5.29E6	8.40
a7	23.4	4.33E9	4.53E7	4.48E7	4.43E7	4.07E7	3.52E7	7.74E6	5.23
a8	15.6	4.51E9	4.71E7	4.66E7	4.61E7	4.24E7	3.66E7	8.05E6	5.38
a9	15.5	4.05E9	4.23E7	4.19E7	4.14E7	3.81E7	3.29E7	7.24E6	6.36
a10	23.4	3.21E9	3.36E7	3.32E7	3.29E7	3.02E7	2.61E7	5.74E6	6.88
b1	33.1	2.72E9	2.84E7	2.81E7	2.78E7	2.56E7	2.21E7	4.86E6	4.80
b2	22.4	3.97E9	4.15E7	4.11E7	4.06E7	3.74E7	3.22E7	7.10E6	5.71
b3	26.7	3.75E9	3.91E7	3.87E7	3.83E7	3.52E7	3.04E7	6.69E6	6.65
b4	41.5	2.41E9	2.52E7	2.49E7	2.47E7	2.27E7	1.96E7	4.31E6	10.87
b5	21.4	4.03E9	4.20E7	4.16E7	4.12E7	3.79E7	3.27E7	7.19E6	5.07
b6	38.3	3.03E9	3.16E7	3.13E7	3.09E7	2.85E7	2.46E7	5.41E6	8.45
b7	27.8	3.56E9	3.72E7	3.68E7	3.64E7	3.35E7	2.89E7	6.36E6	7.49
b8	21.6	4.25E9	4.44E7	4.40E7	4.35E7	4.00E7	3.45E7	7.60E6	5.71
b9	21.6	3.79E9	3.95E7	3.91E7	3.87E7	3.56E7	3.07E7	6.76E6	6.76
b10	27.8	3.13E9	3.27E7	3.24E7	3.20E7	2.95E7	2.54E7	5.60E6	6.02
c1	35.6	2.49E9	2.60E7	2.58E7	2.55E7	2.34E7	2.02E7	4.45E6	4.64
c2	26.0	3.53E9	3.69E7	3.65E7	3.61E7	3.32E7	2.87E7	6.31E6	3.72
c3	29.8	3.76E9	3.93E7	3.89E7	3.84E7	3.53E7	3.05E7	6.71E6	6.75
c4	43.6	2.55E9	2.66E7	2.63E7	2.60E7	2.39E7	2.07E7	4.55E6	9.19
c5	25.2	3.91E9	4.08E7	4.04E7	3.99E7	3.67E7	3.17E7	6.98E6	7.72
c6	40.5	2.49E9	2.60E7	2.58E7	2.55E7	2.34E7	2.02E7	4.45E6	9.85
c7	30.7	3.35E9	3.49E7	3.46E7	3.42E7	3.15E7	2.72E7	5.98E6	6.31
c8	25.3	3.65E9	3.82E7	3.78E7	3.74E7	3.43E7	2.96E7	6.52E6	7.58
c9	25.3	3.49E9	3.65E7	3.61E7	3.57E7	3.28E7	2.83E7	6.24E6	5.31
c10	30.7	3.15E9	3.29E7	3.25E7	3.22E7	2.96E7	2.55E7	5.62E6	9.16

2.2 Comparison Between Configurations

2.2.1 Dimensions of the 98-Mo Targets and Reflectors

Four configurations were tested at the same time, using two different thicknesses for the 98-Mo targets (0.1 and 1 cm) and two different thicknesses for the corresponding reflectors (2.5 cm and 5 cm). The configuration presented in Figure 19 corresponds to a target with a thickness of 0.1 cm and 2.5 cm of graphite on each side. The results, presented in Figure 21, show that there is no advantage in using 1 cm thick targets: increasing the thickness of the targets increases the total activity but decreases the specific activity. Concerning the size of the reflectors, the choice between using 2.5 cm or 5 cm is not clear. For some positions, there is advantage in using 5 cm of graphite on each side of the targets, like at position "a5", just behind the target. In other positions, the activities are higher with 2.5 cm of graphite. Since the decision is not clear in this case, the configuration proposed in Figure 19 was chosen as baseline configuration.

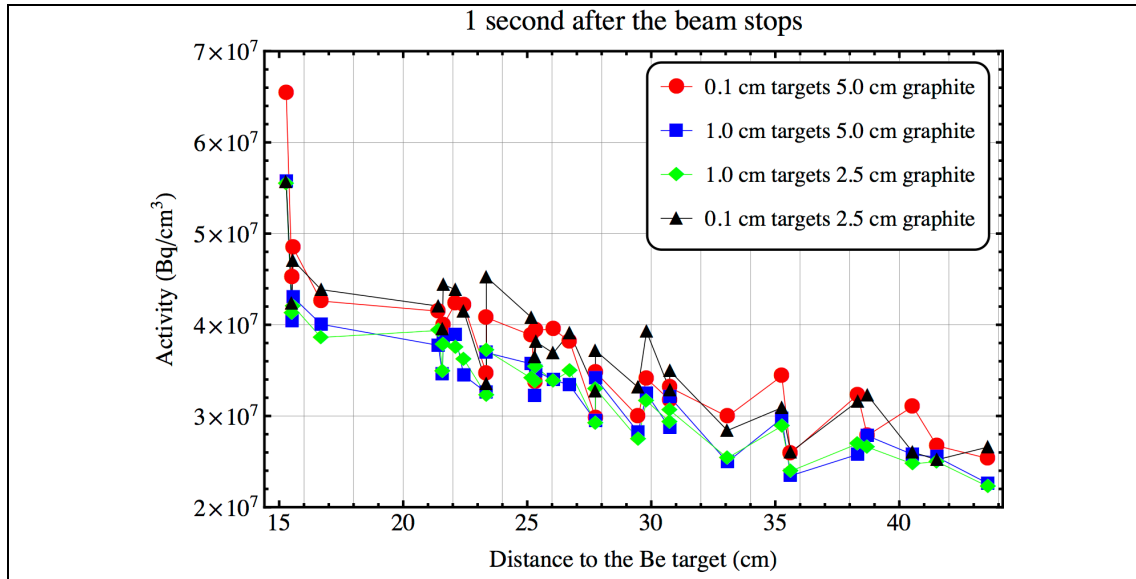
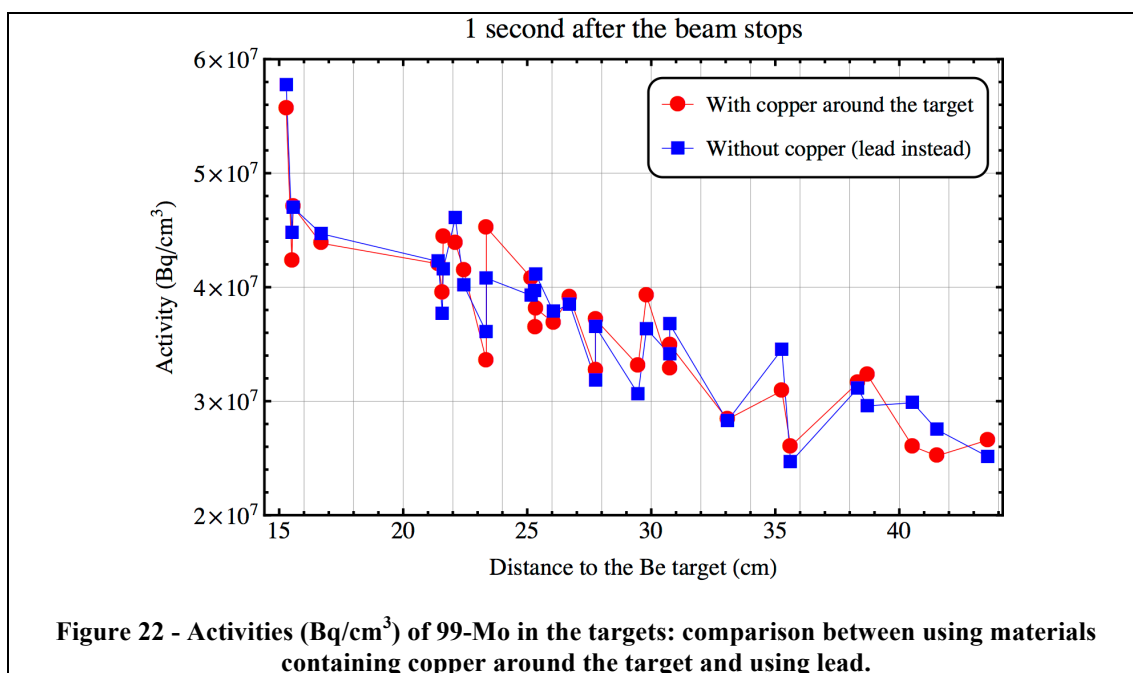


Figure 21 - Activities (Bq/cm³) of 99-Mo in the targets: comparison between different configurations.

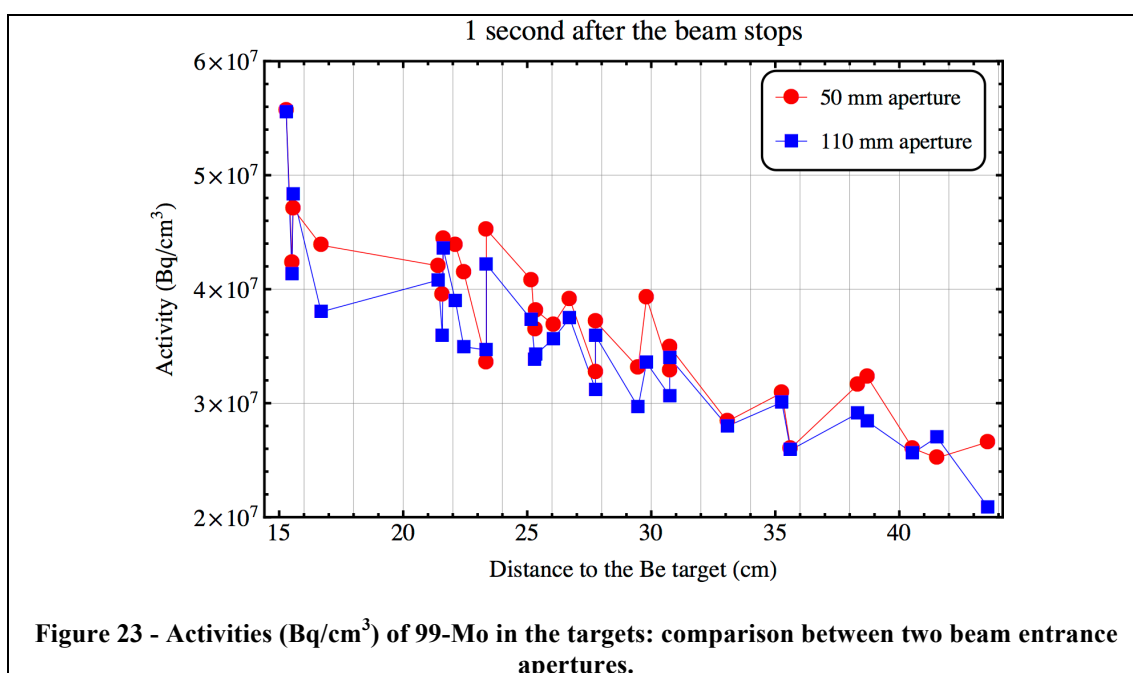
2.2.2 Copper-based Materials vs. Lead around the target

The effect of using materials containing copper around the target was also assessed. In the baseline configuration, the target is surrounded by two materials: pure copper (refrigerator circuit) and copper with 2.55% cobalt and 0.55% beryllium. To test if the quantity of captured neutrons might affect the activation of molybdenum, these materials were replaced by lead in the simulations. Figure 22 shows a comparison between the two configurations; it can be concluded that copper does not capture enough neutrons to have a visible effect on the activation of molybdenum.

