

**EVALUATION OF RADIOISOTOPE
PRODUCTION CAPABILITIES IN
MYRRHA**

MASTER THESIS

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DIPARTIMENTO ENERGIA

Corso di Laurea Magistrale in Ingegneria Energetica e Nucleare

TESI DI LAUREA MAGISTRALE

Evaluation of radioisotope production capabilities in MYRRHA

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**Anno Accademico 2015–2016
Sessione I**

This Master Thesis is the result of researches carried out during an internship, of about nine months, conducted at SCK·CEN in Mol, Belgium.



Abstract

The purpose of this work is to demonstrate what are the radioisotope production capabilities of MYRRHA, thought as the heir of the BR2, one of the most powerful research reactors in Europe. At SCK·CEN where the MYRRHA project is under design, the production of ^{99}Mo by fission of ^{235}U and of ^{227}Ac and ^{228}Th by thermal neutron capture of ^{226}Ra has already been evaluated showing good results. Since MYRRHA is a fast spectrum reactor, moderation has to be provided to neutrons in order to make these reactions occur. The IPS are inserted in the periphery of the core, where it is easier to thermalize neutrons, and are provided of a system with circulating water for both moderating and cooling; Beryllium baskets are foreseen for both hosting the targets inside the IPS and moderating the incoming neutrons. The configurations for the productions just mentioned are used in this work for the production of several radioisotopes. It is investigated the possibility of: extracting ^{89}Sr , ^{90}Sr ($/^{90}\text{Y}$), ^{95}Zr , ^{131}I , ^{133}Xe and ^{140}Ba ($/^{140}\text{La}$) from the same UAl_x plate used for Mo production; producing by (n,γ) reactions ^{32}P , ^{79}Kr , ^{89}Sr , ^{90}Y , ^{140}La , ^{153}Sm , ^{177}Lu , ^{188}W ($/^{188}\text{Re}$), ^{192}Ir and ^{203}Hg , which are already produced in the BR2, and ^{131}I , from two targets of Tellurium in natural and oxide form, and $^{191}\text{Os}/^{191m}\text{Ir}$; performing (n,α) , (n,n') and (n,p) fast reactions in the inner IPS for the production of ^{24}Na , ^{32}P , ^{89}Sr , ^{90}Y , ^{117m}Sn and ^{153}Sm . In order to estimate the produced activities, the core is modelled in MCNP and the simulations are run in ALEPH2, a Monte-Carlo Burn-up Code developed by SCK·CEN itself. The obtained results, not all positive, show on the whole that the designed irradiation rigs work well for the production of these mentioned radionuclide too.

to my first and timeless teachers,
my parents.

Acknowledgements

I really would like to thank my mentor, Dr. Edouard Malambu Mbala, that during the months elapsed at SCK-CEN taught me so much about the issues related to Radioisotopes, following my steps along the path. My grateful thanks to all the researchers of the Nuclear System Physics Group in the name of its head, Dr. Gert Van den Eynde, and to Radu Popescu, that have helped me whenever I needed it.

I would like to thank my advisor, Prof. Piero Ravetto, not only for his availability, but especially for having passed on to us in classroom his love for what he does.

Special thanks to my friends and colleagues, Marco De Lorenzo, Massimo Papa, Michela Magnanimo, Giacomo Alessi, and to my friends Marianna Damiani and Francesca Raimondi for all the support they provided me in the realization of this work.

Dulcis in fundo, I thank my parents, to whom this work is dedicated, my brother, my sister and her family for being my *safe haven*, starting point and, often, destination.

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List of Acronyms

ADS Accelerator Driven Systems

BR# Belgian Reactor #

CR Control Rods

EOI End of Irradiation

FA Fuel Assemblies

GUINEVERE Generator of Uninterrupted Intense NEutrons at the lead
VENus REactor

HEU Highly Enriched Uranium

IPS In-Pile Section

IRF Irradiation at constant Flux

IT Isomeric Transition

JEFF Joint Evaluated Fission and Fusion File

LBE Lead-Bismuth Eutectic

MCNP Monte Carlo N-Particle transport code

MYRRHA Multi-purpose hYbrid Research Reactor for High-tech Appli-
cations

PET Positron Emission Tomography

List of Acronyms

SCK·CEN Studiecentrum voor Kernenergie · Centre d'Étude de l'Énergie
Nucléaire

SPECT Single Photon Emission Computed Tomography

SR Safety Rods

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Chapter 1

Introduction

1.1 Radioactivity genesis

Nuclear is one of the most controversial technologies nowadays known in the world: even if it is synonym of Progress, it does not find often the approval of the majority. However, contrarily to what happens for what concerns the Nuclear Power Plants, the great importance of the applications of radiation in the biomedical and industrial fields is recognized worldwide.

Radioactivity was officially discovered in 1896 by Henri Becquerel, but its name was coined only some years later by Marie Curie. Thanks to her studies conducted together with the husband Pierre, she was able first to announce the existence of Radium (1898) and then to isolate it in a pure form from an Uranium ore, in 1902. In the following year, the Curies were awarded the Nobel Prize in Physics for this discovery; they shared this prize with Becquerel for the extraordinary services he rendered by his discovery of spontaneous radioactivity. In the same year, Rutherford and Soddy postulated their *Theory of Radioactivity Disintegration*, according to which radioactivity is a phenomenon that occurs in the nucleus of the atom: emitting an α -particle (a Helium nucleus) or a β -particle (an electron), the nucleus decays spontaneously in another element, called daughter, which in turn could emit a γ -ray (a photon) if in an excited state. At the end of the 19th century, X-radiation was discovered too. The X-rays are photons as well as γ ; they are

distinguished from each other for their origins: the firsts are emitted by the electron shells and the others by the nucleus itself. Since the first experiments, the huge potentiality of radiation and, in particular, of the X-rays for imaging in medical field was largely understood so that in the first years of the 20th century Radiology was born. Another little curiosity about the work of Madame Curie: during World War I, after having studied quickly radiology, anatomy and automotive mechanism, she developed mobile radiography vehicles and units (known as *petites Curies*) to help the battlefield surgeons. [1]

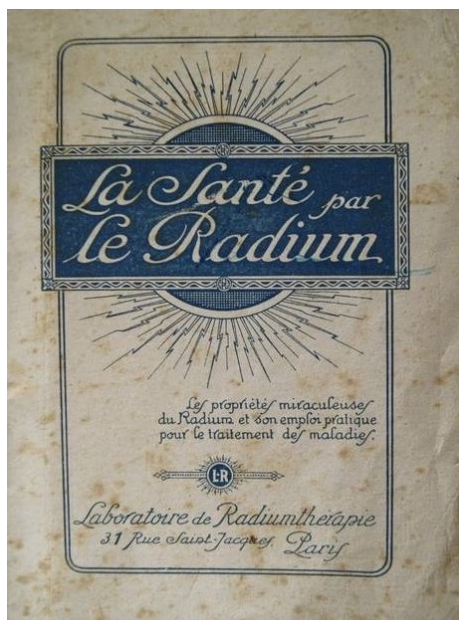


Figure 1.1: *Health from Radium*,
Laboratoire de Radiumthérapie,
Paris (1925)

At that time, the discovery of the Radium and the promises that this carried with itself generated a certain state of euphoria for its extraordinary properties: Figure 1.1 shows an information booklet of a Parisian laboratory in which all the healing properties of their own Radium-based products are listed. The scientists of that era started to investigate the therapeutic effects of the radiation on the human diseases: it was the dawn of the *Nuclear Medicine*.

An important step forward was taken when Curie's firstborn, Irène, and her husband Frederic Joliot discovered *artificial radioactivity* in 1934:

"In our study of the transmutations with emission of neutrons produced in the light elements irradiated with alpha rays, we have noticed some difficulties in interpretation in the emission of neutrons by fluorine, sodium, and aluminium. Aluminium can be transformed, by the capture of an alpha

particle and the emission of a proton, into a stable Silicon atom.¹ On the other hand, if a neutron is emitted the product of the reaction is not a known atom." [2]

That unknown atom was ^{30}P , an isotope of Phosphorus with one neutron less than the naturally occurring isotope; this lack makes it an unstable atom: it emits a positive electron (β^+) to become a stable isotope of Silicon, ^{30}Si . Since that time, the possibility to synthesize new radioelements subjecting the stable isotopes to radiation (α , p , n , etc.) was investigated. [3] This enabled to understand that the key of the stability of atoms is a good proportion of protons and neutrons inside the nucleus. All the isotopes created so far are collected in the so-called *Chart of the Nuclides*, in which all those characteristics related to their radioactivity (half-lives, major radiation and the energy associated to it, and so on) are listed too.

1.2 Artificial Radioactivity technology

Thanks to the easiness with which they are traced, radioactive isotopes (Radioisotopes) are really powerful tools for industrial and biomedical fields. When used with this purpose, the radioisotopes are referred as *Radiotracers*: this word has to suggest both the possibility to *trace* their dissemination and the fact that they are present in very small amount in the used solution, in *traces*. Anyway, radioisotopes have several uses in addition to tracing and the following sections are aimed to give a brief presentation of the most common (of course, this cannot be an exhaustive list of all the uses).

1.2.1 Industrial Uses

In the industrial field, Radioisotopes find a wide use, especially for the environmental management. The continuous development of the nuclear tech-

¹Rutherford was the first who demonstrated the effective possibility of transmutation of elements. In 1919, he was able to transmute Nitrogen into Oxygen bombarding it with α ($^{14}\text{N} + \alpha \rightarrow ^{17}\text{O} + p$).

niques results in increasing product quality and reliability. The uses for industrial purposes are various [4]:

- since even small amounts of radioisotopes can be detected easily, it is possible to *trace* the progress of some of them through complex path, or in chemical and other processes to understand the behaviour of the elements or species. These studies are based on the fact that the chemical behaviour of radioactive isotopes is identical with that of their stable isotopes in any chemical process. In this kind of investigations, the half-life of the radiotracer is chosen to be just long enough to obtain the required information without having remaining radioactivity after the process. Typical examples of tracing are:
 - the measurements of the flow rate of solutions and suspensions with ^{24}Na and ^{82}Br or of gases using ^{41}Ar , ^{85}Kr , ^{133}Xe and rare gases.
 - the studies of the mixing processes. When in mixing solid particles are involved, the best tracer is a fraction of the same material made radioactive by neutron activation.
 - troubleshooting, which provides for localization of a leak monitoring the radiotracer leaking out of the system after being applied to the suspected part of the system itself. Used radioisotopes are ^{41}Ar and ^{82}Br .
 - the monitoring of the erosion of the refractory bricks in blast furnace at which sources of ^{60}Co are embedded during lining. The constant detection of the γ rays emitted by the Cobalt sources allows to determine the level of erosion in relation with their extinction.
 - testing of corrosion resistance of materials. The material to be tested is bombarded, for example, with Krypton in order to incorporate radioactive atoms (^{85}Kr in this case) in its matrix; in this way, a Kryptonate of the material is created. This sample is then exposed to corrosive atmosphere and the residual activity of the

radiokrypton is measured. Sometimes, surfaces are labelled with a radiotracer by isotope exchange, electro- or vacuum-deposition techniques.

- measuring the material loss at the surface by monitoring an activated thin-layer created irradiating the surface with charged particles. The estimation of the wearing of this superficial layer is made converting the measured activity in "wear depth" by a calibration curve obtained on a standard sample. With this technique is possible to study all types of iron, steel, non-ferrous metals and alloys. The usual involved reactions are: $^{56}\text{Fe}(p,n)^{56}\text{Co}$, $^{58}\text{Ni}(p,pn)^{57}\text{Ni} \rightarrow ^{57}\text{Co}$, $^{65}\text{Cu}(p,n)^{65}\text{Zn}$ and $^{95}\text{Mo}(p,n)^{95m}\text{Tc}$.

- radioisotopes emitting both neutrons and photons are used for ***Analysis***.

Neutron emitters are used in case we want to study a substance or a material to know which elements compose it: in this way, we can characterize an ore or, for instance, measure the soil water. In order to reach the goal, the sample is irradiated with neutrons, which, interacting with the atoms of the sample, cause the emission of γ rays; these are, then, analysed for characteristic energy and intensity to identify the types and quantities of the element composing the sample itself. This kind of Analysis is based on two main mechanisms: *thermal neutron capture* (TNC), (n,γ) reaction, which occurs immediately after a low-energy neutron is absorbed and *neutron inelastic scattering* (NIS), (n,n') reaction, which occurs as well instantaneously after a fast neutron collides with a nucleus. The kind of used analysers are composed by the neutron source and the detector: the most diffused use ^{252}Cf (neutrons are produced by the spontaneous fission) as source together with a NaI(Tl) Scintillator, which is mainly sensible to TNC reactions; there are then ^{241}Am -Be or ^{227}Ac -Be probes (neutrons are produced by the interactions of the α emitted by the Am or Ac with the nucleus of ^9Be) together with a scintillator, either NaI(Tl) as before or Bismuth Germanate, which can register both TNC and NIS.

Gamma emitters are used, together with a detector, to determine, for instance, the ash content of coal on line on a conveyor belt: this is possible because the interactions involving gamma rays are atomic number dependant and the ash has a higher atomic number than the coal combustible matter; the detector can measure the energy spectrum of gamma rays which have been inelastically scattered from the coal (Compton Profile Analysis) to indicate the ash content.

X-rays due to the source can induce fluorescent x-rays from other non-radioactive materials, which, then, can be measured by a detector: their energy allows to identify the elements present in the material and moreover, thanks to their intensity, it is possible to quantify each element. This technique is used, for instance, to determine the element concentrations in process streams of mineral concentrators.

- small pellets of radioactive material in a sealed titanium capsule are used for ***Gamma Radiography***. The operation is similar to that of X-ray radiography: the object to be screened is placed between the source and a photographic film. The gamma rays, produced by the source, pass through the object and, reaching the film, create an image on it. This inspection technique is non-intrusive, that is, it is possible to inspect critical components for internal defects without damaging them; flaws in metal castings or welded joints are shown. Unlike the X-ray radiography, gamma radiography has no need of electric power but, precisely because the source cannot be turned off, an adequate shielding is always required when it is not in use as well as when it is. Moreover, the γ sources are more transportable than the X-ray equipment, presenting a clear advantage on these when applications in remote areas are considered, and emit a few discrete wavelengths at most in contrast with the broad band of radiations due to the other type of source. Typical sources are ^{169}Yb and ^{192}Ir : the former is used to analyse steels up to 15 mm thick and light alloys to 45 mm; the latter on steel from 12 to 60 mm thick and light alloys to 190 mm.
- radiation is also used to gauge materials. The ***gauging procedure*** is

based on the attenuation of the radiation intensity in going through matter and consists in deducing the amount of a certain material from measuring the difference in intensity between the radioactive source and a detector. The advantage of this technique is that the contact with the material is not needed. Sometimes, it can happen that radiation is scattered back towards the source: detecting the amount of backscattered radiation can give more information about the characteristics of the same material. This technique is used, for instance, in paper manufacturing in which β gauges are used to monitor the thickness of the sheets of paper at speeds of up to 400 m/s; but γ gauges are also used to monitor and control the flow of materials in pipes, distillation columns, etc. Typical examples of radioisotopes used for gauging are ^{90}Sr , ^{204}Tl and ^{241}Am (used in particular in backscatter gauges).

- γ rays are used to **sterilize** medical products, wool, wood, archival documents, food (for its preservation), etc. They can kill bacteria without damaging packaging. For this purpose: ^{60}Co is the most used because its gammas are very energetic; sometimes, it is produced as a by-product of power generation in nuclear reactors. The goal of sterilizing blood for transfusions and for other medical applications is reached with the use of ^{137}Cs , which emits gammas about half as energetic as cobalt's.
- atoms of ^{238}Pu are used in the so called **Radioisotope Thermoelectric Generators**, which are at the basis of the power supply of spacecraft. These generators do not have moving parts, resulting in this way highly reliable; they provide electricity using thermocouples able to convert the heat due to the α emitted by Pu into electricity. The choice of the radioisotope to be used falls on ^{238}Pu since it has a half-life long enough to have a relatively constant energy release rate for reasonable amount of time and it can produce large amount of power per mass and volume. [5] For other purposes, ^{90}Sr and ^{240}Cm are used as well as generator fuel.

1.2.2 Medical Uses

Depending on the purposes, radionuclides can be used in unsealed or sealed form: in the first case, sources are usually liquids directly injected; in the other, radioactive substances are contained in parts of equipment or apparatus, or encapsulated in unbreakable or impervious objects such as "seeds" or needles. To have a better idea of the function of these elements in the different medical applications, a deeper look on these is presented. [6]

- **Diagnosis**

Radionuclides are used in unsealed form both for "in vitro" and "in vivo" applications. In the first case, human biological samples are studied outside of the human body; usually, this application involves kBq activities of aqueous-based radiopharmaceutical (^{125}I , ^{57}Co , ^{58}Co and ^{14}C , ...) used to measure the levels of drugs or hormones in biomedical samples. In vivo investigations of body are based on imaging. In nuclear medicine field, imaging is referred to as Scintigraphy. What differentiates Scintigraphy from the usual Radiography is the completely different approach of these two techniques to the imaging: in both cases photons are used since they are very penetrating and can pass through the body, creating a little number of ionizations per unit of length; but, while in the latter case imaging is based on the transmission of the X-rays emitted by a machine through the body, in the former the radiations are emitted directly by the patient after that a radiopharmaceutical is administered to him. Thus, the patient becomes himself a source of radiation, which is detected by specific gamma cameras.

The usual gamma cameras, a sketch of which is shown in Figure 1.3, consist of a collimator, made generally of Lead, of a crystal in which the photon frees the electrons (called for this reason photoelectrons), of photomultiplier tubes to amplify the signal deriving from that photoelectrons and then the electronic devices which have the scope of transforming the signals in images. These cameras are designed in the energy range for γ of 100-250 keV. For both energy lower of the minimum value of 100 keV and upper of 250 keV, the images may not be of

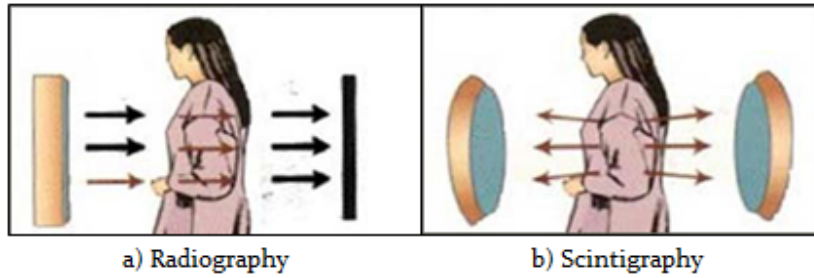


Figure 1.2: Comparison among: Radiography, based on the transmission of the X-ray beam through the body, and Scintigraphy, based on the emission of radiations from the body. (Figure taken from [7])

a sufficient quality because, in the first case, the γ produce too much scatter and in the second case it is difficult to collimate them.

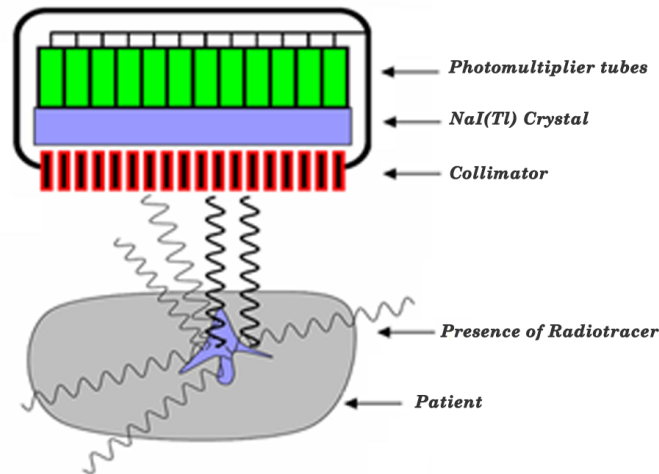


Figure 1.3: Gamma Camera scheme. (Figure from [8])

We have three different ways of scanning: the Scintigraphy diagnosis, **SPECT** and **PET**. The first one differs from the others because gives only bidimensional images. With the others techniques, it is possible to have 3D imagines. The investigation ways of **SPECT** and **PET** are different: even if, as said, in both cases the image is based on the detection of γ rays, the involved sources are not the same: in the first case gamma emitters are used and each "single emission" is detected; in the second, the multi-headed gamma cameras are configured to detect

only "coincidences", that is, the two different photons created by the same annihilation event of the positron emitted by the source in the body and an electron. γ emitters used for [SPECT](#) can be either isotopes that are in a metastable state (e.g., ^{99m}Tc), which emits γ decaying in the ground state (Isomeric Transition ([IT](#))), or radioisotopes (e.g., ^{131}I) that, after having emitted a β^- , decay in an excited state of the daughter nuclide (that is not metastable), which, in turn, reaches the ground state emitting γ ; in this last case, even if the γ is emitted by the daughter nuclide, it is attributed to the parent nuclide that, in this way, is recognized as a γ emitter. Instead, radioisotopes, which undergo β^+ decay (e.g., ^{18}F), are used for [PET](#). A little description of the operation of the two kinds of scanners is proposed below.

Regarding [SPECT](#), the cameras are rotated around the patient to detect the emitted photons. [SPECT](#) imaging is performed by using a gamma camera to acquire the data, from multiple angles, that then, thanks to the powerful computers to which they are linked, are converted in multiple 2-D images (also called projections). Applying a tomographic reconstruction algorithm to the multiple projections, the computers can yield a 3-D dataset. This exam is used to analyse the functions of internal organs, to get information about blood flow to tissues, to look at the chemical reactions in the body and, finally, to reveal where a tumour is located. To achieve this last aim, antibodies are linked to a radioactive substance: if a tumour is present, the antibodies will stick to it; [SPECT](#) scan is done, so, to detect where the radioactive substance is, revealing in this way the location of the tumour.

The mechanism on which [PET](#) scan is based is that of the annihilation of the positron, emitted by the radioisotopes within the patient, and an electron: this phenomenon creates two γ rays, emitted in opposite directions from each other, with an energy of 511 keV whatever the used isotopes. To better understand this concept, consider this procedure step-by-step. The Radiotracer is injected into the living subject

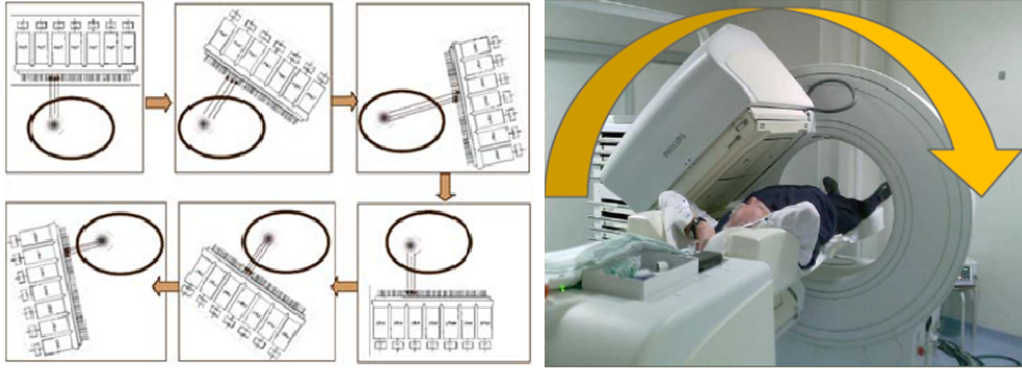


Figure 1.4: SPECT Scanner. (Figure taken from [7])

(usually into blood circulation). The tracer is chemically incorporated into a biologically active molecule. There is a waiting period while the active molecule becomes concentrated in tissues of interest; then the subject is placed in the imaging scanner. The emitted positron travels in tissue for a short distance (dependent on the isotope, but typically less than 1 mm), during which time it loses kinetic energy, until it decelerates to a point where it can annihilate with an electron. The most significant fraction of electron-positron annihilations results in two 511 keV gamma photons being emitted at almost 180 degrees to each other; hence, it is possible to localize their source along a straight line of coincidence (also called the line of response, or LOR). Unfortunately, due to its high energy, the positron may travel a distance of several millimetres from its injection point before encountering an electron: in the little table shown in Fig. 1.5, the average range for ^{11}C , ^{13}N , ^{15}O and ^{18}F is given.