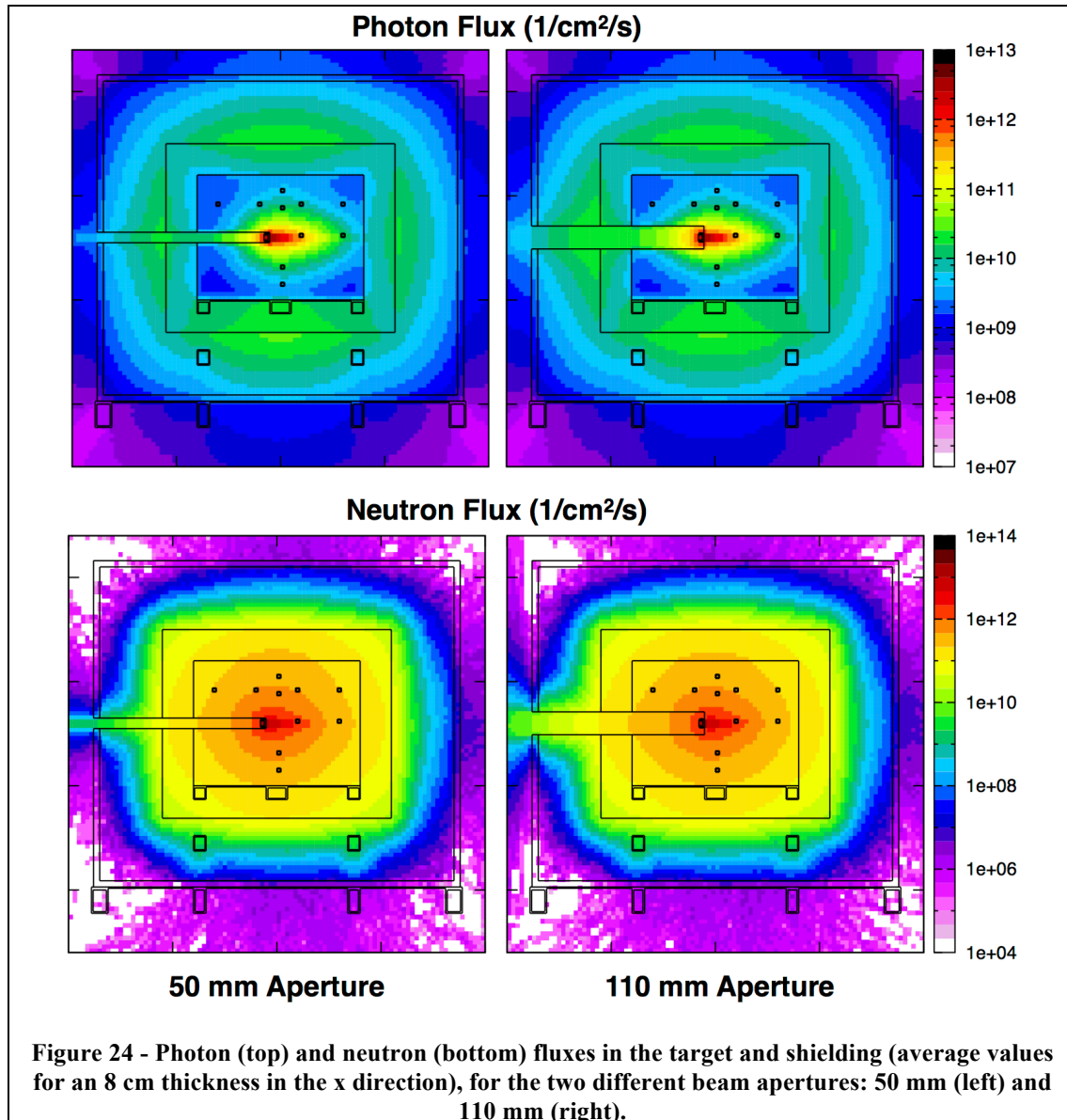


2.2.3 Beam Entrance Aperture

In the baseline design for the simulations, the beam entrance had an aperture with 50 mm of diameter. Due to practical constraints, this diameter had to be increased to 110 mm. Figure 23 shows the impact of this change in the activities, according to the FLUKA simulations. The decrease in the total activity of ⁹⁹-Mo (in the 30 targets) is of approximately 6%. This decrease depends on the distance of the molybdenum target to the beryllium target, going up to 27% in the most distant target. The results presented in the following sections are still for the configuration with a 50 mm aperture.



Regarding the neutron and photon fluxes escaping the system, Figure 24 shows that the leakage of neutrons and photons through the aperture increases with the 110 mm aperture. The same is true for the photon fluxes and equivalent dose rates after the stoppage of the beam, as shown in Figure 25.



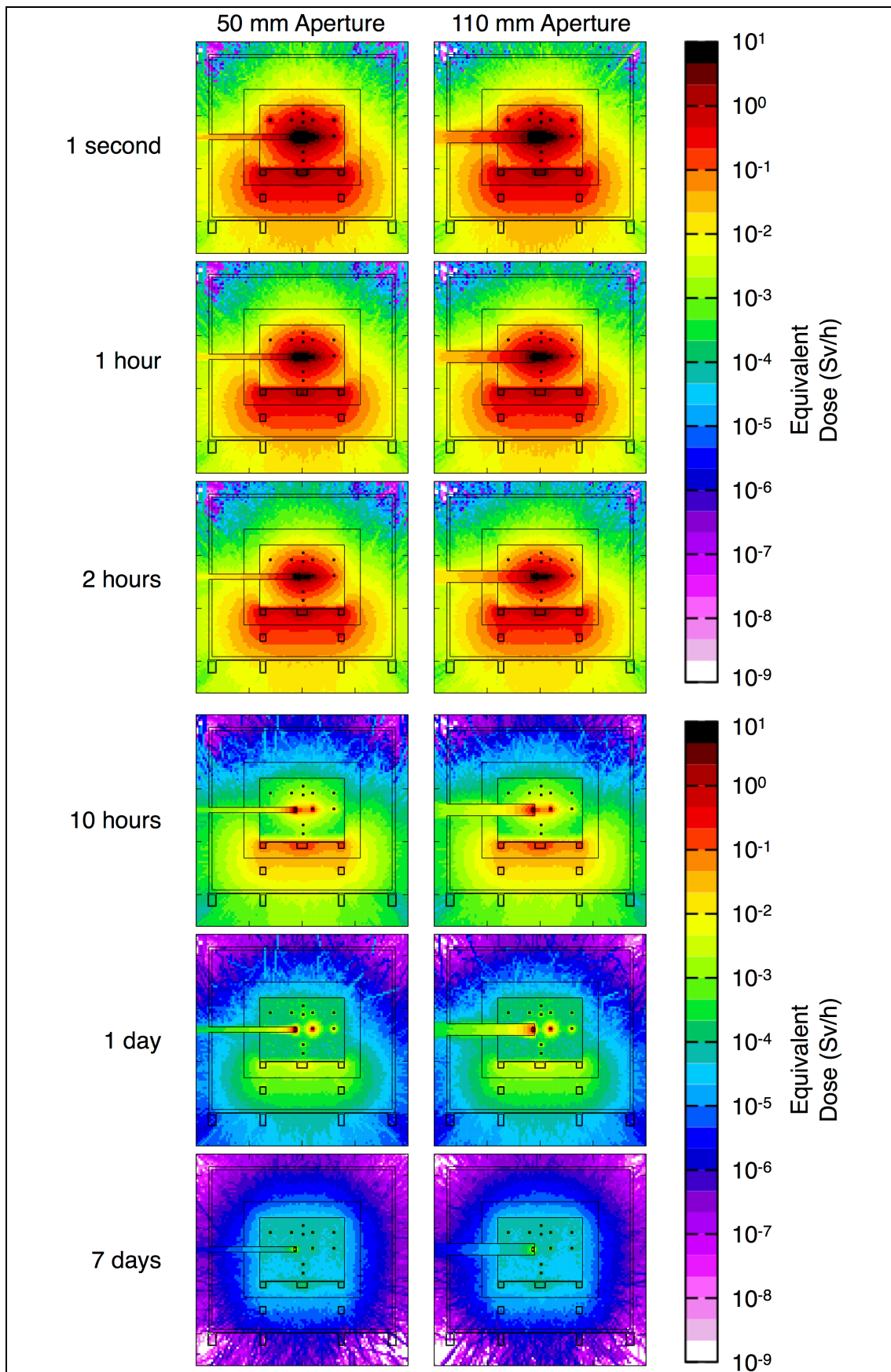


Figure 25 - Ambient equivalent dose rates in the target and shielding (average values for an 8 cm thickness in the x direction). Left - 50 mm aperture. Right - 110 mm aperture.

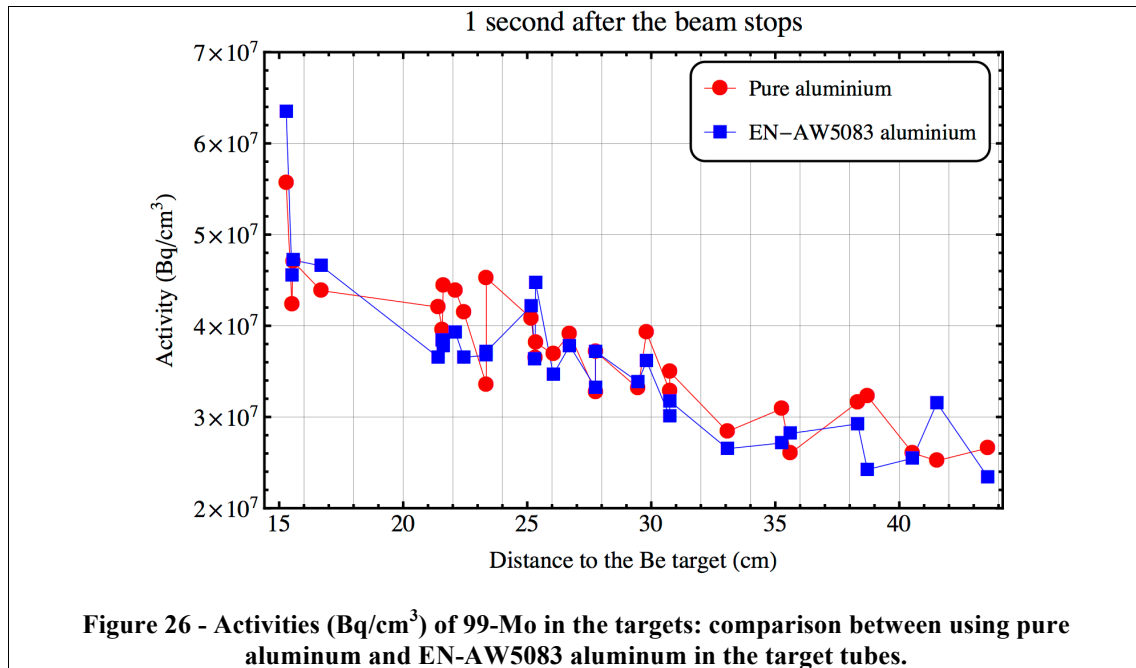
2.2.4 Target Tubes: Pure Aluminum vs. EN-AW5083

In the previous simulations, pure aluminum was used in the target tubes. The composition of the material that ended up being chosen for the tubes, EN-AW5083, is presented in Table 5. New simulations were run using this material, to check the effect on the residual dose rates and on the production of ⁹⁹-Mo.

Table 5 - EN-AW5083 composition.

Si	Fe	Cu	Mn	Mg	Cr	Zn	Ti	Al
0.4%	0.4%	0.1%	1%	4.9%	0.25%	0.25%	0.15%	92.55%

Figure 26 shows the activities of ⁹⁹-Mo, one second after the stoppage of the beam, with the two different types of aluminum. The differences are not significant: with each kind of aluminum, the activities are higher in some targets and lower in others.



As for the residual dose rates in the whole system, Figure 27 shows that there is no visible difference when the two types of aluminum are compared. More important, however, are the results shown in Figure 28, the residual equivalent dose rates produced by one of the ten target tubes (containing targets "a5", "b5" and "c5"), with all the other elements set to vacuum. Once again, there is no visible difference between the two types of aluminum. For the remaining simulations, EN-AW5083 will be used instead of pure aluminum.

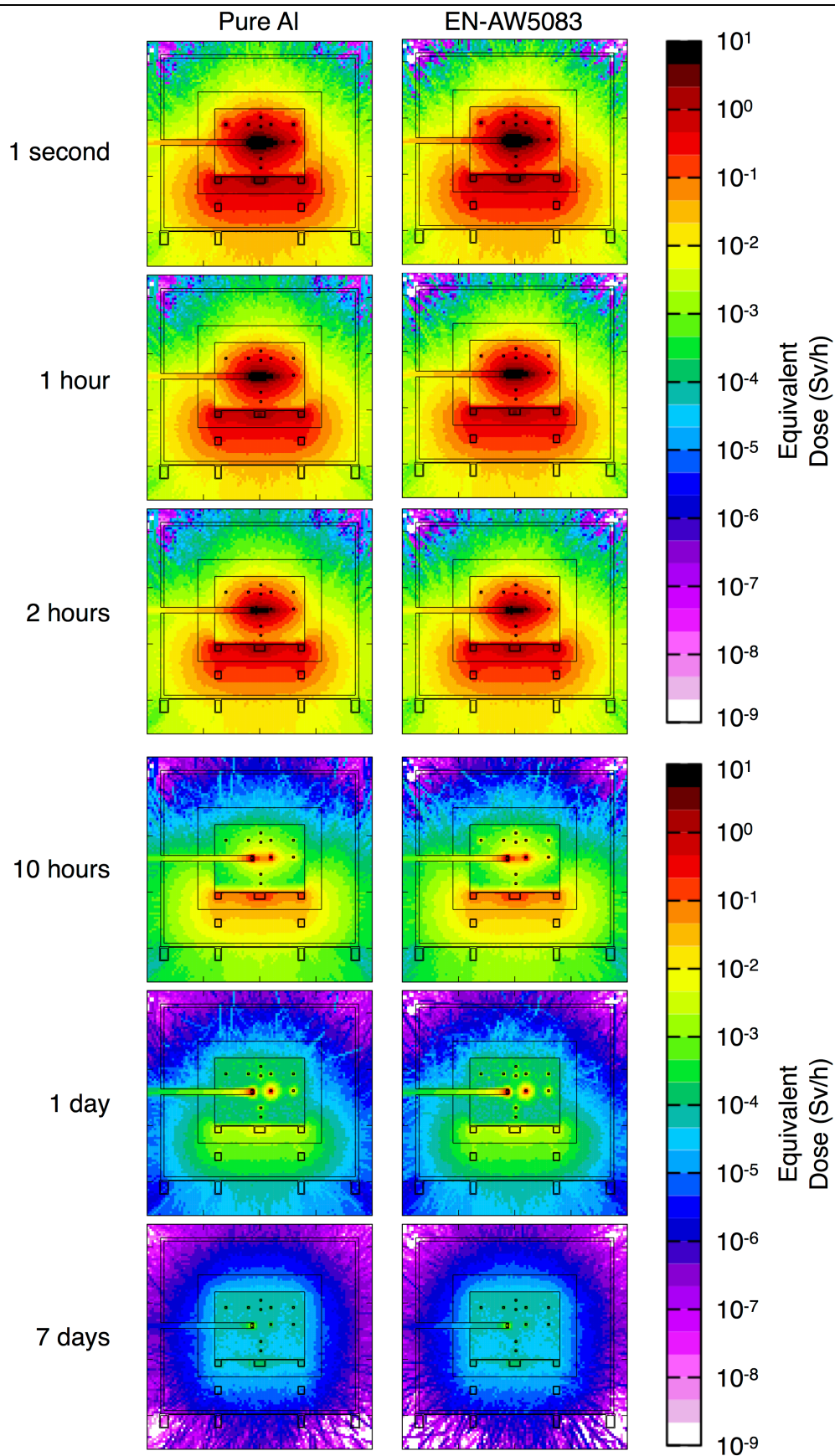


Figure 27 - Ambient equivalent dose rates in the target and shielding (average values for an 8 cm thickness in the x direction). Left - Pure Aluminum. Right - EN-AW5083.