It is important to take in consideration that the final image, corresponding to the sum of the points of impact, will give the statistical distribution of these annihilation points and not the distribution of the points of origin of the positron emission. In Figure 1.6, an example of PET scanner is presented. This kind of imaging gives the possibility to: reveal the size, shape, position, and some function of organs; diagnose

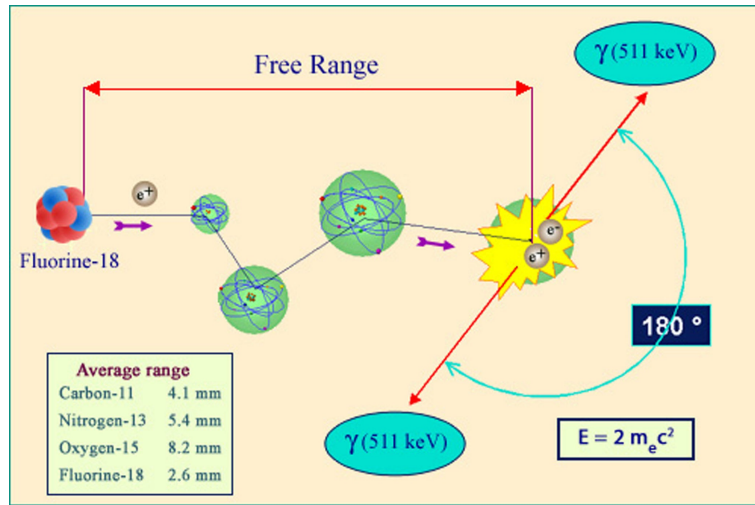


Figure 1.5: Annihilation of the positron emitted by ^{18}F . In the table, the average distance that it travels is listed for different isotopes. (Figure from [9])

cancer, heart problems, and brain disorders; see how far cancer has spread. Typical examples of used radionuclides in unsealed form are: ^{99m}Tc , ^{131}I , ^{133}Xe for **SPECT**; ^{15}O , ^{18}F , ^{79}Kr for **PET**. Sealed sources are used for bone densitometry, anatomical marking and calibration and reference standards. Typical sources are several GBq of ^{125}I , ^{153}Gd and ^{241}Am .

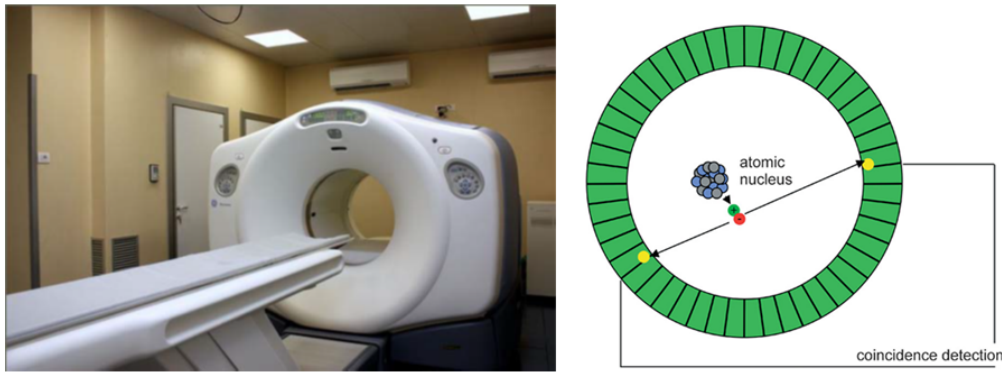


Figure 1.6: PET Scanner. (Figure taken from [7])

- **Treatment**

Radiations are used to destroy the DNA of carcinogenic cells in order to avoid their reproduction. The use of radiotherapy is justified by the Bergonié-Tribondeau law, which guarantees that the radiosensitivity of the cells of the human body is proportional to their mitotic index, that represents the velocity with which cells reproduce. As well known, cancerous cells reproduce faster than healthy ones having a higher mitotic index; this means that they are more radiosensitive, that is, the effects of the radiations on them are higher compared to those on the others. Two different ways of therapy exist: Teletherapy, based on the use of an external source of γ rays, and Brachytherapy, which uses an internal source of α and β . The last ones are little penetrating but, especially α , have a high LET and, so, are very effective against a cancer. In dosimetry, LET (*Linear Energy Transfer*) is defined as the energy transferred by an ionizing radiation to the matter along its path; it is a measure of the ionization density too. Thus, when the radiopharmaceutical reaches the cancer site, after being injected in the patient's body, the emitted radiation deposits its energy just in the area of interest. Typical examples of unsealed radioactive source are ^{131}I (thyroid cancer), ^{32}P or ^{90}Sr (bone cancer). Several radionuclides are used in sealed form during manual brachytherapy, remote after-loading brachytherapy, teletherapy, blood irradiation and other purposes. Since the involved activities are rather high, attention should be paid to proper shielding, storage and security as soon as the sources are taken into use. Typical examples are ^{60}Co , ^{137}Cs and ^{192}Ir .

The potentialities of β -emitters in radionuclide therapy were the first to be recognized: this kind of radioisotopes is still the most frequently used for targeted radiotherapy. As known, β are not emitted with a precise energy: the values of the energy of this particles represent a continuous spectrum ranging from zero to a maximum value; the average is about one-third of the maximum energy. For therapeutic applications, these radioisotopes are classified in three groups according to the *mean range* of the emitted β in tissues: *short range* ($<1000\text{ }\mu\text{m}$), *medium*

range (1000 - 2000 μm) and *long range* ($>2000 \mu\text{m}$). The range represents the distance travelled by the electron in passing through matter, in this case, human tissue; of course this is an energy-dependent parameter. In Table 1.1, the mean range for some isotopes is listed.

Table 1.1: Classification of some β -emitters according to their range in human tissue. [10, pag. 332]

	Short-range		Medium-range	Long-range			
Radioisotope	^{131}I	^{177}Lu	^{153}Sm	^{32}P	^{89}Sr	^{90}Y	^{188}Re
Mean Range [μm]	910	670	1200	2900	2500	3900	3500

• Research

In health care biological research centres and pharmaceutical development facilities, many unsealed and sealed radionuclides are used: in the first form we find especially ^3H , ^{32}P and ^{33}P , ^{35}S for DNA sequencing but also ^3H and ^{14}C in GBq quantities for organic synthesis; in sealed form, ^{60}Co , ^{63}Nb , ^{137}Cs for gas chromatography, small calibration sources for gamma and liquid scintillation counting.

1.3 Belgium: a worldwide leader in Radioisotope production

All the applications listed above require the supply of several artificial Radioisotopes, the production of which is a crucial issue. One of the world's major producing countries of radioisotopes is Belgium. What guarantees to Belgium this leadership is the combined effect of the work of important companies, such as the IRE & IRE ELiT, the IBA and the SCK-CEN.

Located in Fleurus, the National Institute for Radioelements (IRE) produces radiochemicals needed especially for the medical applications, i.e., ^{90}Y in the form of YCl_3 , ^{99}Mo as Na_2MoO_4 , ^{131}I as NaI and ^{188}Re in the form of NaReO_4 . The interest of IRE is selling those radioelements to the pharmaceutical companies that manufacture radiopharmaceuticals, in the form

of radionuclide generators (see 2.2.4), solutions or capsules, which then will be used in the hospitals or in the healthcare centres. IRE exports more than 95% of its products in Europe, North and South America and Asia, contributing, in this way, to about the 20% of the global demand. [11]

Founded in 1986 in Louvain-la-Neuve, the Ion Beam Applications (IBA) S.A. focuses its work in the fight against cancers promoting the development of integrated and innovative solutions for the diagnosis and the therapy. IBA deals mainly with proton therapy, considered today to be the most advanced and targeted cancer treatment. IBA SA owns jointly with SK Capital Partners (USA) the IBA Molecular. This one has the aim of producing radio-pharmaceuticals of high quality starting from the specific radioelements.

SCK·CEN, the national Nuclear Research Centre, was established in the 1952 by the Belgian Government. It has been built in the Flemish part of Belgium, precisely in Mol in the province of Antwerp. The acronym comes from the Dutch *Studiecentrum voor Kernenergie* and the French *Centre d'Étude de l'Énergie Nucléaire* and remarks the fact that Belgium is a bilingual country. For further details about the history of SCK·CEN, please look [12]. SCK·CEN, works in many fields concerning the applications of nuclear energy and radioactivity. Concerning the production of artificial radioisotopes, SCK·CEN operates the BR2 (Belgian Reactor 2), reaching about the 25% of the world's total production of the most important radioisotopes. For instance, this allows to Belgium to be the second supplier of ^{99}Mo , the most useful radionuclide for medical applications. [13]

To better understand how these companies work, let us consider the case of production of the $^{99}\text{Mo}/^{99m}\text{Tc}$ Generator: a Highly Enriched Uranium (HEU) Target ($^{235}\text{U}/\text{U} \simeq 90\%$)² is irradiated for 7 days in BR2; the irradiated target is, then, shipped to IRE in order to separate the ^{99}Mo from the other elements contained in the target itself; after this processing, the radionuclide, in the form of Na_2MoO_4 , is shipped to IBA Molecular that, finally, creates

²The usual route for producing it is by fission, but an alternative one is the thermal neutron capture by atoms of ^{98}Mo . Since the obtained yields with this reaction are low compared with that of fission of Uranium, it results preferable producing ^{99}Mo by fission even if this involves the use of targets made of HEU, which generate high-level radioactive waste, and requires a complex chemical processing.

the Generator, which will be sold, for instance, to the hospitals.

1.4 Research and irradiation facilities at SCK

So far, SCK·CEN has operated five reactors: the *BR1*, the *BR02*, the *BR2*, the *BR3* and *VENUS*, a brief presentation of which is given below. [14] Other realized facilities are: the Laboratory for High and Medium level Activity, *LHMA*, where the damages and the ageing processes of the irradiated materials are analysed; *HADES* (High Activity Disposal Experimental Site), a laboratory, located, as well as the Greek Hades, underground (at a depth of 225 metres), where feasibility researches of storing high-level radioactive waste in the deep layers of clay are undertaken; other nuclear and non-nuclear laboratories working in the area of radiobiology, microbiology, radio-ecology, space-travel, radiochemistry and dosimetry. SCK·CEN has a long list of research partners, both Belgians and foreign. Every year, it attracts many students and researchers from all over the world and the results of its work are often innovative and forward-looking applications for medical word, industry and the energy sectors. Hence, in order to guarantee the continuity of its services, since 1997, SCK·CEN is working on the design of a multifunctional research reactor of the IV Generation, MYRRHA, able to replace the *BR2*, the operating life of which is expected until the 2023. This project is the major challenge with which SCK·CEN is facing but must be seen really as a stride towards the future that the centre takes. [13]

1.4.1 BR1

The *BR1* was the first reactor operated by the centre and has been called in this way because it was the first research reactor of the country (BR stays for Belgian Reactor). In operation since the 1956, it is an air-cooled graphite reactor fueled with natural Uranium. Even if its maximum thermal power is 4 MW, at the present, it works at 700 kW and for short periods of time it reaches 1 MW. In all these years of operations, it consumed barely 1% of its original fuel, hence, the costs related to it are very limited. It is used

for the calibration of measurement instruments, testing the materials under irradiation and for the training of nuclear experts.

1.4.2 BR2

The BR2 was commissioned in 1961, after that a full size zero power model of its core, the BR02, has been operated for about two years. It produced its first chain reaction on July 6, 1961 but became operational only after further tests in December, 1962. The used fuel is [HEU](#). The moderation is provided mainly by water, that is used as coolant too; the Beryllium matrix acts as an additional moderator. This reactor allows operation at power higher than 100 MW_{th} , but usually it is operated at a power level of 60 MW_{th} . Unlike the BR1, it has been refurbished once in the beginning of the eighties, a second time in the mid-nineties and is currently undergoing its third refurbishment. It is a pool-type reactor: the aluminium pressure vessel is immersed in demineralised water. Because of this, the safety system is mostly passive and the radiation is shielded from the environment; moreover, in case of damage occurring to the fuel, the water acts as an additional buffer against radioactive releases. In the BR2 core, the channels are distributed in this way: there is a central vertical 200 mm diameter channel (H1) and all the others are inclined forming, in this way, a hyperboloidal arrangement around it. The compactness of this design is sought to reach high fission power density. In this geometry, compactness is combined to easy access at the top and bottom covers to insert/withdraw the complex irradiation devices. A top view of the reactor is shown in [Figure 1.7](#). The BR2 was conceived as a Materials Testing Reactor (MTR), so with the major task of analysing the behaviour of the materials in nuclear reactors when irradiated with a high neutron flux (up to $10^{15} \text{ cm}^{-2} \text{ s}^{-1}$). At present, the BR2 is operated, moreover, for the production of both doped Silicon and radioisotopes in accordance with an annual regime based on five or six irradiation cycles, for a total of 120/140 operating days, consisting of 3 or 4 weeks of operation at an operative power between 55 and 70 MW_{th} according to the irradiation programme. Silicon is doped by neutron irradiation in order to achieve a high-grade semiconduc-

tor necessary to be used as the basic substance for electronic components of hybrid vehicles, locomotives of high-speed trains, solar energy systems and wind turbines. Radioisotopes are produced in the reactor by the transmutations occurring when the specific target, provided each time by the customer, undergoes neutron irradiation. For this purposes, BR2 has several dedicated channels: the central one said before (H1), other four peripheral channels (H2-H5) with the same diameter (200 mm) and several other experimental positions with diameters of 84, 50 and 33 mm. The central H1 beryllium plug presents seven sub-channels (six with a diameter of 33 mm and one of 84 mm). There, a peak thermal neutron flux up to $10^5 \text{ cm}^{-2} \text{ s}^{-1}$ is available and allows the production of several radioisotopes characterized by high specific activities, such as ^{75}Se , ^{89}Sr , ^{188}W , ^{192}Ir . For the production of radioelements such as ^{79}Kr , ^{90}Y , ^{99}Mo , ^{131}I , ^{133}Xe , ^{153}Sm , the targets are placed in peripheral reflector channels where the peak thermal neutron fluxes are between 10^{14} and $3.5 \cdot 10^{14} \text{ cm}^{-2} \text{ s}^{-1}$. Moreover, it is possible to produce radioisotopes with reactions that need fast neutron fluxes loading the targets into irradiation baskets inside a fuel element where the neutron flux above an energy of 0.1 MeV is between 10^{14} and $5 \cdot 10^{14} \text{ cm}^{-2} \text{ s}^{-1}$ (it depends on the burnup and the location of the fuel element). It is exactly thanks to its high neutron flux that it is possible to achieve high yields, so that nowadays BR2, alone, is responsible of about the 30% of the global demand of radioisotopes for medical and industrial applications. In particular, with respect to the only Molybdenum-99, BR2's averaged yearly production and the irradiation capacity peak are respectively of about 25% and 65% of the world demand.³ Because of the severe crisis in 2008-2010 caused by the temporary shutdown of the HFR (Netherlands) and the NRU (Canada), in 2010, the ^{99}Mo production was increased by 50% and, to secure this new level, an additional new equipment was installed with a dedicated cycle to operate. [15]

All this makes of the BR2 the most powerful reactor of the Western Europe. [16]

³The world demand is estimated to be approximately 12000 Ci "6-day" calibrated weekly for North America (53%), Europe (23%), Asia (20%) and the rest of the world (4%).

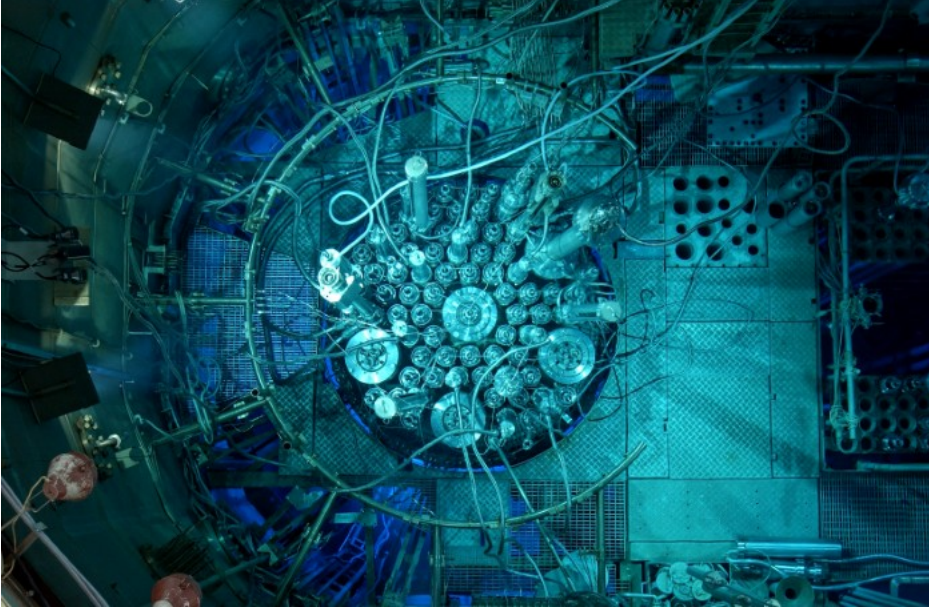


Figure 1.7: Top view of BR2. (Figure from [13])

1.4.3 BR3

The BR3 has, instead, a different story. Beside the construction of these two research reactors, the centre was interested in building a power plant, with a Pressurized Water Reactor (PWR). This technology was developed by the American company Westinghouse, on the basis of the experience obtained from powering submarines with nuclear reactors (nuclear submarines). Hence, a contract was concluded by the two parts in 1956. For Westinghouse, this was the second order recorded with a view of peaceful application. The original idea was that of installing the power plant in Schaarbeek in order to supply the 1958 World Exhibition with nuclear energy, the *energy of the future*. Since this plan failed, it was decided to install the reactor in Mol. When the BR3 became operational in 1962, it was the first PWR power plant built in Europe. It was conceived as a demonstration unit for both the construction and the exploitation and the development and the improvement of the related techniques. It played an important role in the training of the operators and the specialized personnel for power plants. It was definitely shut down in 1987 after having concluded its eleventh operation campaign.

During the operating years it has produced about 964.6 million kWh and delivered 851.7 million kWh. In 1989, the European Commission decided that the decommissioning of the BR3, together with that of three other reactors, should enter in its five-year RTD (Research and Technological Development) programme on decommissioning of nuclear installations. It was completely dismantled at the end of the 2013. For further information about it, look [\[14\]](#); more attention to its decommissioning is given in these websites [\[17, 18\]](#).

1.4.4 VENUS

Venus, a zero power reactor, was built under the Vulcain Project in 1964. The name stands for Vulcain Experimental NUClear Study. This project was carried out to evaluate the feasibility of a reactor concept with a variable neutron spectrum: in order to do that, the heavy water (D₂O) was gradually replaced with the light water (H₂O) as the fuel was consumed. In two years, several types of experiments were carried out in this reactor until when, in 1966, the Vulcain core was finally installed in BR3. Since then, VENUS was converted in order to allow neutron studies for new reactor core configuration and for the embrittlement of reactor vessels under irradiation. In this context, in 2008 SCK·CEN reconfigured VENUS reactor in order to launch the new project "Generator of Uninterrupted Intense NEutrons at the lead VEnus REactor ([GUINEVERE](#))" faced to demonstrate the feasibility of [ADS: GUINEVERE](#), started in 2010, is the first scale model of a subcritical reactor with a total lead core driven by a particle accelerator and can be seen as the real milestone for the MYRRHA project.

1.4.5 MYRRHA Project

MYRRHA Project concept is a subcritical fast reactor with MOX fuel, cooled by an eutectic alloy of Lead and Bismuth (LBE), driven by a proton accelerator (hosted in the central channel). The accelerator is needed to have high energy protons, which, bombarding the heavy-metal target nuclei of the spallation source, generate neutrons that, entering in the subcritical core ($k_{eff} = 0.955$), cause fissions. Because of the deposition of about 65% of

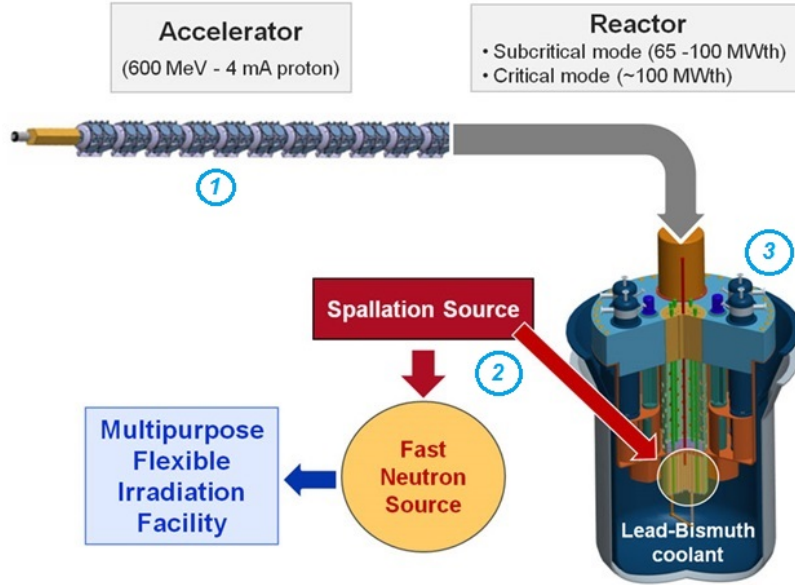


Figure 1.8: MYRRHA ADS concept. (Figure from [19])

the total beam power as heat in the spallation target, this one has to be able to evacuate heat. Moreover, the target must be small enough and as compact as possible in order to fit in the central hole of the core assembly and to create the highest possible neutron fluxes. The need of evacuate the heat deposited by protons in the target combined with the space limitations for optimal neutronic performance require forced convection heat removal by liquid metal. At last, the lifetime of the target must be appropriate compared to the operations of MYRRHA. For all that, LBE has been chosen as spallation target since it guarantees both to obtain a high neutron gain and to operate in forced convection. Figure 1.8 shows a scheme of this concept. Since it results convenient to profit from the thermal inertia provided by a large LBE volume, MYRRHA is designed to be a pool-type reactor: the primary system operates in forced circulation and the thermal-hydraulic design of the core allows to provide every position inside the core, except the peripheral ones occupied by the reflectors, the needed full mass flow for a fuel assembly; a very flexible core management is, thus, possible as well as necessary for working in dual mode. The components of the primary loop, shown in Figure 1.9, are inserted in from the top by fully-remote handling

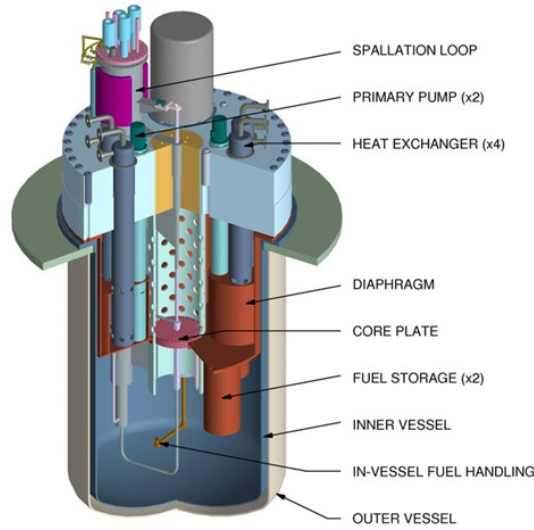


Figure 1.9: MYRRHA: Primary System. (Figure from [19])

systems and the fuel assemblies from underneath thanks to the In-Vessel Fuel Handling system (IVFH). These handling systems have been designed for both operation and maintenance (O&M) and In-Service Inspection & Repair (ISI&R) and allow to MYRRHA to be a very flexible irradiation facility. Being an ADS, it guarantees more safety and a high controllability because, when the particle accelerator is switched off, no more neutrons are generated and the chain reaction stops due to the subcriticality of the core. Moreover, regarding safety, MYRRHA has all the advantages of a Lead-cooled reactor: LBE cannot explode and in case of LOCA, for instance, the leaked coolant will solidify soon, shielding most of the radiations, without contaminating the environment, guaranteeing a very low emission; since its melting point is low (124.5 °C), LBE allows the operation at rather low temperature. The idea with which MYRRHA is designed is that of replacing the BR2, so as well as this reactor it has to preserve, first of all, those functions that makes of the BR2 a so important reactor. It has to guarantee the continuity in: testing and studying the materials, with special regard to those materials necessary for fusion technology; producing both doped silicon and radioisotopes. Thanks to the use of ADS technology, it will be possible to transmute long-lived radioisotopes in short-lived ones: as much as half of MYRRHA

core is foreseen to be loaded with high-level radioactive waste. Transmutation is a great advantage concerning the storage of waste. Even if processing waste on an industrial scale with MYRRHA is not even thinkable, surely, this facility will give the opportunity of testing and improving this technology. Furthermore, being a lead-bismuth cooled fast reactor driven by an accelerator, it will demonstrate the feasibility and the performance assessment of these technologies contributing to their development. The flexibility of MYRRHA allows to operate the reactor in both subcritical and critical mode. To control the criticality of the reactor, it is sufficient to vary the number of fuel assemblies inserted in it. Moreover, since the neutron source is not necessary, in the critical configuration the central channel hosts an [IPS](#) assembly instead of the proton beam tube: the central [IPS](#) can host samples for material testing or targets for radioisotope production that needs a higher neutron flux.