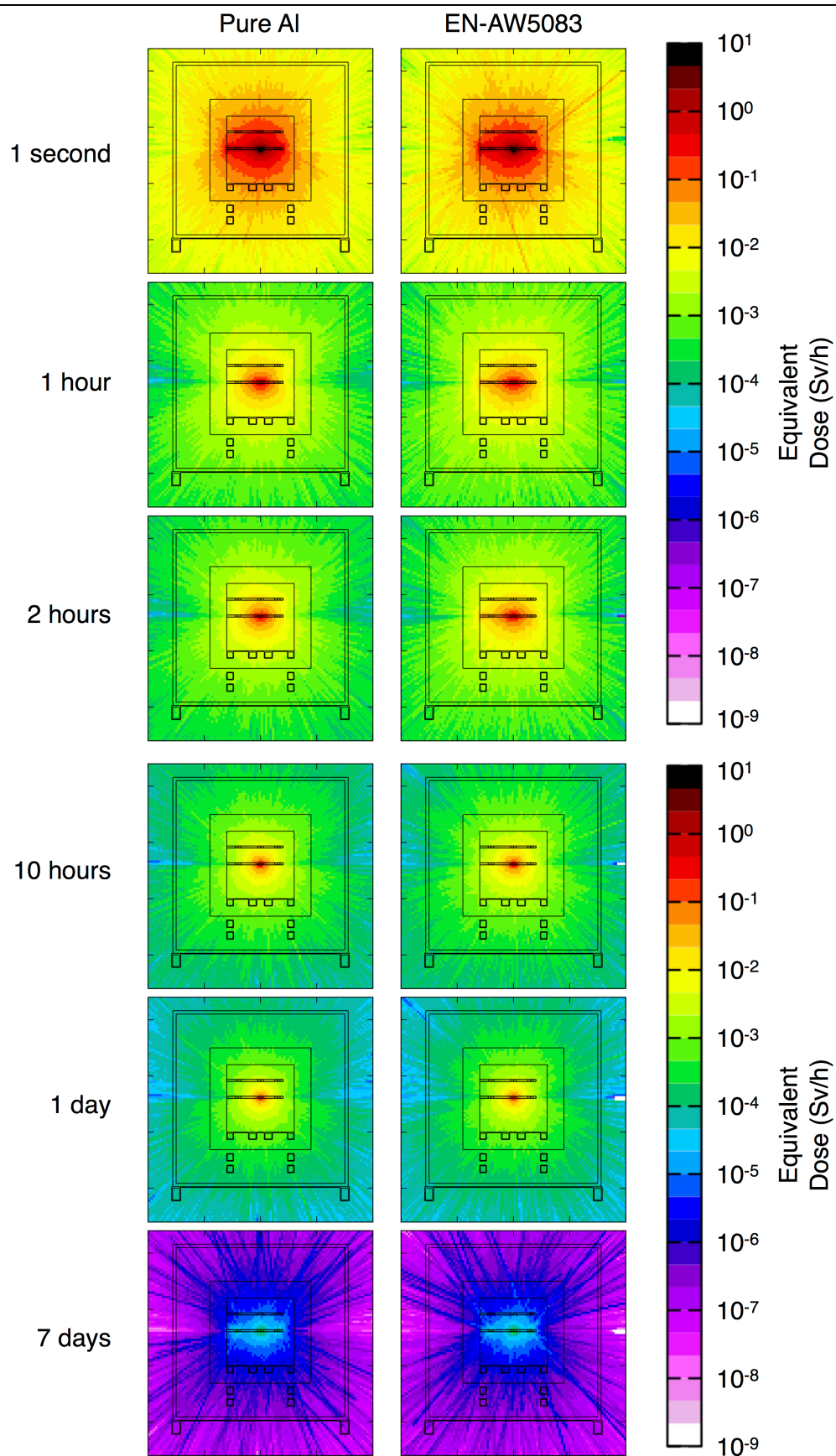


Figure 28 - Ambient equivalent dose rates (8 cm thickness), contribution from one tube (remaining components set to vacuum). Left - Pure Aluminum. Right - EN-AW5083.

2.2.5 Diffuser Material: Lead vs. Sodium

The next step consisted in comparing the production of 99-Mo using sodium and lead as neutron diffusers. The results, presented in Figure 29, show that using sodium instead of lead reduces the production of 99-Mo to a great extent. For this reason, this option was not further pursued.

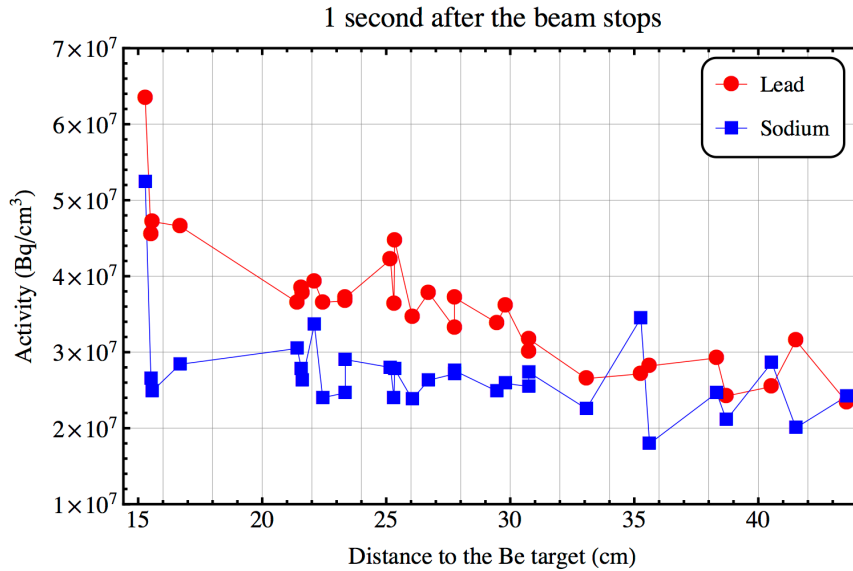


Figure 29 - Activities (Bq/cm³) of 99-Mo in the targets: comparison between using lead and sodium as diffuser material.

2.2.6 Extra Graphite Layer in the Diffuser

Another approach aiming at improving the production of 99-Mo was to insert an additional layer of graphite inside the diffuser, in order to further moderate the neutrons. Four configurations were tested (Figure 30):

- (10, 5, 15) - 10 cm of lead, followed by 5 cm of graphite and 15 cm of lead, (measured from the center of the assembly, in all directions).
- (15, 5, 10) - 15 cm of lead, followed by 5 cm of graphite and 10 cm of lead;
- (10, 10, 10) - 10 cm of lead, followed by 10 cm of graphite and 10 cm of lead;
- (10, 15, 5) - 10 cm of lead, followed by 15 cm of graphite and 5 cm of lead;

The activities in the targets one second after the stoppage of the beam for the four configurations are presented in Figure 31. When compared to the standard configuration, these results show that the introduction of the new graphite layer is indeed a good option, increasing the activities in all samples, and mainly on those closer to the beryllium target. Of the four configurations, the one with the highest production of 99-Mo is the one with 10 cm of graphite, followed by the one with 15 cm. An optimized value for the thickness of graphite should be between 10 and 15 cm (closer to 10). For target "a5", the one closest to the beryllium target, using a 10 cm layer of graphite triplicates the production of 99-Mo.

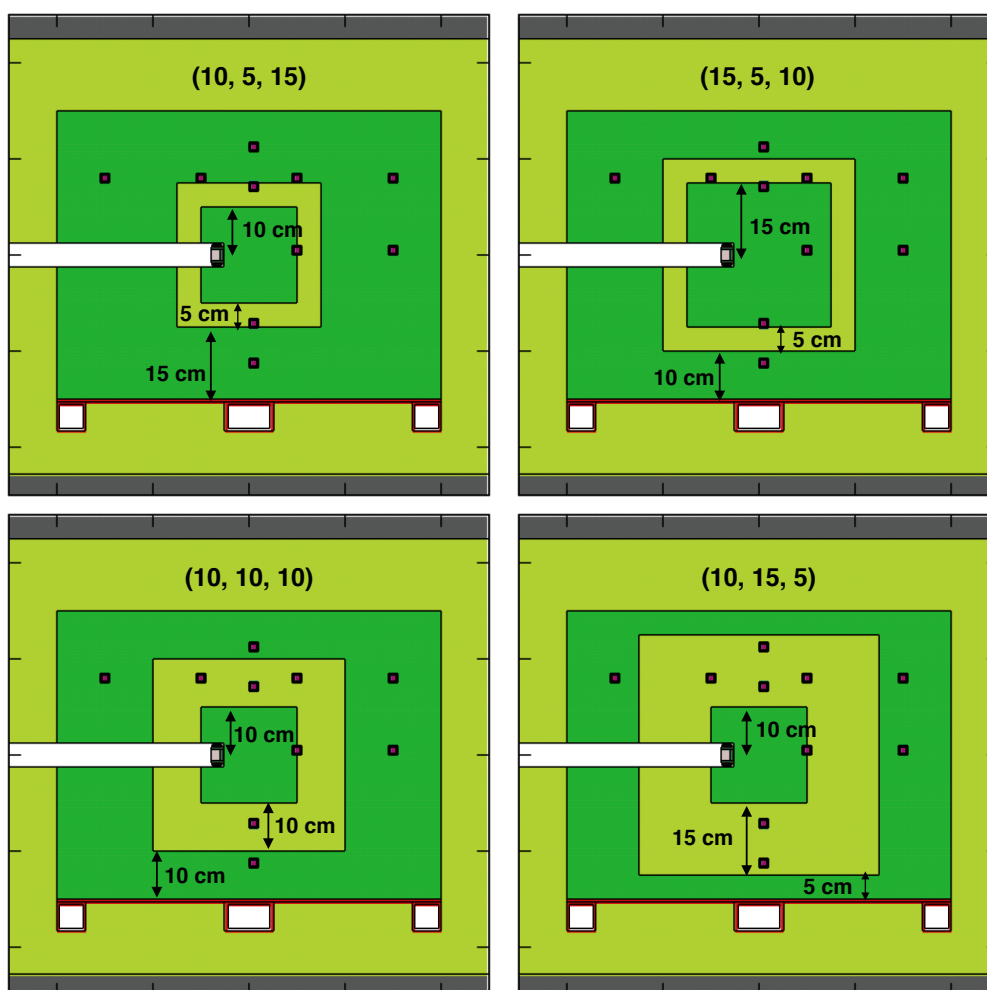


Figure 30 - Extra graphite layer in the diffuser: four possible configurations.