All this makes of the Multi-purpose hYbrid Research Reactor for High-tech Applications a real pioneering project. [\[19\]](#)

1.5 Master Thesis subject

The present work is the result of an internship, of about nine months, at SCK·CEN aimed to demonstrate the effective potentiality of MYRRHA, operated in critical mode, in producing a certain number of radioisotopes used in industrial and medical field. The feasibility study started with the selection of these radioisotopes and is carried out using ALEPH2 code, a Monte-Carlo Burn-Up code developed by SCK·CEN itself. Since, as said, MYRRHA is intended as the heir of the BR2, the production of many of those radioisotopes nowadays produced in BR2 is evaluated in order to guarantee a certain continuity. Thus, considering the reactions with which we want to produce them, targets are designed and located in the suitable irradiation rigs. The concepts to produce ^{99}Mo and $^{227}\text{Ac}/^{228}\text{Th}$ in this facility are already under design; so, here the possibility of obtaining a certain number of other radioelements using these same concepts is investigated. In particular, it is studied the production of:

- ^{89}Sr , ^{90}Sr (parent nuclide of ^{90}Y), ^{95}Zr , ^{131}I , ^{133}Xe and ^{140}Ba (parent nuclide of ^{140}La) as byproducts of the production of ^{99}Mo by fission of a High Enriched Uranium target;
- ^{32}P , ^{79}Kr , ^{89}Sr , ^{90}Y , ^{131}Te (parent nuclide of ^{131}I), ^{140}La , ^{153}Sm , ^{177}Lu , ^{188}W (parent nuclide of ^{188}Re), ^{191}Os (parent nuclide of $^{191\text{m}}\text{Ir}$), ^{192}Ir and ^{203}Hg by thermal capture reactions performed in the irradiation rig designed for $^{227}\text{Ac}/^{228}\text{Th}$ production.

Furthermore the production of ^{32}P , ^{24}Na , ^{89}Sr , ^{90}Y , $^{117\text{m}}\text{Sn}$ and ^{153}Sm by fast neutron capture reactions is evaluated performing the reactions in one of the inner [IPS](#) where the neutron flux is higher.

Eventually, the resulting activities per gram of target have been compared (when possible) with the values of the current activities produced in BR2.

In the following pages: more information about the characteristics and the production of radioisotopes are given in [Chapter 2](#); the methodology and the simulation tools where the study is implemented together with the description of the model of MYRRHA critical core are outlined in [Chapter 3](#); then, the evaluated performances of MYRRHA are reported in [Chapter 4](#); eventually, the obtained results are discussed in the last chapter.

Chapter 2

Radioisotopes: Characteristics and Production

2.1 Characterization of the Radiation

A radioisotope can be chosen for carrying out a particular purpose according to the nature of the emitted radiation and its physical and chemical properties. As known, in a medium, the radiation behaves differently according to its kind (α, β, γ). Dealing with radiation means knowing its behaviour in the different media on which operating. For this, it becomes very important to consider a quantity defined Penetration Power, which tell us how far the particle penetrates in the medium before losing all its energy, which is linked to the Ionization Power of the radiation itself, that is the measure of how many ions are formed when it passes through a given area. Obviously, the further the radiation can penetrate, the more spread out the caused ionization will be; this means that the less the penetrating power is, the more localised the ionizations are. The Penetration Power dependence on the energy of the radiation should also be considered, because radiation is stopped, and so absorbed by the matter, only when it loses all of energy: the higher

it is, the longer the distance travelled in the matter is. This means that the knowledge of the energy of the emitted radiation is fundamental.

In the choice of a radioisotope, we do not just look at the decay mode but also to its physical properties. Among these, the half-life ($T_{1/2}$) plays a role of primary importance together with the energy of the emitted radiations as just said. To better understand the reason why the $T_{1/2}$ is a crucial parameter, just think, for instance, about how radioisotopes are used in medical field: since these are injected in the patients' bodies, on the one hand, it is necessary to administrate to patients the smallest amount possible of radiopharmaceuticals ensuring, at the same time, the maximal uptake at the limited tumour cell surface antigen sites, and on the other one, the radionuclides must be expelled in the shortest possible time in order to not expose the patients to useless doses. At this point, it is simple to understand that, to reach the two said goals, radionuclides with a short half-life are needed.

Chemical properties of the radionuclides are useful especially in medical field: according to the chemical behaviour of an isotope it is possible to determine in which organ of the body it will accumulate and in which proportion. This allows to consider that organ as a target for the radioisotope in exam. For instance: ^{90}Sr , being similar (chemically speaking) to Calcium, will accumulate in bones, its target organ; ^{131}I will accumulate in the Thyroid; instead, ^{99m}Tc has the total body as target organ, accumulating all over it. For this reason, the former is used for the treatments and the diagnosis related to bones diseases, the latter in case of Thyroid disease and the last for several cases.

2.1.1 Constraints for Medical Uses

Radioisotopes are used in biomedical procedures for Diagnosis, Treatment and for Research. In this context, radiopharmaceutical drugs are prepared. These are composed by three important elements:

- the *Radionuclide*, which is chosen according to the purpose of the drug,

taking into account the nature of the emitted radiations and the physical and chemical properties as previously mentioned;

- a *peptide* or an *antibody*, which has the scope of driving the radioisotope to the site of interest, especially when its characteristic affinity to an organ is not enough to reach the specific site;
- a *linker* used to connect the radionuclide to the peptide or to the antibody.

In Figure 2.1, an illustrative scheme is shown.

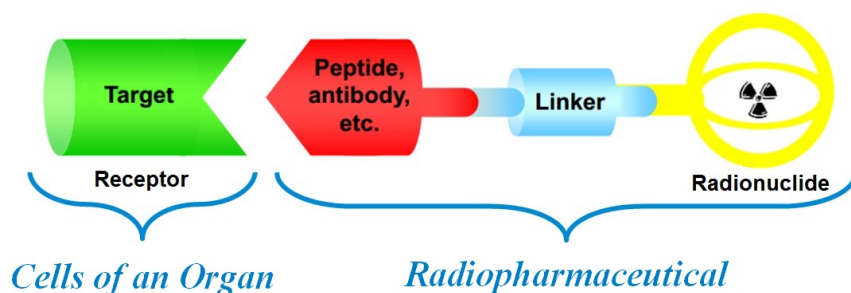


Figure 2.1: Scheme of a radiopharmaceutical.

Beyond the particular scope, not all the existing radionuclides can be injected in the human body. It is important to underline that they must respect some constraints, that is:

- a low half-life in order to be easily eliminated from the body without exposing the patient to useless radiations;
- non-radioactive progeny, to not contaminate the patient's body with other radioisotopes;
- the possibility of reaching high specific Activity, which allows to inject very small amount of radioactive substances inside the patient's body.

It must be specified that when a radioisotope is inside the body, not only its activity decreases because of its physical decay (dictated by its half-life) but also because of the natural elimination from the body due to the

biological functions of the body itself. This allows to specify a characteristic time that says how long it will take to have its amount halved just as a result of the biological functions of the body: it is referred to as *Biological Half-Life*. At this point, it is possible to define an *Effective Half-Life* that says which is the velocity with which the activity disappears from the body:

$$T_{1/2,eff} = \frac{T_{1/2,p} \cdot T_{1/2,b}}{T_{1/2,p} + T_{1/2,b}}$$

meaning with p physical, with b biological and with eff effective.

Since the $T_{1/2,b}$ are, usually, really long (of the order of one or several days or even more), looking at this formula it is possible to understand the reason why radionuclides with short $T_{1/2,p}$ are preferred for medical purposes. In Table 2.1, both the physical and biological half-life together with the effective half-life are listed for some radioisotopes of interest for this work.

Table 2.1: Physical, Biological and Effective Half-Lives of some radionuclides considered in this work. For the $T_{1/2,p}$ refer to [20]. For the $T_{1/2,b}$ refer to [21], [22], [23].

Radionuclide	$T_{1/2,p}$	$T_{1/2,b}$	$T_{1/2,eff}$
^{32}P	14.26 d	1155 d (Bone) 257 d (Total Body)	14.09 d 13.55 d
^{90}Y	64.00 h	49 y (Bone)	63.99 h
^{99m}Tc	6.02 h	24 h	4.81 h
^{131}I	8.03 d	80 d (Thyroid) 12 d (Extra Thyroid)	7.30 d 4.81 d
^{133}Xe	5.24 d	30 s	30 s
^{188}Re	17.00 h	8.2 h (Blood) 107.8 h (Total Body)	5.53 h 14.68 h

With particular reference to the case of imaging, another constraint must be considered. We know that the image is reconstructed thanks to the photons that reach the detectors. Since we deal with photons' transmission through the body both in case of Radiography and in case of Scintigraphy, when we design the devices we must consider all the possible interactions

that they can have. The imaging takes advantage of the fact that photon radiation follows a straight line carrying with itself the information related to the particular points encountered along this path. This means that we have to avoid or, at least, limit scattering events that affect the direction of the beam itself otherwise scattered photons, depositing their information elsewhere with respect to the right points, will generate useless unfocused pictures. In order to do this, it results sufficient to restrict the energy range of the used photons to values in which scattering is not so important. For instance, in the Chapter 1 we have already seen that the gamma cameras usually used are set to energy range of 100-250 keV.

2.2 Production

It is clear that, since the necessary radioisotopes have a short half-life, they cannot exist in nature. This means that, in the choice of the radionuclide for performing a generic operation, its production process plays an important role as well as its characteristics. For the production processes, great importance must be given to the reactions and to the target to be used for creating the desired isotope. In the following, some explanation is given about these but to have more details and examples refer to [10].

2.2.1 Nuclear Reactions

The used Radionuclides are created irradiating stable nuclei with neutrons, protons, alphas, deuterons, gammas or other nuclear particles. In order to achieve this goal, the reactions are carried out in nuclear reactors as well as in cyclotrons. In the first case, the reactions are of the type (n,fission), (n, γ), (n,n'), (n,p), (n, α); in case of the accelerators the involved reactions are of the type (p,n), (p,2n), (p, α), (d,n), (d, α), etc. Since the reactor produced radionuclides are generally neutron rich, they decay by β^- emission; example of reactor produced isotopes are ^{90}Sr , ^{99}Mo , ^{131}I , etc. On the other hand, the cyclotron produced radioisotopes, being usually neutrons poor, decay mainly by EC or β^+ emission; typical examples are ^{11}C , ^{15}O , ^{18}F , etc.

For the purposes of this work, the attention is focused only on the reactions taking place in nuclear reactors. This kind of reactions involves the neutron irradiation of nuclei to transmute them in the desired radioisotopes; those nuclei compose the so-called *Target*. Hitting the nucleus, the neutron is absorbed by it causing the occurrence of either *Fission* (n,f), or *Radiative Capture* (n, γ) or *Capture and consequent emission of supplementary particles* (n,n'), (n,p), (n, α) reactions, according to the kind of involved isotope. To describe these reactions and, in this way, the interaction of neutrons with matter, the quantities known as neutron cross sections (σ) are defined. These quantities take into account the probability of an interaction since, properly, the cross section represents the area around the target particle (the nucleus) that a projectile particle, in our case a neutron, has available to hit and, in this way, interact with it. This is why it has the dimensions of an area; it is commonly represented with an unit measure introduced exactly to describe it, the barn (b): $1 \text{ b} = 10^{-24} \text{ cm}^2$. The neutron cross section depends strongly on:

- the target, since the way in which the elements interact with neutrons is not the same;
- the type of reaction: we have σ_s for scattering, σ_f for fission, σ_γ for neutron capture, and so on;
- the incident neutron energy, represented as the neutron velocity before the interaction (usually reported in electronVolts (eV)).

To a lesser extent, a neutron cross section depends also on both the angle of incidence of the neutron on the target and the temperature of the target itself, which influences the vibrational state of the atoms that compose it.

It results useful to define the so-called *Reaction Rate*: as said σ represent the probability of interaction for a neutron with an atom of the target; multiplying it times the neutron flux, which represents the number of neutrons per unit area per unit time, we obtain the number of interactions per unit time:

$$\text{Reaction Rate} = \sigma(\mathbf{r}, E) \phi(\mathbf{r}, E, t) \left[\frac{\text{reactions}}{\text{s}} \right]$$

The formula shows explicitly that σ and ϕ depend on the position ($\mathbf{r}$) and the energy of the incident particle (E) and the latter on the time too. Concerning the radioisotope production, which is the topic of this work, it is better to use directly the *Production Rate*, a quantity that expresses how many reactions occur per second in the target, that is how many target nuclei, absorbing the incident neutron, are transmuted in the desired isotope. It is given by the following expression (neglecting the dependencies):

$$R = N_0 \sigma \phi \left[\frac{\text{reactions}}{s} \right] \quad (2.1)$$

where:

- N_0 refers to the number of atoms of the target and is evaluated as the ratio of the grams (g) of the target times the Avogadro constant (N_A) and the molecular mass (MM) of the involved specie:

$$N_0 = \frac{g N_A}{MM}$$

- σ is the cross section of the considered reaction [in cm^2];
- ϕ represents the incident neutron flux [$cm^{-2}s^{-1}$].

So, concerning the radioisotope production in nuclear reactors, the first quantity, N_0 is known having the grams of the target, and the other two are due to the operations of the reactor itself and, for a feasibility study like the one of these pages, must be evaluated by computational codes.

2.2.2 Target

The choice of the appropriate target material is of particular importance. It has to be composed in a good percentage by the isotope parent of the radionuclide in which we are interested and has to respect the given specifications about the contents of impurities. This is required in order to have a final product with a high level of purity, that is, with the minimum presence of radiocontaminants due to the activation of the same impurities. For this

reason pure metals or elements are preferred because, in this way, the amount of waste is limited. However in some cases, it is necessary to enrich isotopically a material to suppress competing reactions on other naturally occurring isotopes. But the choice is influenced also by the thermal and the chemical properties of the involved materials that can prejudice its use rather than that of another. So, in addition to the purity specifications, it is required:

- a good thermal conductivity and a high melting point since the target can be heated up by the energy deposition of fast neutrons, by the exothermic reactions that can take place and by the so-called " γ -heating" due to the absorption of the energy of prompt gammas from (n,γ) reactions occurring in the sample, in the fuel elements, in the reactor containment and etc. This last phenomenon is proportional to the mass of the sample itself, so it is considerable for target heavier than a gram. Moreover, in order to prevent the overheating of the target that may result in possible destruction, it is necessary to provide cooling;
- a chemical stability, which means that the target does not decompose with either high Temperature or radiation damages and does not evolve in gases, which would pressurize the containment capsule causing the rupture of the same. Instead, reactions of any kind are permitted between it and its container;
- a form compatible with the post-irradiating processing of separation.

For these reasons: organic materials and compounds together with aqueous solutions cannot be used; easily dissolving compounds are sometimes preferable over metal targets. Thus, oxides, hydrates, carbonates, halides, and other compounds are used when it is not possible to use pure metallic targets. In this case, the activation of all atoms must be considered because partner atoms do not lead to the targeted nuclides: these reactions could affect the final yield, reducing it, since a high specific activity is reached when the final concentration of the radionuclide of interest in the target is high. Examples of targets considered in this work are: Tellurium, Tungsten,

Gadolinium, Aluminium in elemental form; as oxides, La_2O_3 , Sm_2O_3 , ZrO_2 ; as hydrates, $La(C_2H_3O_2)_3$; as carbonates, $SrCO_3$; and finally, as alloy, UAl_x .

The target has to be sealed in a containment capsule for both safety and preventing cross-contamination. The container should guarantee no leaks and not corrode or overheat during irradiation. Usually, small plastic tubes (e.g., polypropylene) are used in case of short period irradiations at low neutron flux and high-purity quartz ampoules in case of longer irradiation times at higher flux. The formers are cheap and above all do not activate appreciably, the latters present some unavoidable activation of ^{30}S (3.3% natural abundance) in ^{31}S ($T_{1/2}=2.62$ h, $E_\gamma=1266$ keV (0.07%), $\sigma(^{30}S)=0.11$ b). The quartz ampoules are placed in aluminium or magnesium holders specific to the irradiation facility. There is some activation of these materials but it does not appear as a problem since the cross-sections are small and the products have very short half-lives.

2.2.3 Separation methods

Once the target has been irradiated, the required radioisotope must be extracted from the target itself and separated by the induced radioactive byproducts in order to achieve high specific activity. To reach this goal, the usual chromatography, solvent extraction, distillation and precipitation techniques are used as well as electrolysis, electro-deposition and separation based on sublimation. All these processes must respect some radiochemistry requirements: they must be adapted to the radiological hazards of exposure and possible contamination, allow the separation of very small amounts (*carrier-free*) of product from the bulk target and, at last, the time spent for the process must be as little as possible to minimize the decay losses. In this field, research is moving towards the developing of techniques specific for each application: for instance, solvent extraction is usually used in case of purification of the reactor-produced ^{191}Os and ion exchange to separate transition metals from each other and from rare earth elements. When (n,γ) reactions are involved in the production processes, the so-called *Isotope Effects* must be used for separation: in this case, target and product differ just

for one neutron; thus, to separate them we can make advantage only of the differences in the physical and chemical properties of atoms and molecules resulting from isotopic substitution ($T_{1/2}$, energy of radiation and cross section are not considered). For further details about the chemical and isotope separation procedures refer to [24, 25]. Obviously, these processes for radioisotope production are used as well for the treatment of the nuclear waste since the basic concepts are the same.

As example, let us consider the extraction method of ^{99}Mo from a Uranium target carried out by IRE; ^{131}I and ^{133}Xe result as byproducts of this process. The U-target is dissolved in an alkaline solution (3M NaOH and 4M NaNO_3), method well-adapted for aluminium-containing targets as the one we refer. Formed already as gas inside the target, Xe is released at this step and thus trapped in Cu clippings to be then purified. Hydroxides of Uranium and of most of the fission products precipitate in the dissolving solution, whilst Molybdenum and radioiodine are still present as MoO_4^- and I^- , in ionic form. To extract them, the solution is first filtrated and then acidified with concentrated nitric acid (HNO_3). At this step, I is released and trapped in Platinum asbestos to be then purified. Instead, the acidified solution is passed through an alumina column. There, Mo is retained and to elute it NH_4OH is used. Eventually, to purify the Mo solution, ion-exchange resin and activated carbon is used. [26]

2.2.4 Radionuclide Generators for Medicine

The radioisotopes used in Nuclear Medicine have short half-lives, usually of the order of days or few hours and, just in rare cases, of the order of few minutes or seconds (e.g., ^{191m}Ir used for angiography has a $T_{1/2} = 4.94$ s). Since the involved half-lives are so short, sometimes it is more convenient to produce not directly the radionuclide of interest but its parent nuclide: the parent decays, as time goes by, so it is possible to recover from it the required isotope. This is made considering that, after the irradiation in the reactor or in the cyclotrons, the targets must be transported to the processing facilities to separate the radionuclide from the rest and, then, to be

delivered to the client. In the meantime, radionuclides with very short half-lives could rapidly disappear. Having a higher half-life, the parent nuclide can be easily transported over long distances to hospitals and there separated from the (required) daughter nuclide: this procedure gives the necessary time to process and transport the required activity where it needs. In this case, what is delivered to the client is the so-called *Radionuclide Generator*.

When the parent half-life is higher than the daughter one, between them *transient equilibrium* is established. This is shown in Figure 2.2: starting from a point in which we have only the parent activity, the daughter activity begins to grow until it reaches a maximum; after that, it starts to decrease showing the same behaviour of the parent.

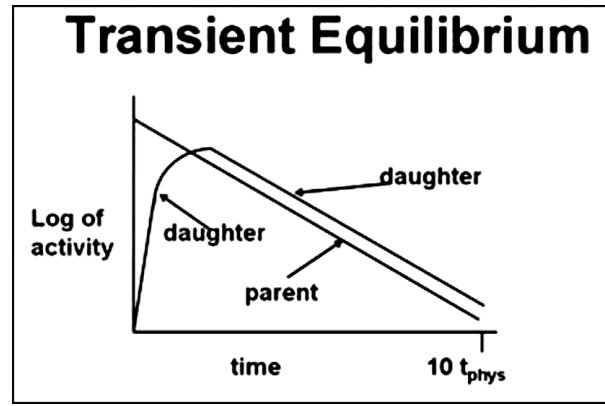


Figure 2.2: Transient Equilibrium. (Figure from [27])

In Table 2.2 the values of the half-lives of both parent and daughter nuclide of the generators considered in this work are listed.

Table 2.2: Half-lives of both parent and daughter nuclide of the considered generators. [20]

	$^{90}\text{Sr}/^{90}\text{Y}$	$^{99}\text{Mo}/^{99m}\text{Tc}$	$^{140}\text{Ba}/^{140}\text{La}$	$^{188}\text{W}/^{188}\text{Re}$	$^{191}\text{Os}/^{191m}\text{Ir}$
Parent	28.79 y	65.98 h	12.75 d	69.78 d	15.40 d
Daughter	64.00 h	6.01 h	40.29 h	17.00 h	4.90 s

To better understand this behaviour, Bateman equations are used indicating with 1 and 2 namely the parent and the daughter nuclide:

$$\begin{cases} \frac{dN_1}{dt} = -\lambda_1 N_1 \\ \frac{dN_2}{dt} = \lambda_1 N_1 - \lambda_2 N_2 \end{cases}$$

The solution to the proposed system of differential equations is:

$$\begin{cases} N_1 = N_{1,0}e^{-\lambda_1 t} \\ N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_{1,0}(e^{-\lambda_1 t} - e^{-\lambda_2 t}) + N_{2,0}e^{-\lambda_2 t} \end{cases}$$

As said, to have transient equilibrium, the parent nuclide is longer-lived than the daughter, $T_{1/2,1} > T_{1/2,2}$, that is $\lambda_1 < \lambda_2$. This means that, when t becomes sufficiently large, $e^{-\lambda_2 t}$ is negligible compared to $e^{-\lambda_1 t}$, so:

$$N_2 = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_{1,0}e^{-\lambda_1 t}$$

The ratio of the numbers of the two radionuclides is:

$$\frac{N_1}{N_2} = \frac{\lambda_2 - \lambda_1}{\lambda_1}$$

and, consequently, the ratio of their activities is included in the range $[0, 1]$:

$$\frac{A_1}{A_2} = \frac{\lambda_2 - \lambda_1}{\lambda_2} = 1 - \frac{\lambda_1}{\lambda_2}$$

This means that, when equilibrium is reached, the daughter activity is greater than the parent one by the factor $\frac{\lambda_2}{\lambda_2 - \lambda_1}$. A radionuclide generator is a concept based on the possibility of easily separating the decaying parent and daughter radionuclides taking advantage of the different chemical properties.

The targeted nuclide is, in this way, obtained in a pure radionuclidic and radiochemical form. Since parent and daughter nuclides are in the condition of transient equilibrium, it is possible to elute the daughter activity, separating it from the parent before it reaches equilibrium, when its activity reaches the maximum; then it will grow again until it reaches a maximum and is in equilibrium with the parent. It goes without saying that the elution does not compromise the parent decay. This elution and regrowth can be continued as long as there are useful amounts of the parent radionuclide available. In Figure 2.3, the case of $^{99}\text{Mo}/^{99m}\text{Tc}$ is represented: it is the most widely used radionuclide generators and allows to elute ^{99m}Tc every 24h from the alumina column (Al_2O_3) on which the ^{99}Mo is immobilized due to its very high affinity for alumina in the form of sodium molybdate (Na_2MoO_4).