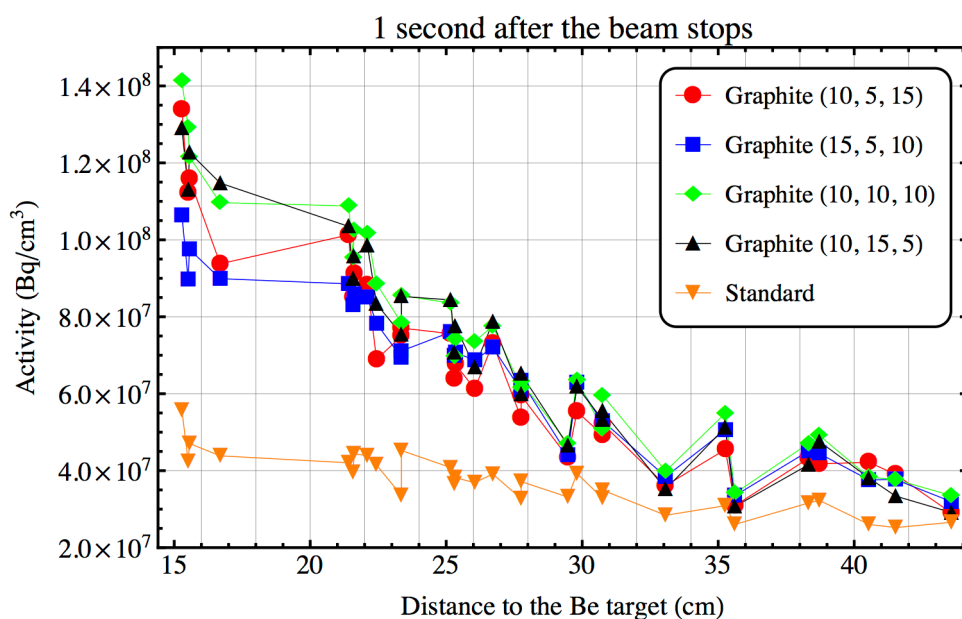
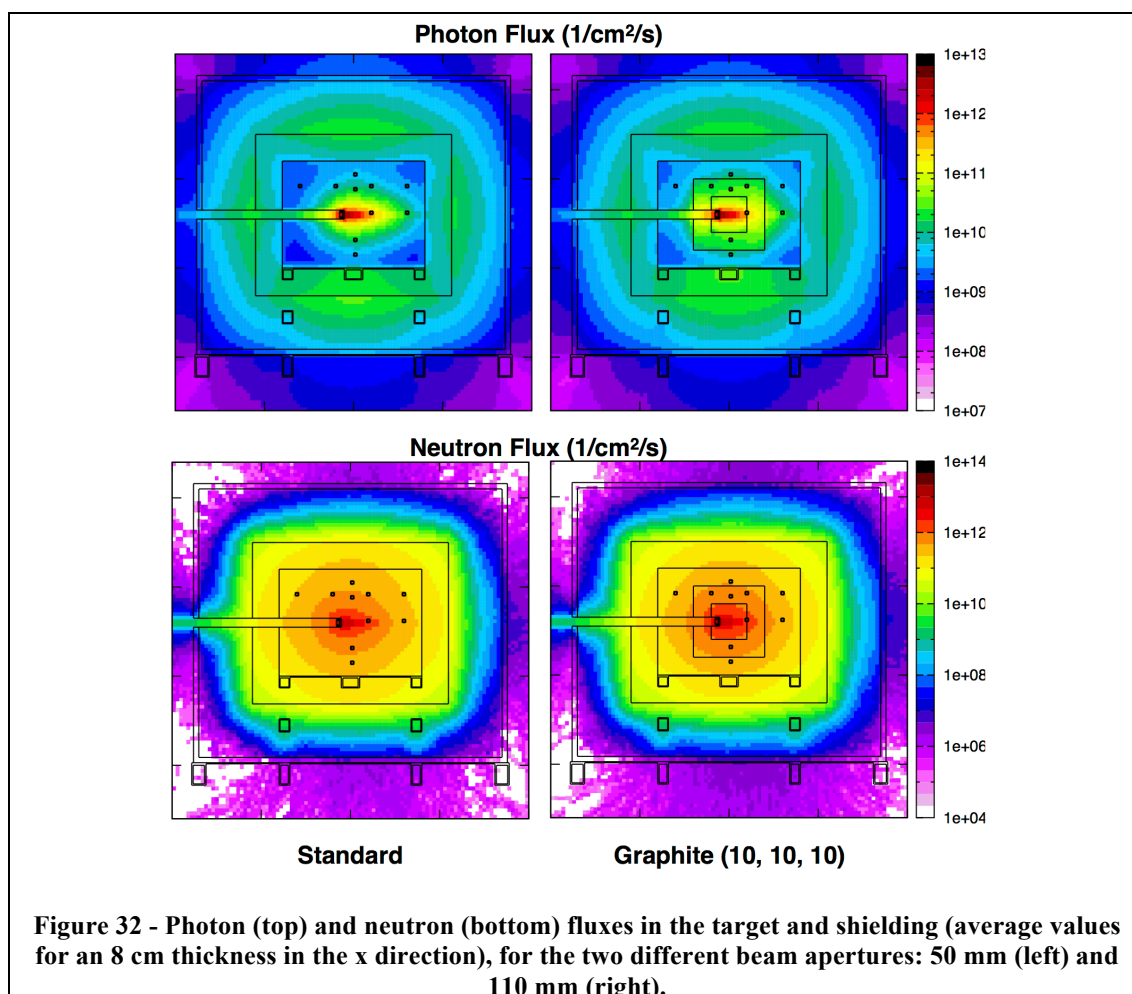
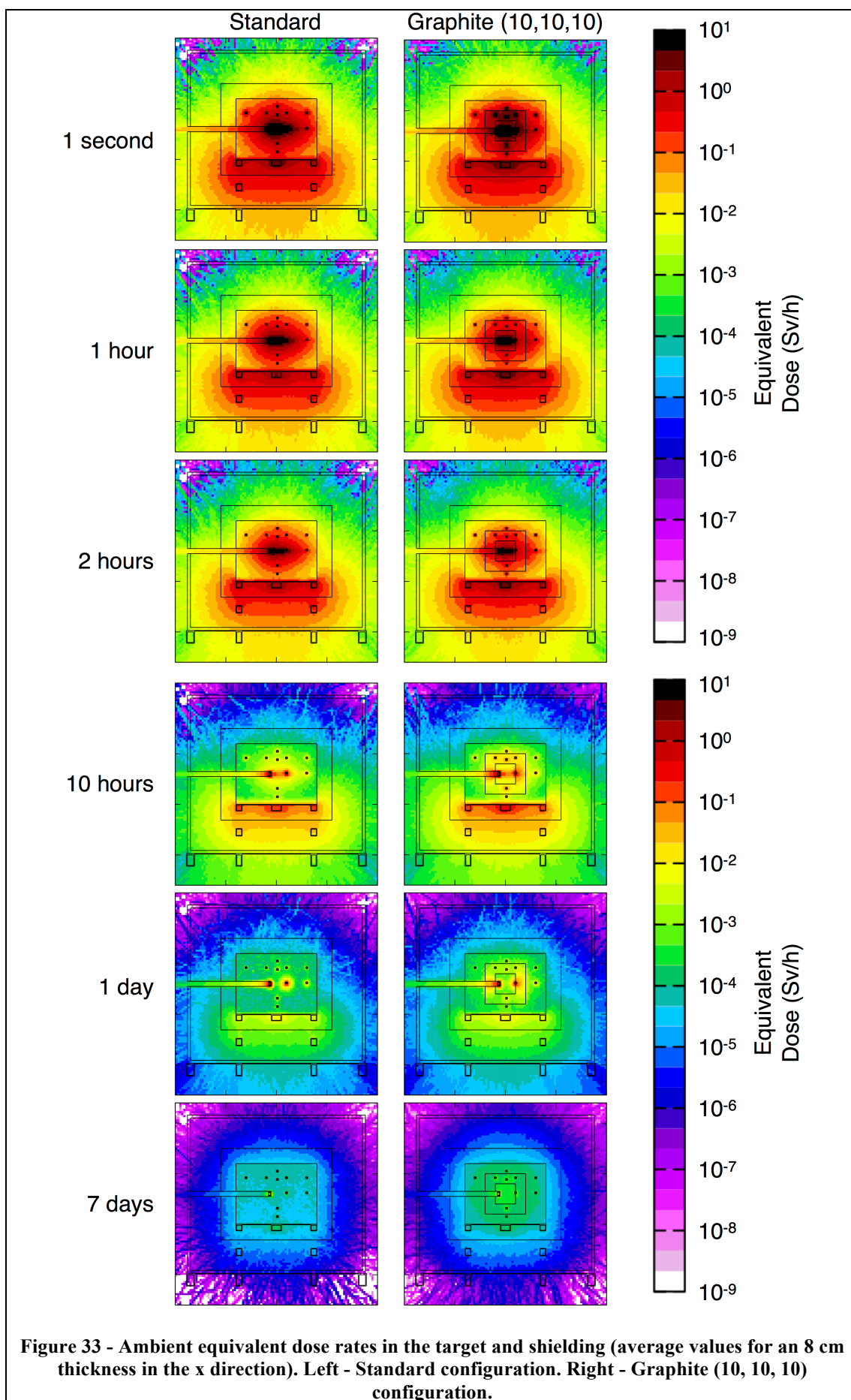


Figure 31 - Activities (Bq/cm^3) of ^{99}Mo in the targets with a slice of graphite inside the lead: comparison between different configurations.

It is also important to assess the effect of using the extra layer of graphite on the fluxes and residual dose rates outside the shielding. Figure 33 shows the photon and neutron fluxes in the standard configuration compared to the ones obtained with the extra layer of graphite. The 10 cm layer was chosen for the comparison, since it produces the highest activities of 99-Mo.

The neutron fluxes are increased outside the shielding when the additional layer of graphite is introduced. This is due to the removal of lead, which has a much higher density than graphite; although the neutrons lose less energy per collision, there are many more collisions in lead and the neutrons are more efficiently diffused (on average, they end up crossing an increased thickness of polyethylene, losing more energy and increasing the probability of capture in the outer layer of borated polyethylene). The photon fluxes outside the shielding seem slightly reduced, an unexpected result since lead is much more efficient to shield photons than graphite. And the photons coming from the target are indeed better shielded when there is no extra graphite layer. The explanation for the slight reduction of the photon fluxes outside the shielding seem to lie on the fact that with more lead there is more neutron diffusion, which, on average, increases the path length of the neutrons in polyethylene, increasing also the number of capture reactions and making polyethylene a brighter secondary source of photons.

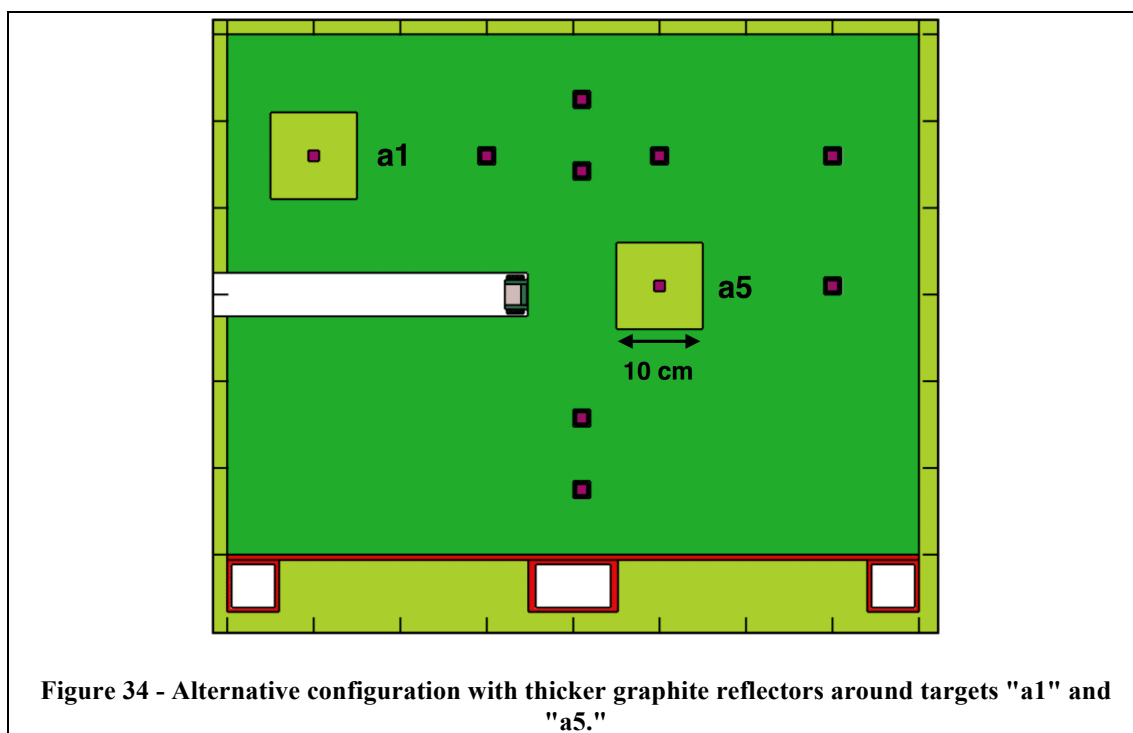




As for the residual dose rates, Figure 33 shows that they are increased in the new configuration. This is due to the increased activities of ^{99}Mo and also from an increased activation of graphite when compared to lead.

2.2.7 Extra Graphite Layer vs. Increased Graphite Thickness around ^{98}Mo

An additional configuration was tested, to check if the increase of the activities with the extra graphite layer could be replicated with graphite reflectors of the same thickness around the individual ^{98}Mo targets. This configuration is shown in Figure 34: two targets were chosen for this test, one located close to the target ("a5") and the other more distant ("a1"). In the previous section, target "a5" was located inside the layer of graphite, while target "a1" was located outside. Both targets are now placed inside 10-centimeter-sided cubes of graphite.



The comparison between the activities one second after the stoppage of the beam is presented in Figure 35, for the four configurations of the previous section and the new configuration. It is evident that the block of graphite around an individual target cannot replicate the results achieved with any of the four configurations of the previous section. As shown before, the advantage of using a single layer of graphite inside the lead is more pronounced when the ^{98}Mo target is located closer to the beryllium target. Although the new configuration produces better results than the standard configuration, a single layer of graphite inside the lead, in comparison, can increase the yields of ^{99}Mo by almost a factor of 2.

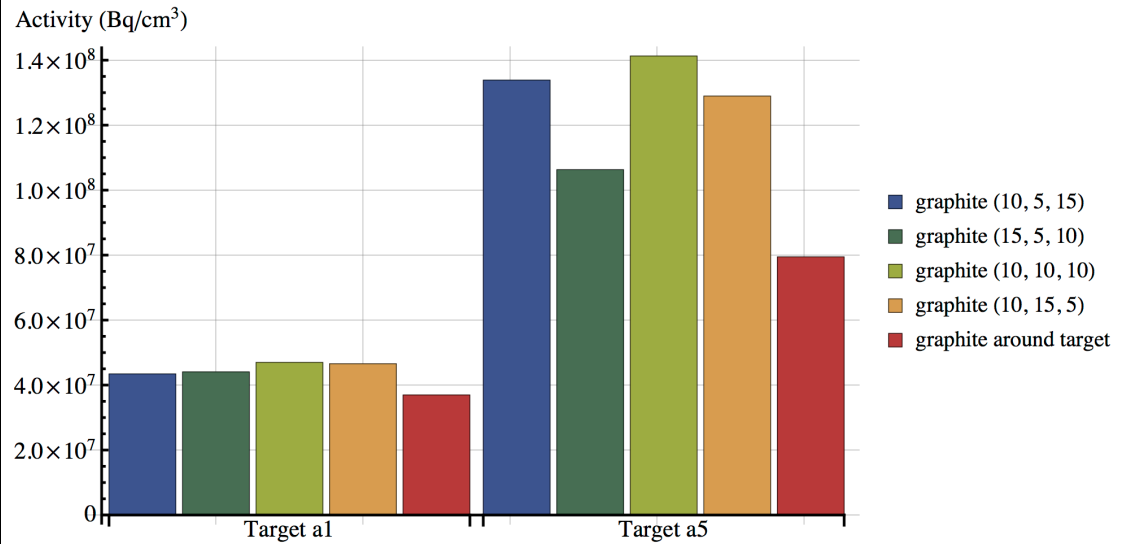


Figure 35 - Activities (Bq/cm³) of 99-Mo in targets "a1" and "a5" one second after the beam stoppage: comparison between configurations.