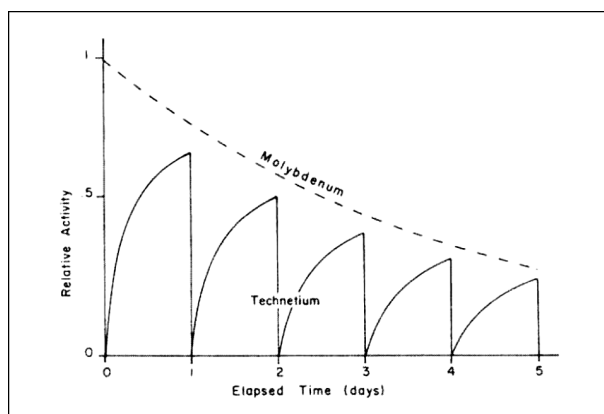


Figure 2.3: Elution sequences of ^{99m}Tc from ^{99}Mo parent nuclide. (Figure from [28])

The elution process is known as "milking the cow": passing through the column, the eluant, a 0.9% saline solution, allows extracting the anion $^{99m}\text{TcO}_4^-$ in the form of sodium pertechnetate (NaTcO_4), taking advantage of the total lack of affinity of it for alumina, whereas molybdate remains completely bound to the alumina; the eluate is then collected in a shielded, evacuated sterile vial and calibrated prior to use. The Figure 2.4 shows the main components of a $^{99}\text{Mo}/^{99m}\text{Tc}$ Generator.

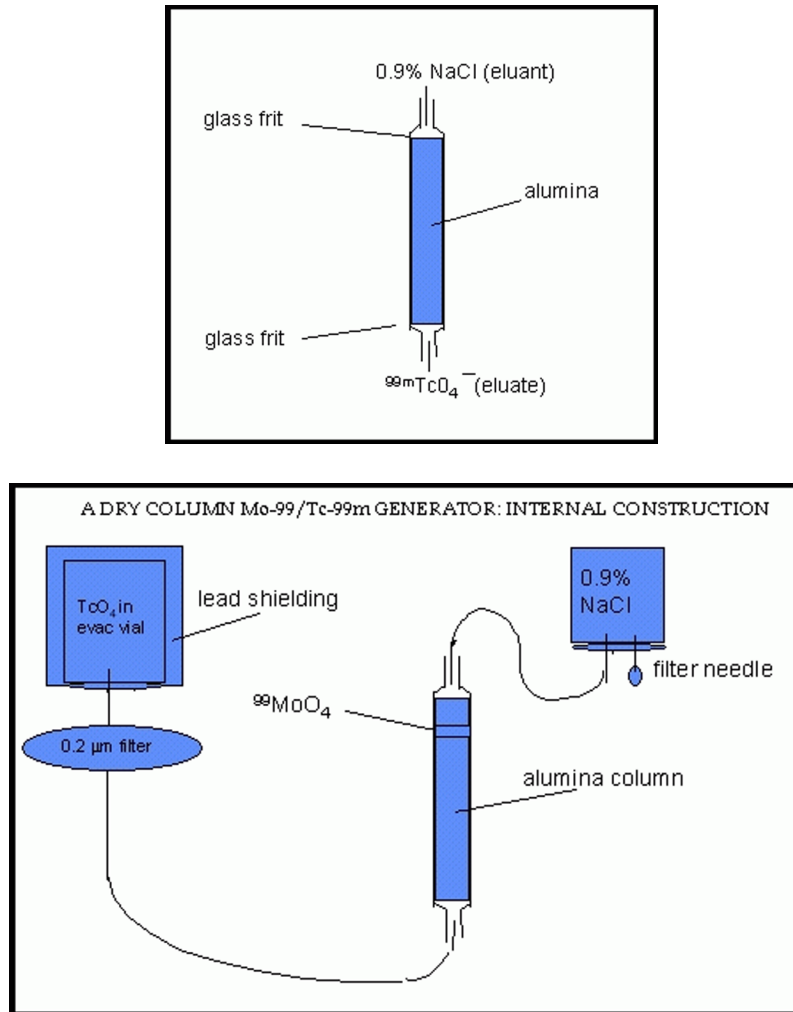


Figure 2.4: Alumina column and the internal components of a $^{99}\text{Mo}/^{99m}\text{Tc}$ Generator. (Figure from [27])

Chapter 3

Simulation tools and model of MYRRHA critical core

The study presented in these pages, apt to evaluate the radioisotope production capabilities of MYRRHA, operated in critical mode, deals, obviously, with the neutronics of the reactor itself. In this context, we can see the reactor just as a source of neutrons, necessary to ensure that the desired reactions occur. Equation 2.1 shows a simplified way for the evaluation of the production rates; but, even in this case, we need to determine the neutron flux and the neutron cross section for the specific involved reaction, not easy work given the dependencies of these on the position in the reactor, on time and on the neutron spectrum. In order to have a good estimation of the transmutation rate of a certain nuclide i , we need to solve the first-order integro-differential equation showed below:

$$\begin{aligned} \frac{dN_i}{dt} = & - \sum_{j \neq i} \left[\lambda_{ji} + \int \varphi(E, t) \sigma_{ji}^{tr}(E) dE \right] N_i + \\ & + \sum_{j \neq i} \left[\lambda_{ij} + \int \varphi(E, t) \sigma_{ij}^{tr}(E) dE \right] N_j \end{aligned}$$

The subscripts represent the direction of the transmutation course: ij

considers the transformation of nuclide i into nuclide j and ji the opposite. Of course, the λ s represent the product of the decay constant of the parent nuclide times the involved branching ratio for obtaining the considered daughter nuclide. The other terms have obvious meaning. It can be noticed that: the first sum in the right-hand side shows the consumption rate of nuclide i through both decay and reactions; the second, the accumulation rate of nuclide i . Since we have to integrate both the neutron flux φ and the transport cross-section σ^{tr} over energy, a full energy-scale neutron transport simulation is needed. To have this result, we need very powerful tools to model the operation of the core considering the high-energy neutron cascade in it, the production of neutrons by different reactions in the reactor and the transport of all these. To deal with such a problem, we cannot use deterministic algorithms since the geometry of the reactor and the transport of particles are too complex. So, we resort to the so-called Monte Carlo methods, which are numerical statistical methods able to simulate complex systems knowing their basic physical phenomena.

The use of Monte Carlo method is really widespread because it is a very general statistical method and, consequently, it can be applied to any statistical phenomenon and to those deterministic phenomena that can be described statistically. Using these methods, we do not have the problem of solving complex integro-differential equations because they do not use deterministic equations but simulate the basic statistical phenomena to evaluate the average value of the involved statistical quantities together with their statistical uncertainties. Before starting the numerical calculation of the system behaviour, it is necessary to represent the involved quantities as expected values of random variable distributions; these values have to be defined consistently with the requested results. It is important to underline another great advantage of using Monte Carlo methods: they do not need discretization schemes, which are not able to reproduce exactly the system geometry. Considering our case of study, after having exactly defined the geometry of MYRRHA, what we do using Monte Carlo method is tracking the neutron life, considering all the interactions that occur in the medium, averaging it on the behaviour of all the particles involved in the simulation. To better

understand the difference among Deterministic methods and Monte Carlo methods, consider that the firsts solve the transport equation for the average particle behaviour but for the seconds the results are obtained simulating individual particles and by focusing on precise aspects of their average behaviour. The obtained information is affected mostly by the uncertainties coming from the nuclear data, since the statistical errors of the computation can be made negligible with respect to those due to the said uncertainties running sufficient neutron histories. This means that, to carry out reliable calculations, the codes must refer to reliable data; usually, these data are evaluated experimentally and organized in tables (*libraries*).

The computational codes adopted for carrying out the simulations at SCK-CEN are MCNP and MCNPX. These codes are able to handle and model the complex geometry of the systems to be studied providing very rigorous treatment of various particles, relevant for designing of nuclear systems. Using Monte Carlo methods, we can determine the average total neutron flux and the average neutron cross sections in a specific moment in time: running MCNP(X), we are carrying out a steady-state calculation. But, since for this work we are interested in producing radionuclides, we have to focus on their evolution in time. As seen, this is dictated by the difference between the production rate, from the depletion of the target nuclei, and their decay rate (because we do not have to forget that, being radioactive nuclides, they vanish in time according to the specific half-life). What we need is the coupling of the MCNP(X) code with an efficient Depletion code to solve Bateman equations. Since 2004 the researchers at SCK-CEN are developing ALEPH, a Monte Carlo Burn-up code that allows all that.

In the following pages, more details about the used codes, especially of ALEPH2, and all the necessary tools for running the necessary simulations are given. Eventually, the description of the MYRRHA critical core design is presented.

3.1 MCNP(X)

What is exactly MCNP and what does it allow to do is explained in the following lines:

MCNP is a general-purpose, continuous-energy, generalized-geometry, time-dependent, coupled neutron/photon/electron Monte Carlo transport code. It can be used in several transport modes: neutron only, photon only, electron only, combined neutron/photon transport where the photons are produced by neutron interactions, neutron/photon/electron, photon/electron, or electron/photon. The neutron energy regime is from 10^{-11} MeV to 20 MeV for all isotopes and up to 150 MeV for some isotopes, the photon energy regime is from 1 keV to 100 GeV, and the electron energy regime is from 1 KeV to 1 GeV. The capability to calculate k eigenvalues for fissile systems is also a standard feature. [33, chap. 1, pag. 1]

The structure of an input file is the following:

- Title Card
- Cell Cards [*Block 1*]
- Surface Cards [*Block 2*]
- Data Cards [*Block 3*]

Block 1 and *Block 2* are used to model all the components of the system to be studied in terms of geometry and material. The description of all the used material is given in *Block 3*, which contains, furthermore, the *Source specifications*, in terms of position, energy and direction of the first neutrons introduced in the core before the occurring reactions create others, and *Tally specifications*. The tally cards are used to specify what type of information we want to gain from the calculation. There are several types of tallies (current across a surface, flux at a point, heating in a region...) but we are interested mainly in the one giving the *Neutron Flux Averaged over a cell*, type *F4:N* [particles/cm²]. The mathematical expression of F4:N is:

$$F4 = \frac{1}{V} \int_V dV \int_E dE \int_{4\pi} d\Omega \Phi(\mathbf{r}, E, \Omega) \quad (3.1)$$

where $\Phi(\mathbf{r}, E, \Omega)$ is the energy and angular distribution of the fluence as a function of position. The formula represents the sum of all the contributions WT/V to the flux in the cell of all track-length T within the specified cell volume V made by a neutron of weight W and energy E . So, to obtain the average flux over a cell, we have just to specify in the tally card the use of *F4:N tally* on the desired cell, which for our purpose is the one where the targets are located. For a more detailed discussion about the formulation of a MCNP input file, refer to [33, 34].

3.2 ALEPH, a Monte Carlo Burn-up Code

The simulations for the study carried out in this work have been made mainly with ALEPH2.5 [35]. This is a Monte Carlo Burn-up code under development at SCK-CEN that, by the addition of only several extra cards (16 in version 1 reduced to 5 in version 2.5) to an usual MCNP(X) input file, allows to evaluate, for instance, the fuel depletion or the evolution of radioisotopes within the considered cells in a certain timespan. This is made possible by the coupling of MCNP(X) with efficient solvers of Bateman equations. The depletion code used for the version 1 was ORIGEN 2.2; this has been replaced by the advanced depletion module based on RADAU5 implicit Runge-Kutta algorithm in version 2.5. This change enables the predictor-corrector mechanism, calculation of heating during irradiation and decay, calculation of neutron sources due to decay, fuel management.

The five necessary extra cards needed to run an ALEPH2 input are:

- *DATN*, where indicating the Datapath to the folder containing neutron-induced data.
- *MCNP*, the MCNP(X) executable.
- *BURN*, in which specifying which are the materials to be followed in the problem (these materials must be specified in the Material Card of

the MCNP(X) part).