Annex 2

Radiation protection controls during activator runs

Radiation protection controls were undertaken for each of the 20 runs carried out from the 17th September 2014 to the 22th May 2015. This document reports the methods and main results.

1. Methods

Before and after a run, the dose rates coming from activation at several points across the proton beam line were measured. Measurements were performed with the proportional counter FH40 (table 1). After the run, due to the higher dose rates, measurements were often performed with the external teleprobe FHZ612 (2 GM-tubes) associated to the FH40 detector in order to increase the distance to the measurement point. Both devices provide the ambient equivalent dose ($H^*(10)$) and are calibrated in the energy of ^{137}Cs . On the beam line dose rates were measured at contact (figure 1). The reading provided by the monitor may vary approximately within a 10% depending on the orientation and position of the detector.



Figure 1: Positions for dose rate chart measurements.

Characteristics	FH40	FHZ612
Precision	+/- 20%	+/- 20%
Energy range	33 keV – 3 MeV	>60 keV
Dose rate range	0.01 $\mu\text{Sv/h}$ – 1 Sv/h	0.5 $\mu\text{Sv/h}$ – 10 Sv/h
Measured quantity	$H^*(10)$	$H^*(10)$

Table 1: Detector characteristics.

Ten points were chosen across the beam line (figure 2): four after the first bending magnet, located near the Faraday cups or right upstream the quadrupoles (A, B, C, D), two on the second bending magnet (E) and on the main line upstream the quadrupole (F), two upstream the activator (G and H) and two remaining points, one at about 2 m from the activator beam line (balise 4) and another in front of the activator, on the accessible side.

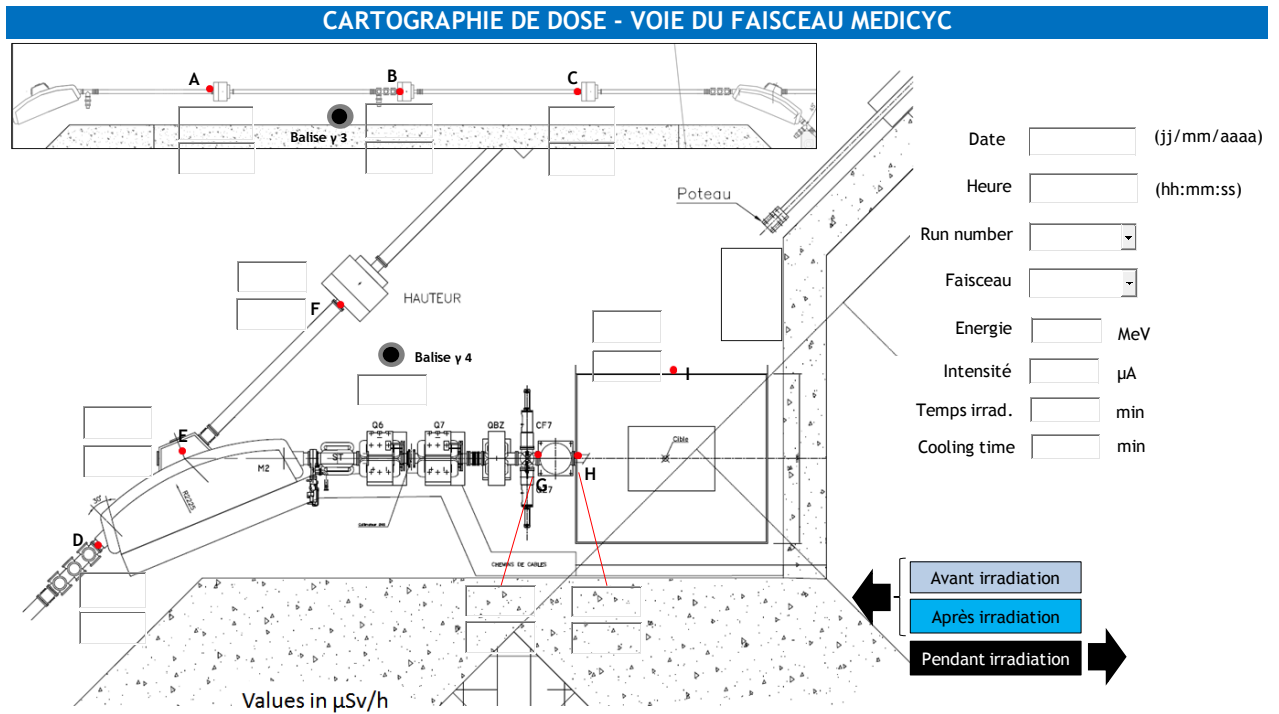


Figure 2: Positions for dose rate chart activation measurements.

From February 2015 dose rate values measured by the 5 fixed gamma detector located inside the cyclotron bunker were also retrieved during the run (figure 3). Among the 5 detectors detector number 4 (balise 4) is located closer to the activator and is thus the more representative of the secondary gamma dose rate generated during the run.

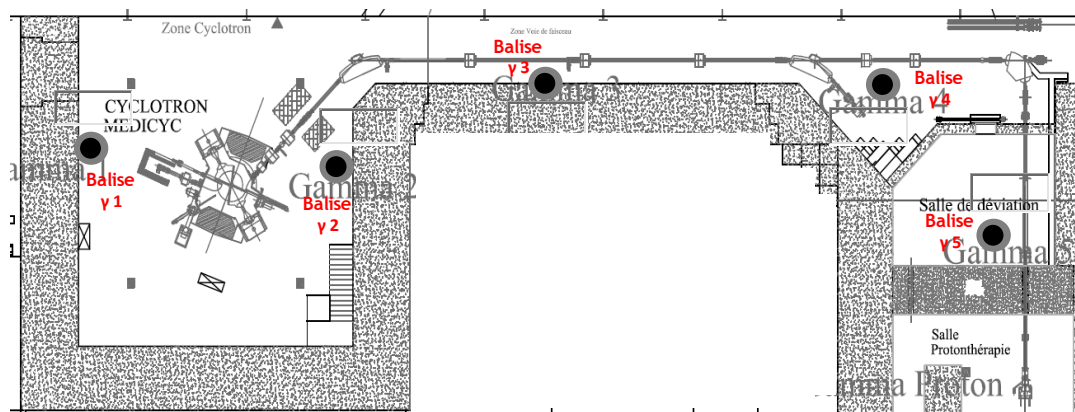


Figure 3: Positions of gamma dose fixed detectors in Medicyc bunker.

2. Results

Dose rates have been reported in individual reports after each run. Figures 4 to 6 summarize the global results for 4 μA proton runs (figure 4), 1 μA proton runs (figure 5) and deuteron runs (figure 6). Points A to I and “balise 4” show the activation measurements after each run and on the right, “balise 1” to “balise 5” show the gamma dose rates during the run.

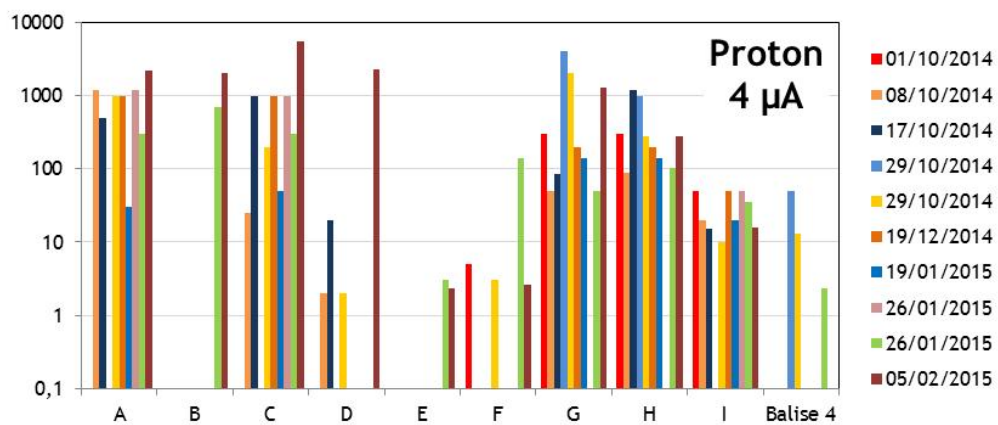


Figure 4: Positions of gamma dose fixed detectors in Medicyc bunker.

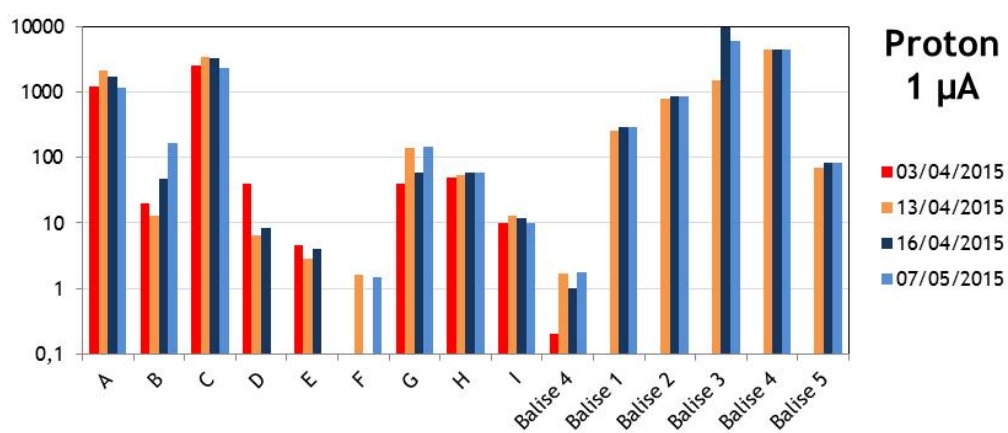


Figure 5: Positions of gamma dose fixed detectors in Medicyc bunker.

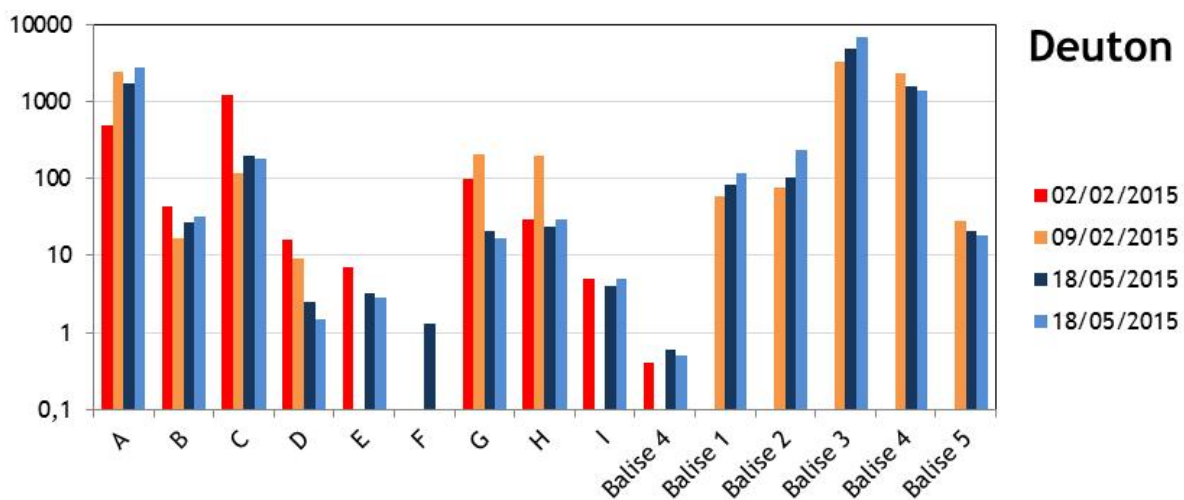


Figure 6: Positions of gamma dose fixed detectors in Medicyc bunker.

Figure 7 shows a comparison between deuteron and proton runs in terms of activation dose rates after the run (left plot) and gamma dose rates during the run (right plot).

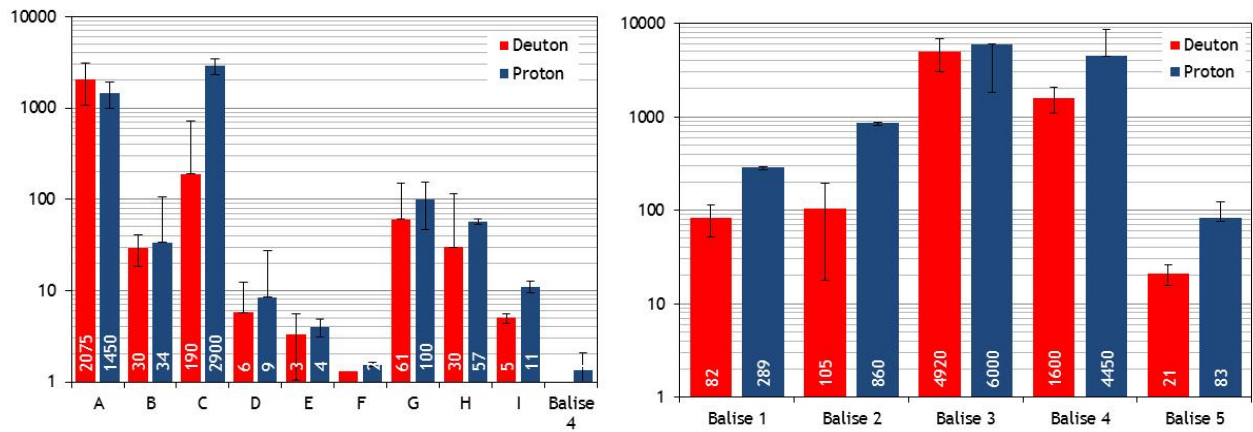


Figure 7: Activation dose rates.

Ratios between 1 μ A proton and deuteron runs for the activation dose rate after the run and the dose rate during the run are provided in table 2.

Position	Activation dose rate after run (proton run / deuteron run)	Detector	Dose rate during run (proton run / deuteron run)
A	0.7	Balise 1	3.5
B	1.1	Balise 2	8.2
C	15	Balise 3	1.2
D	1.5	Balise 4	2.8
E	1.2	Balise 5	4.0
F	1.2		
G	1.7		
H	1.9		
I	2.2		
Balise 4	2.7		

Table 2. Ratio of activation dose rates and dose rates during the run, for 1 μ A proton runs over 1 μ A deuteron runs.

The activation on the right and left sides of the beam pipe (figure 8) differed significantly in points A and C. Figure 9 shows a comparison of the values on both sides of the beam pipe for 5 different runs (values measured before the run).

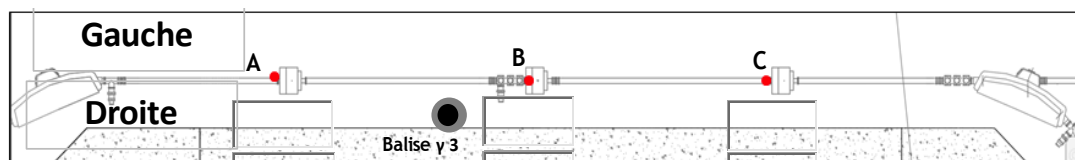


Figure 8: Identification of right (droite) and left (gauche) sides of the beam pipe.

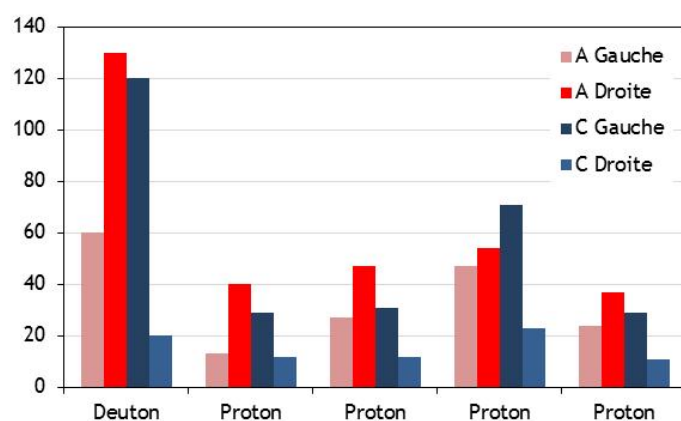


Figure 9: Activation dose rates before run for 5 different runs, showing the activation values on the right and left sides of the beam pipe.

Point A was always more activated on the right side whereas point C left side was more activated than the right side.

Annex 3

IBEL SA Survey of RadiolIsotope for Clinical and Research Use in Switzerland

IBEL SA Survey of RadioIsotope for Clinical and Research Use in Switzerland

IBEL SA
Geneva April, 2014



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for a better life

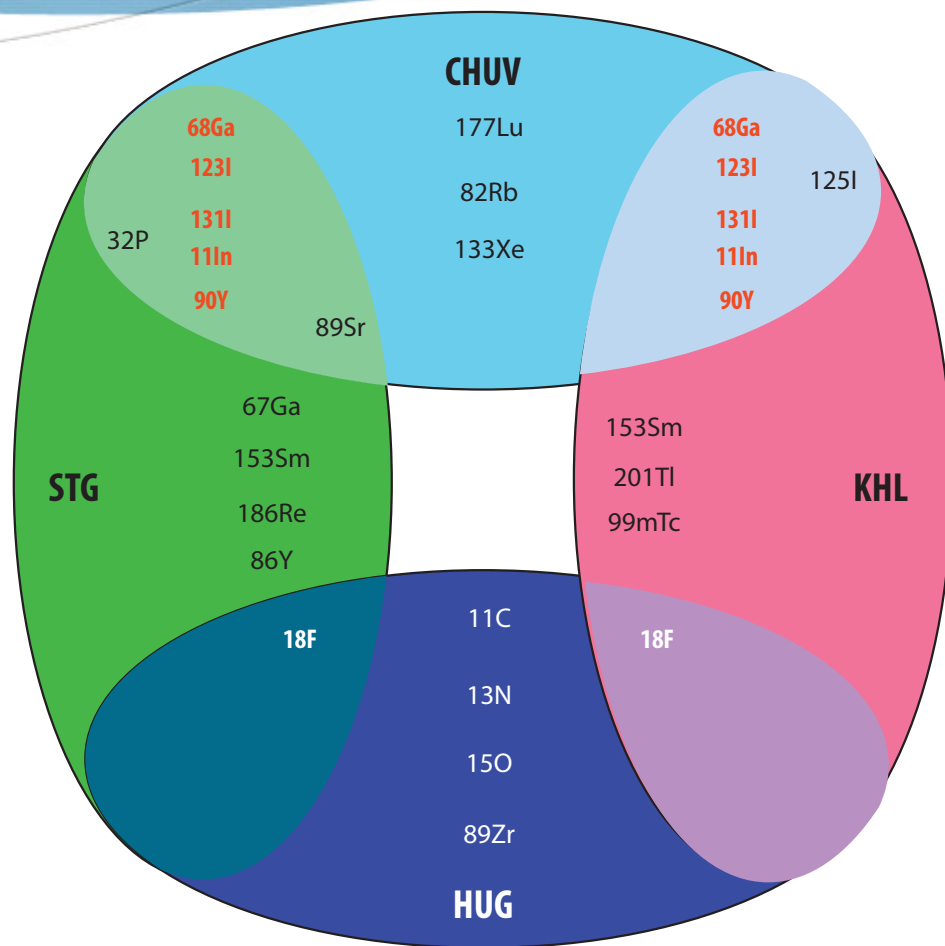


IBEL Online Survey

- ◆ The Survey was launched at the end of January.
- ◆ It was sent to 34 hospitals and research institutions in Switzerland
- ◆ Response rate: 12%
- ◆ Three hospitals HUG, CHUV, HKLuzern, and other institution from St. Gallen.
- ◆ It was used the free Monkey Survey application on the web.

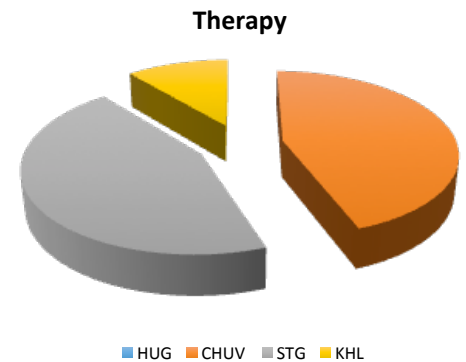
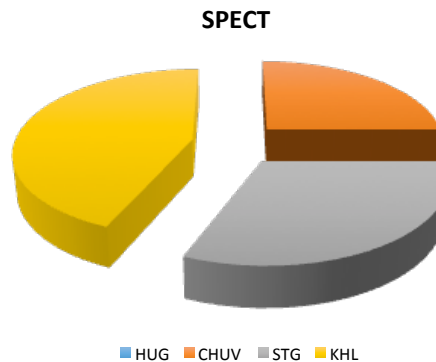
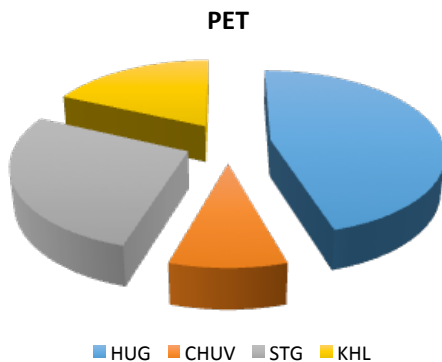
Question 1a: Which radionuclides do you and your co-investigators use in your current Clinical Use?

- CHUV & HUG use different radio-isotopes for their clinical activities.
- HUG is oriented to the use of PET isotopes,**
- CHUV is more oriented to SPECT** (123I, 125I, 131I, 111In) & **Therapy** (32P, I125, 89Sr & 90Y) and PET (68Ga),
- STG:
 - PET (18F, 68Ga, 86Y),
 - SPECT (123I, 131I, 111In, 67Ga, 153Sm)
 - Therapy (32P, 186Re, 89Sr, 90Y).
- KHL:
 - PET (18F, 68Ga),
 - SPECT (123I, 125I, 131I, 111In, 153Sm, 201Tl, 99mTc),
 - Therapy (90Y).
- CHUV & STG common radio-isotopes for:
 - PET (68Ga),
 - SPECT (123I, I125, 131I, 111In),
 - Therapy (32P, 89Sr, 90Y).
- CHUV & KHL common radio-isotopes for:
 - PET (68Ga),
 - SPECT (123I, I125, 131I, 111In),
 - Therapy (I125, 90Y).
- HUG & STG common radio-isotopes: for PET (18F)
- HUG & KHL common radio-isotopes: for PET (18F)



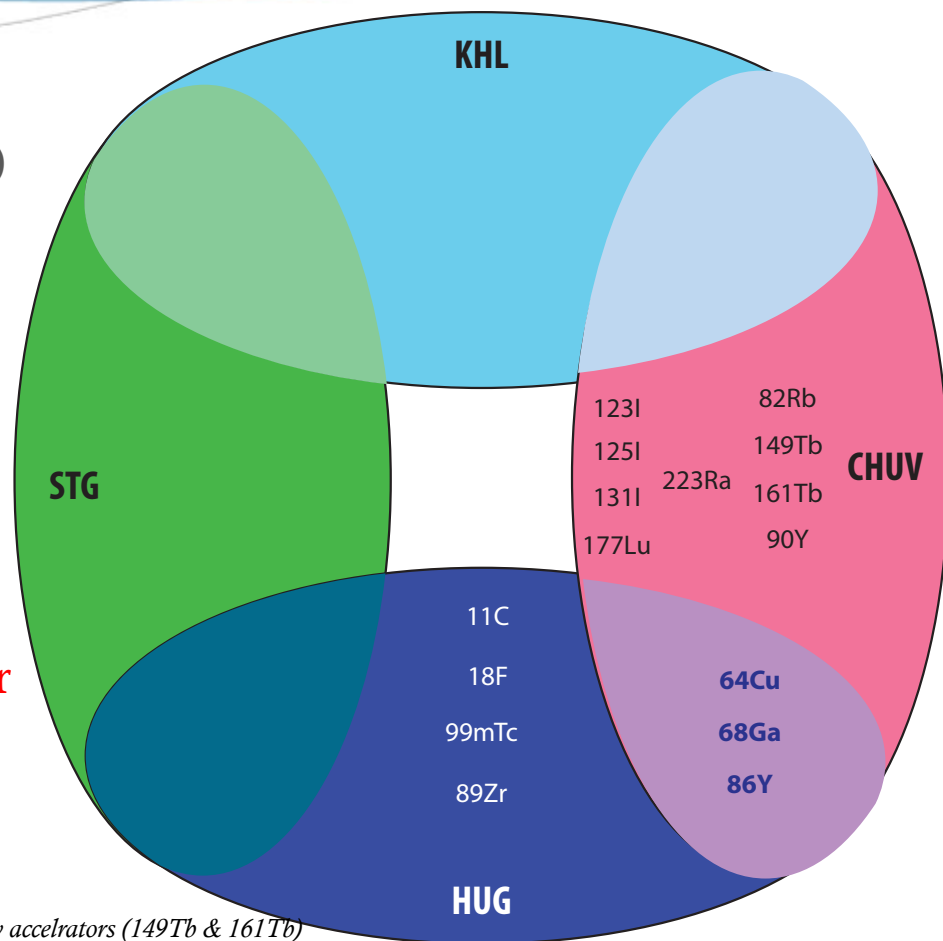
Question1a: Conclusion

HUG is more oriented to offer **PET Services** while the other institutions are using radioisotopes for SPECT and Therapy.



Question 1b: Which radionuclides do you and your co-investigators use in your current Research Activities?

- HUG research radioisotopes for
 - PET(11C, 18F, 89Zr, 64Cu, 68Ga, 86Y)
 - SPECT (99mTc).
- CHUV research radioisotopes for
 - PET (64Cu, 68Ga, 86Y, 82Rb),
 - SPECT (123I, 125I, 131I, **161Tb***)
 - Therapy (223Ra, 177Lu*, 149Tb*, 90Y)**
- CHUV& HUG common radio-isotopes: **for PET** (64Cu, 68Ga & 86Y)
- STG & KHL: No R&D



(*) Promising Radioisotopes, all of them are at present produced by reactors and high energy accelerators (149Tb & 161Tb)

Question 2: Which of these radionuclides are produced in adequate quantities on site, in your institution?

◆ Clinical Use:

◆ HUG:

- ◆ PET (11C, 18F, 13N, 15O)

◆ CHUV:

- ◆ PET (68Ga, 82Rb)

- ◆ SPECT (99mTc)

◆ KHL:

- ◆ SPECT (99mTc)

◆ STG:

- ◆ SPECT (99mTc)

◆ Research Activities:

◆ HUG:

- ◆ PET (11C, 64Cu, 18F, 86Y)

◆ CHUV:

- ◆ PET (68Ga, 82Rb),

- ◆ SPECT (99mTc)

◆ KHL:

- ◆ No

◆ STG:

- ◆ No

Made on site Radioisotopes:

PET (11C, 18F, 13N, 15O, 68Rb, 82Rb) → For Clinical Use

PET (11C, 64, 18F, 86Y, 68Ga, 82Rb) → For Research

99mTc: *It is very hard to believe this radioisotope is produced in these institutions, since 99mTc is produced in a reactor and commercialized in 99Mo/99mTc Generators*

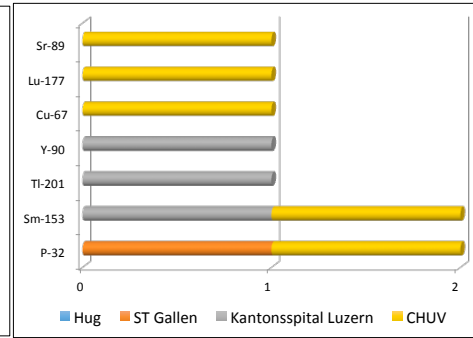
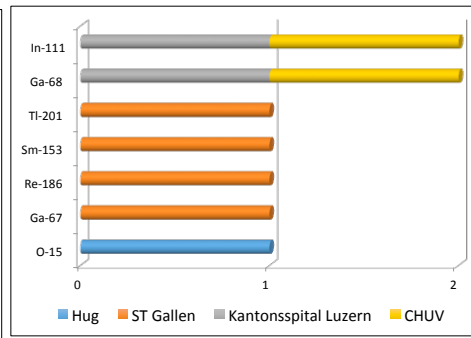
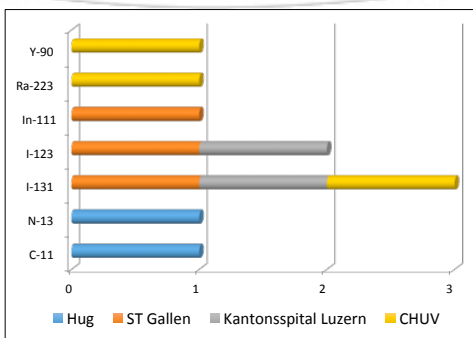
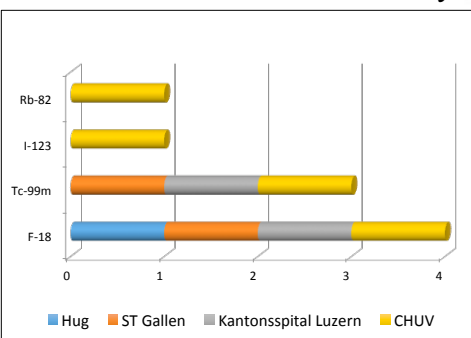
Question 3: For each isotope listed below, please indicate the frequency of use for Clinical Use

Daily

Weekly

Monthly

Yearly



All: PET(18F)

CHUV: PET (82Rb),
SPECT (123I, 99mTc)

HUG: PET (11C & 13N)

CHUV: SPECT (131I),
Therapy (223Ra, 90Y)

STG: SPECT (123I, 131I,
111In)

KHL: SPECT (123I, 131I,
111In)

HUG: PET (15O)

CHUV: PET (68Ga),
SPECT (111In)

STG: SPECT (67Ga,
153Sm), Therapy (186Re)

KHL: PET (68Ga),
SPECT(111In)

HUG:

CHUV: Therapy (89Sr,
177Lu, 67Cu, 153Sm,
32P)

STG: Therapy (32P)

KHL: SPECT (201Tl,
153Sm), Therapy (90Y)

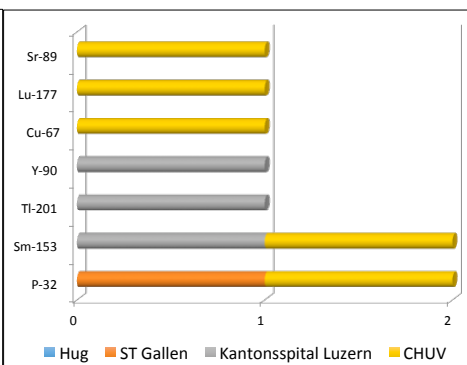
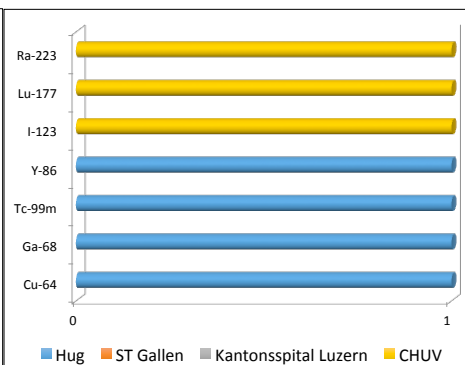
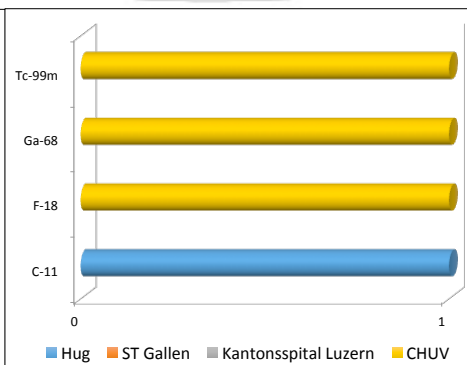
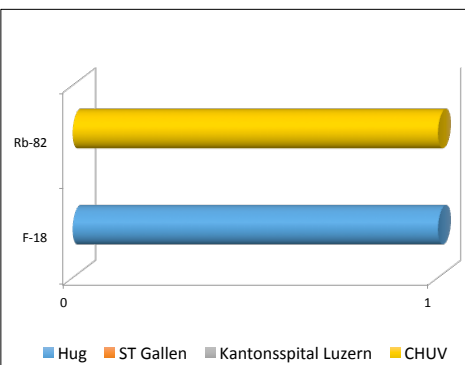
Question 4: For each isotope listed below, please indicate the frequency of use for Research Activities

Daily

Weekly

Monthly

Yearly



HUG:

PET (18F)

CHUV:

PET (82Rb)

HUB:

PET (11C)

CHUV:

PET (18F, 68Ga)

SPECT (99mTc)

HUG:

PET (64Cu, 68Ga, 86Y),

SPECT (99mTc)

CHUV:

PET (123I),

Therapy (177Lu, 223Ra)

CHUV:

Therapy (32P, 153Sm, 67Cu, 177Lu, 89Sr)

STG:

Therapy (153Sm)

KHL:

Therapy (153Sm, 201Tl, 90Y)

Question 5a: Please prioritize the radionuclides you use in decreasing order of Clinical Use

HUG:

- PET (18F, 11C, 13N, 15O)

CHUV:

- PET (18F)
- SPECT (99mTc)
- PET (82Rb)

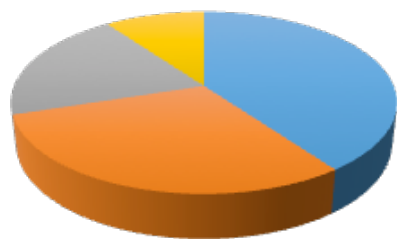
STG:

- SPECT (99mTc)
- PET (18F)
- SPECT (131I, 111In, 123I, 67Ga)
- Therapy (186Re, 90Y, 153Sm)
- SPECT (201Tl)

KHL:

- PET (18F)
- SPECT (99mTc)
- SPECT (131I, 111In, 123I)
- PET (68Ga)
- Therapy (90Y)

Hug



F-18 C-11 N-13 O-15

CHUV



F-18 Tc-99m Rb-82

ST Gallen



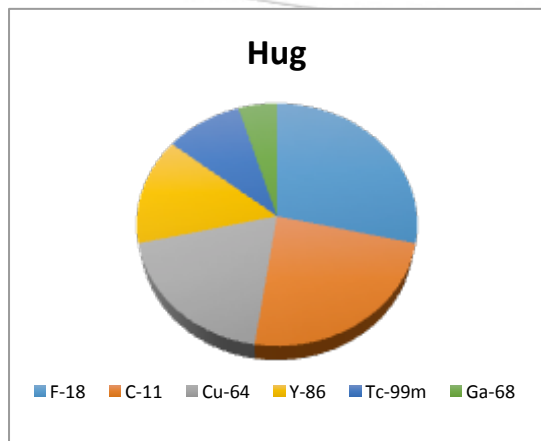
Tc-99m F-18 I-131 In-111 I-123 Ga-67 Re-186 Y-90 Sm-153 Tl-201

Kantonsspital Luzern



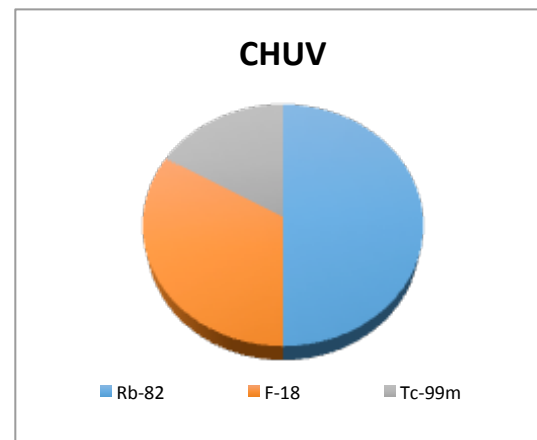
F-18 Tc-99m I-131 In-111 I-123 Ga-68 Y-90

Question 5b: Please prioritize the radionuclides you use in decreasing order of Research Activity

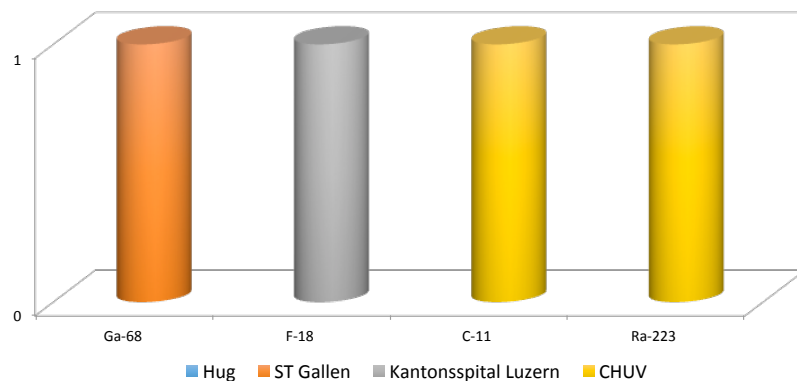


- ◆ HUG:
 - ◆ PET (18F, 11C, 64Cu, 86Y)
 - ◆ SPECT (99mTc)
 - ◆ PET (68Ga)

- ◆ CHUV:
 - ◆ PET (82Rb, 18F)
 - ◆ SPECT (99mTc)



Question 6: Which of your currently utilized radionuclides would you use more often, if it were more available?



For Research Activities

CHUV:

- PET (11C, 64Cu)
- Therapy (166Ho)

For Clinical Use

CHUV:

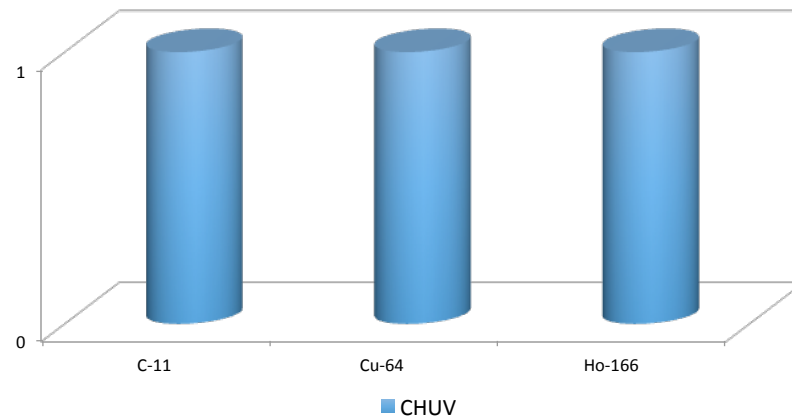
- PET (11C)
- Therapy (223Ra)

STG:

- PET (68Ga)

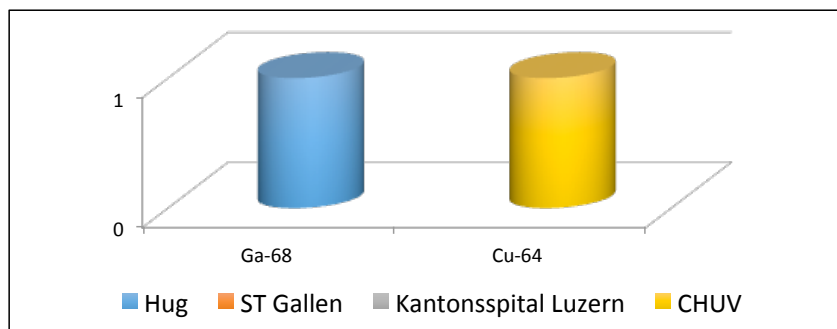
KHL:

- PET (18F)

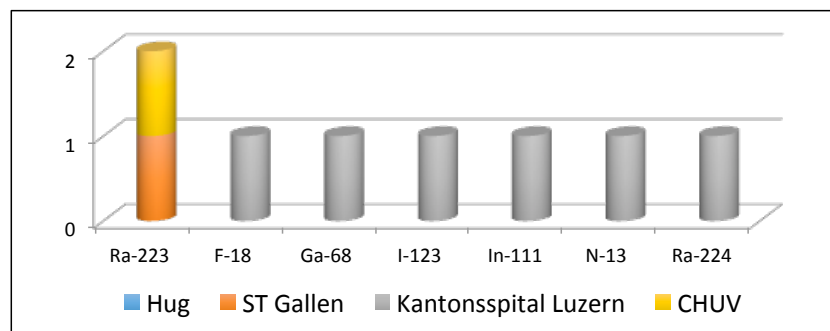
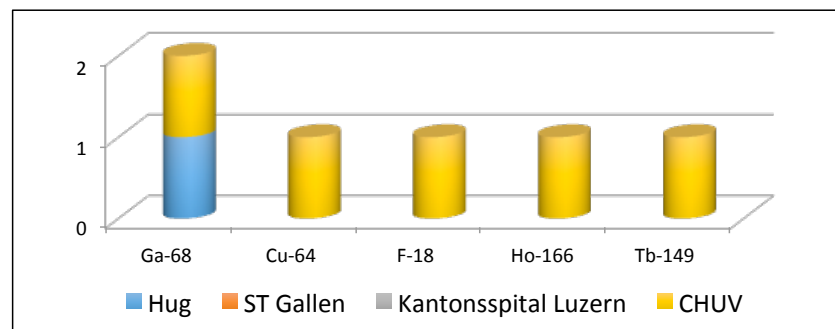


Question 7: What other radionuclides would you use on a routine basis, if they were available at a reasonable cost?

For current Research Activities



For upcoming Research Activities



For Clinical Use

Resumen 1

- ◆ In Switzerland it is used 31 different radioisotopes. PET (10), SPECT (10) & Therapy (11)
- ◆ These centers (CHUV & HUG) for their **research activities** are using the following radionuclides (in total 16): PET(11C, 18F, 89Zr, 64Cu, 68Ga, 68Y, 82Rb), SPECT (99mTc, 123I, 125I, 131I, 161Tb) & Therapy (223Ra, 177Lu, 148Tb),
- ◆ These centers (CHUV & HUG) for their **research activities** produce in situ 6 different radionuclides (positron emitters): PET (11C, 64Cu, 18F, 86Y, 68Ga, 82Rb)
- ◆ The most frequently used radionuclides **in Clinical activities** are: Daily use = {PET(18F, 82Rb), SPECT (123I, 99mTc)}, Weekly use = {PET (11C, 13N), SPECT (123I, 131I, 111In) & Therapy (223Ra, 90Y)}
- ◆ The most frequently used use radionuclides in **research** are: Daily use = {PET(18F, 82Rb)}, Weekly use = {PET (11C, 68Ga), SPECT (99mTc)}

Resumen 2

- ◆ For Clinical Use, the most important radioisotopes in Switzerland are the following: PET (18F, 11C, 13N, 15O, 82Rb, 68Ga), SPECT (99mTc, 131I, 111In, 123I, 67Ga), Therapy (186Re, 153Sm, 90Y, 201Tl)
- ◆ For Research the most important radioisotopes in Switzerland are the following: PET (18F, 11C, 64Cu, 86Y, 82Rb), SPECT (99mTc)
- ◆ Radioisotopes that would be used more often in **current research** if they were more readily available include: PET (64Cu, 11C), Therapy (166Ho)
- ◆ Radioisotopes desired most **frequently for current and upcoming projects**:
 - ◆ R&D: PET (18F, 68Ga, 64Cu), Therapy (166Ho, 149Tb)
 - ◆ Clinical: PET (18F, 68Ga, 13N), SPECT (123I, 111In) & Therapy (223Ra, 224Ra)

Conclusions

- Most of the Radioisotopes used in Switzerland can be produced in a accelerator center of 30-70 MeV (see slide Survey radioisotopes at different energies).
- The radioisotopes produced by Nuclear Reactors (^{67}Cu , ^{131}I , ^{133}Xe , ^{223}Ra , ^{177}Lu , ^{186}Re , ^{153}Sm , ^{90}Y , ^{32}P , ^{33}P , ^{166}Ho) can also be produced by such a center, as for example ^{166}Ho and others.
- Research radioisotopes and other radioisotopes of emerging importance, can also be produced.

Survey radionuclides at different energies

The short-lived PET isotopes are produced via the (p, n) process, the related p-energy is ~ 15 MeV.

Proton energy (MeV)	Radionuclide easily produced
0 – 10	^{18}F , ^{15}O , ^{86}Y , ^{89}Zr
11 – 16	^{11}C , ^{64}Cu , ^{18}F , ^{13}N , ^{15}O
17 – 30	^{123}I , ^{67}Ga , ^{111}In , ^{11}C , ^{18}F , ^{166}Ho , ^{125}I , ^{13}N , ^{15}O , ^{82}Sr (^{82}Rb), ^{186}Re , ^{201}Tl
30+	^{11}C , ^{64}Cu , ^{18}F , ^{67}Ga , ^{68}Ga (^{68}Ge), ^{123}I , ^{111}In , ^{13}N , ^{15}O , ^{82}Rb , ^{82}Sr (^{82}Rb), ^{89}Sr , ^{86}Y , ^{90}Y , ^{89}Zr
100+	^{149}Tb , ^{161}Tb (PSI, ISOLDE @ CERN)
Reactor Produced	^{67}Cu , ^{131}I , ^{133}Xe , ^{223}Ra , ^{177}Lu , ^{186}Re , ^{153}Sm , ^{90}Y , ^{32}P , ^{33}P , ^{166}Ho

The classical SPECT isotopes are produced via the (p, 2n) process, the related p-energy is ~ 25 MeV

PET Radioisotopes (from Survey)

1	C-11	20.3m	To study normal/abnormal brain functions
2	Cu-64	12.7h	PET scanning , planar imaging , SPECT imaging dosimetry studies , cerebral and myocardial blood flow , used with Cu-62 , treating of colorectal cancer.
3	F-18	110m	Brain studies, PET imaging
4	Ga-68	68.1m	Study thrombosis and atherosclerosis, PET imaging, detection of pancreatic cancer, attenuation correction.
5	I-124	4.17d	Radiotracer used to create images of human thyroid, PET imaging.
6	N-13	9.97m	PET imaging, myocardial perfusion.
7	O-15	122s	Water used for tomographic measuring of cerebral blood flow, PET imaging, SPECT imaging.
8	Rb-82	1.27m	Myocardial imaging agent, early detection of coronary artery disease, PET imaging, blood flow tracers. From ⁸² Sr.
9	Y-86	14.7h	Can be imaged by positron emission tomography and used as an isotopically matched surrogate radionuclide for (90)Y radiation doses estimations.
10	Zr-89	78.4h	The ⁸⁹ Zr radioisotope is used in the field of tumor diagnostics, tumor therapy and the investigation of the bio-kinetic (Uddin et al., 2005). Also, zirconium-89 has been suggested as a suitable positron emitter for labeling antibodies.

SPECT Radioisotopes (from Survey)

1	Ga-67	78.3h	Imaging of abdominal infections, detect Hodgkins/non-Hodgkins lymphoma, used with In-111 for soft tissue infections and osteomyelitis detection, evaluate sarcoidosis and other granulomatous diseases, particularly in lungs and mediastinum.
2	I-123	13.1h	Brain, thyroid, kidney, and myocardial imaging, cerebral blood flow (ideal for imaging), neurological disease (Alzheimer's).
3	I-125	59.9d	Osteoporosis detection, diagnostic imaging, tracer for drugs, monoclonal antibodies, brain cancer treatment (I-131 replacement), SPECT imaging, radiolabeling, tumor imaging, mapping of receptors in the brain, interstitial radiation therapy (brachytherapy) for treatment of prostate cancer.
4	I-131	8.04d	Lymphoid tissue tumor/hyperthyroidism treatment, antibody labeling, brain biochemistry in mental illness, kidney agent, thyroid problems, alternative to Tl-201 for radioimmunotherapy, imaging, cellular dosimetry, scintigraphy, treatment of Graves disease, treatment of goiters, SPECT imaging, treatment of prostate cancer, treatment of hepatocellular carcinoma, treatment of melanoma, locate osteomyelitis infections, radiolabeling, localize tumors for removal, treatment of spinal tumor, locate metastatic lesions, treat neuroblastoma, internal (systemic) radiation therapy, treatment of carcinoma of the thyroid.
5	In-111	2.81d	Detection of heart transplant rejection, imaging of abdominal infections, antibody labeling cellular immunology, used with Ga-67 for soft tissue infection detection and osteomyelitis detection, concentrates in liver, kidneys, high specific activity, white blood cell imaging, cellular dosimetry, myocardial scans, treatment of leukemia, imaging tumors. Brain studies, infection and colon transit studies.
6	Sm-153	2.00d	Cancer treatment/diagnostics, monoclonal antibodies, bone cancer pain relief, higher uptake in diseased bone than Re-186, treatment of leukemia.
7	Tb-161	6.9d	The radiolanthanide (161)Tb ($T_{1/2} = 6.90$ days, $E_{\beta(-)} = 154$ keV) was recently proposed as a potential alternative to (177)Lu ($T_{1/2} = 6.71$ days, $E_{\beta(-)} = 134$ keV) due to similar physical decay characteristics but additional conversion and Auger electrons that may enhance the therapeutic efficacy. The goal of this study was to compare (161)Tb and (177)Lu in vitro and in vivo using a tumour-targeted DOTA-folate conjugate (cm09).
8	Tc-99m	6.01h	Brain, heart, liver (gastroenterology), lungs, bones, thyroid, and kidney imaging, regional cerebral blood flow, equine nuclear imaging, antibodies, red blood cells, replacement for Tl-201.
9	Tl-201	73.1h	Clinical cardiology, heart imaging, less desirable nuclear characteristics than Tc-99m for planar and SPECT imaging, myocardial perfusion, cellular dosimetry.
10	Xe-133	5.25d	Lung imaging, regional cerebral blood flow, liver imaging (gas inhalation), SPECT imaging of brain, lung scanning, lesion detection.

Radioisotopes for Therapy (from Survey)

1	C-67	61.9h	Cancer treatment/diagnostics, monoclonal antibodies, radioimmunotherapy, planar imaging, SPECT or PET. Non-Hodgkins Lymphoma.
2	Ho-166	26.83h	seed source can deliver high radiation doses to the tumor, while the short range of the beta particles limits damage to the adjacent normal tissue. A beta emitter bioglass Ho-166 seed has been proposed for treating small HCC tumors (small hepatocellular carcinomas).
3	Lu-177	6.68d	Heart disease treatment (restenosis therapy), cancer therapy. Various therapies are being explored.
4	P-32	14.3d	Polycythaemia Rubra Vera (blood cell disease) and leukemia treatment, bone disease diagnosis/treatment, SPECT imaging of tumors , pancreatic cancer treatment , radiolabeling. 32P phosphate treatment of myeloproliferative diseases. For bone metastasis to control the pain.
5	P-33	25d	Labeling. Radioactive Phosphorus 33 (33P) radionucleotides are an alternative to 32P or 35S radionucleotides for DNA and RNA studies. Theoretical dosimetry calculations show that 33P offers a 3- to 6-fold therapeutic advantage over 32P, depending on the source and target regions assumed. The low-energy beta-particle emitter 33P appears to offer a substantial dosimetric advantage over energetic beta-particle emitters (e.g., 32p, 89Sr, 186Re) for irradiating bone and minimizing marrow toxicity.
6	Ra-223	11.4d	Alpha emitting. Monoclonal antibody attachment (alpha emitter) used for cancer treatment (RIT). HRPca Bone Metastases - Alpharadin
7	Ra-224	3.6d	Radium (224Ra) is commercially available again for the treatment of ankylosing spondylitis. The 224Ra is an alpha emitter with a physical half-life of 3.6 days and a radiation energy of 5.7 MeV. The effective half life is 1.7d in the human body. It behaves like a homologue of calcium with preferential accumulation in zones of calcification and ossification. The pharamclogocial effects in ankylosing spondylitis are antiosteoblastic and anti-inflammatory with subsequent relief of pain, improvement of mobility, and reduced spinal ossification.
8	Re-186	3.9d	Cancer treatment/diagnostics, monoclonal antibodies, bone cancer pain relief, treatment of rheumatoid arthritis, treatment of prostate cancer, treating bone pain.
9	Sr-89	50d	Bone cancer pain palliation (improves the quality of life), cellular dosimetry, treatment of prostate cancer, treatment of multiple myeloma, osteoblastic therapy, potential agent for treatment of bone metastases from prostate and breast cancer .
10	Tb-149	4.13h	Monoclonal antibody attachment used for cancer treatment (RIT). alpha-emitting, shows promise for radio-immumotherapy.
11	Y-90	64h	Internal radiation therapy of liver cancer , monoclonal antibodies , Hodgkins disease, and hepatoma , cellular dosimetry, treating rheumatoid arthritis, treating breast cancer, treatment of gastrointestinal adenocarcinomas .