- *VOL*, where to list the volumes occupied by the selected materials.
- *Irradiation and/or Decay History*, where declaring the irradiation (at constant flux, IRF, and/or constant power, IRP) and/or the decay (DEC) timesteps in which the considered time interval is subdivided (do not forget that ALEPH allows to evaluate the evolution in time of the amount of desired material). The steady-state irradiation source strength in source particles per second is determined by this formula:

$$S = \frac{P_{tot}}{C \cdot F \cdot \sum_j w_j Q_{f,j}} \quad (3.2)$$

where:

- P_{tot} is the total power of the system at the given step;
- C is the conversion coefficient from MW to MeV/s;
- F is the total number of fission occurred in the system (per source particle);
- w_j is the weight of the fissions occurred globally in the system on a fissionable nuclide j and $Q_{f,j}$ is the total recoverable energy release per fission event occurred on the nuclide j , so that $\sum_j w_j Q_{f,j}$ gives the total energy release in the system due to fission (in MeV).

The flow chart of a typical simulation is shown in Figure 3.1. When ALEPH starts, a steady-state calculation of MCNP(X) is run to obtain the neutron flux and the spectrum that will be used to evaluate the reaction rate and so, thanks to the depletion code the nuclide concentration (step 0). It goes without saying that the values of the neutron fluxes are evaluated thanks to the *F4:N tally*. The reaction rate (RR) is evaluated by ALEPH according to this formula:

$$RR_i = N_i \int \sigma_{i,x}(E) \phi(E) dE = N_i \sum_{g=1}^n \sigma_{i,x,g} \phi_g \quad (3.3)$$

where:

- the subscript i is referred to the considered (target) nuclide of which N_i is the concentration, and the subscript g is related to the neutron energy bin;
- $\sigma_{i,x}(E)$ is the neutron cross section of reaction x averaged over the spectrum, related to the nuclide i , as a function of the energy. In particular, for each group g :

$$\sigma_g = \frac{\int_{E_{g-1}}^{E_g} \sigma(E) \phi(E) dE}{\int_{E_{g-1}}^{E_g} \phi(E) dE}$$

- $\phi(E)$ is the total neutron flux (averaged over the considered cell).

ALEPH allows the possibility of using the Predictor-Corrector: if it is set, at the half of the considered time step the obtained concentrations are used for recalculate the flux and the spectrum and, eventually, the concentrations on the basis of these are given at the end of the same time step. As the flow chart shows, MCNP(X) is updated only if the user specified it, otherwise, the already stored flux and spectrum are used, together with the new concentrations, to evaluate directly the reaction rate. The number and the duration of each time step are defined in the Irradiation History. In the output, the results are organized in table for facilitating the reading; the evaluated activities, which are what we are looking for, are presented in terms of Ci per cm³.

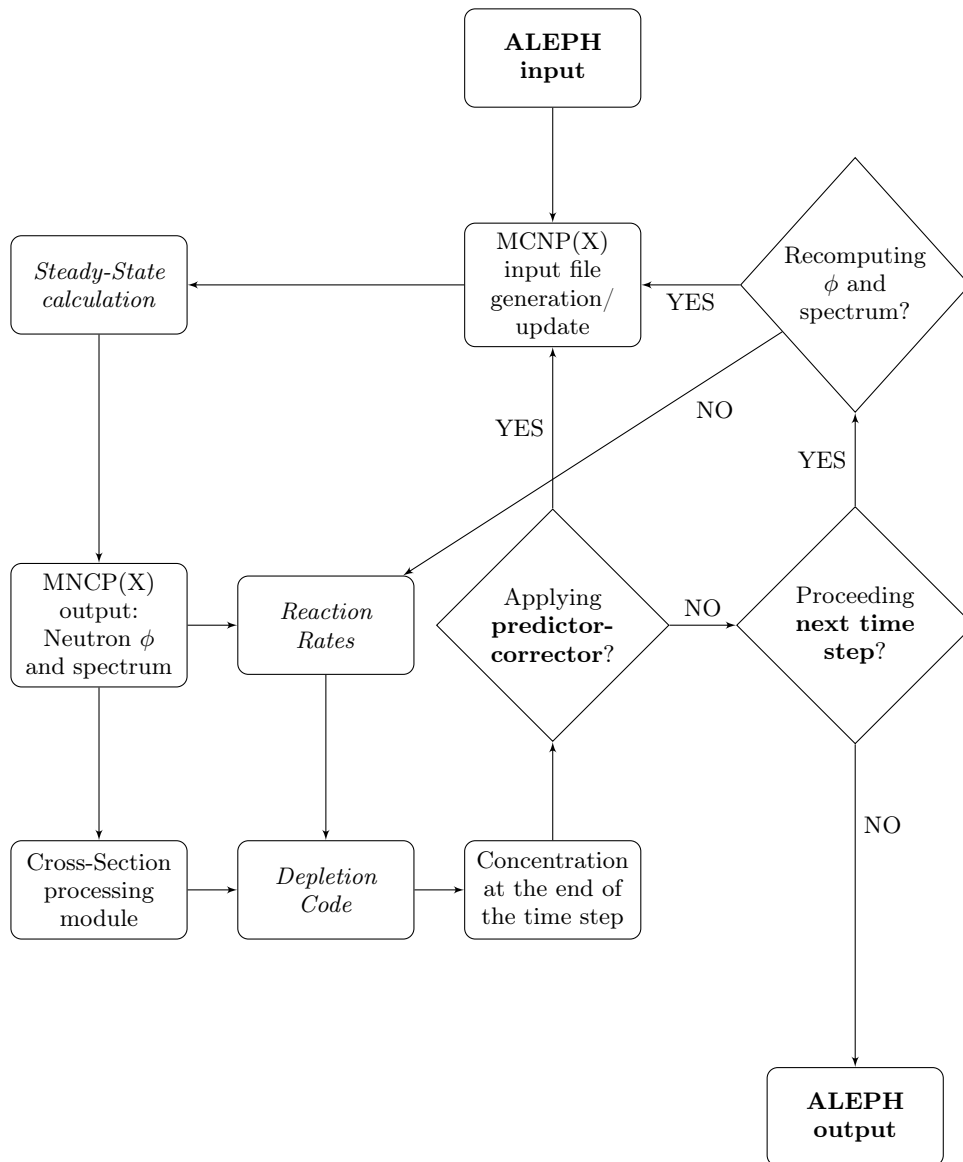


Figure 3.1: ALEPH2 code flow chart.

3.3 Nuclear Data Libraries

In order to perform a Monte Carlo simulation, we need nuclear data: interaction cross-sections, secondary particle energy-angular distributions, multiplicities and so on. What is important is especially the reliability of these data because this affects in a sensible way the results of the computation. MCNP and MCNP(X) are already provided of sets of nuclear data in ACE-format. Worldwide, many *libraries* collecting evaluated nuclear data have been created: the European JEFF and TENDL, ENDF/B in USA, the Japanese JENDL, the Chinese CENDL, and the Russian BROND/RUSFOND.

At SCK·CEN, libraries of neutron cross-sections on a temperature range between 300K and 2100K (every 50K till 1000K, then every 100K up to 1500K and thus every 300K until 2100K) have been created on the basis of JEFF-3.1.2, ENDF/BVII.1, JENDL-3.3 and JENDL-3.4. In particular, the library JEFF-3.1.2 has been chosen as reference for simulating neutron transport and depletion calculations in the MYRRHA core. It is a general purpose library in ENDF-6 format containing incident neutron data for 381 isotopes or elements from Hydrogen (1-H-1) to Fermium (100-Fm-255). The Joint Evaluated Fission and Fusion File ([JEFF](#)) project is mainly an international collaboration of Data Bank member countries coordinated by the NEA Data Bank. The [JEFF](#) library results by the combined efforts of the [JEFF](#) and EFF/EAF (European Fusion File/European Activation File) Working Groups for producing common sets of evaluated nuclear data.

3.4 SCK·CEN Computer Cluster

Monte Carlo simulations require high processing power since, to reach good accuracies, millions of particle histories need to be simulated for reducing the statistical errors. The use of standard sequential computers for such a simulation would involve several weeks or months of wait for obtaining the results. This is the reason why parallel processing platforms have to be used. A low-cost solution is that of *Computer Clusters*, a set of computers (the nodes) with multiple CPUs. At SCK·CEN, the simulations are run in routine

using a cluster operating on LINUX, a parallel computer environment with MPI-multiprocessing. This cluster contains currently 76 nodes for a total of 762 CPU cores.

3.5 MYRRHA Critical Core (rev. 1.6)

The study presented in these pages has been performed considering the reactor at the startup, with a fresh core, working in the critical mode. As it is possible to imagine, with respect to the case of subcritical mode, the main differences in the design of the core consist in the removal of the accelerator, no more necessary for starting the reaction chains, since the core is made critical by increasing the number of Fuel Assemblies (FA) and in the introduction of a secondary shutdown system, which is fully extracted during operation and fully inserted downward only for emergency scram. The layout of the core is strongly influenced by the shutdown systems criteria. The design of MYRRHA critical core (revision 1.6), composed of 211 hexagonal channels, is shown in Figure 3.2. It foresees:

- 78 channels loaded with FA (in orange),
- 10 IPS for material testing and radioisotope production (in yellow),
- 6 Control Rods (CR) (in green),
- 3 Safety Rods (SR) (in light green),
- 74 LBE-filled dummies (in light blue),
- 40 BeO-filled dummies (in blue).

Before concentrating on the irradiation rigs for radioisotope production, a brief presentation of the different assemblies composing the core is given. [19, 36]

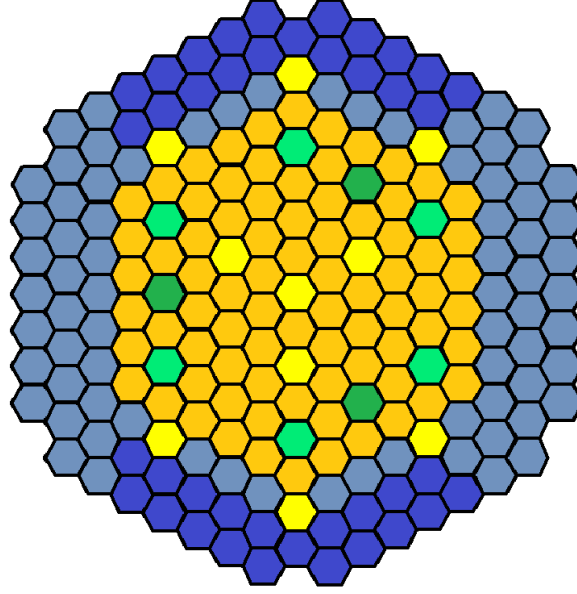


Figure 3.2: MYRRHA Core design.

Fuel Assemblies are composed of 127 fuel pins arranged with a triangular pin lattice. As said, the number of [FA](#) varies according to the mode in which the reactor is operated. A fifteen-step reshuffling scheme is foreseen: the number of fuel assemblies per batch changes between 3 and 6 and, in particular, the elements of a batches are chosen to be 120°-symmetric. The operational cycle lasts 90 days.

Control Rods and Safety Rods are used for the reactivity control of the reactor. The former act as operation and primary shutdown system (control and scram); especially when the reactor works in [ADS](#) mode, the [CR](#) are used as absorbing device to enhance the flexibility of the reactor itself and only during refuelling they are operated manually since the criticality level must be guaranteed below 0.95. The latter, introduced instead into the reactor when the core has been made critical, work only in case of emergency to scram the reactor. These two systems are completely independent, redundant and diverse as required by safety. As a matter of fact, even the insertion into the reactor is different: for the [CR](#), it is from the bottom of the core and takes advantage of the buoyancy effect

of the rods in LBE, due to the different densities; **SR** are inserted by the top of the core, driven by gravity, by a pneumatic assisted fall. In terms of reactivity: in order to limit the number of **CR** to be used, these have been designed to have a high anti-reactivity worth; the **SR** design foresees an extended safety margin and the possibility of sustain a core deformation of about 10 mm. In both cases pellets of B_4C equip the rods. The Boron contained in these pellets has an enrichment of about 90% in ^{10}B , which results a very powerful absorber, useful for the purpose of controlling the reactivity. The design of **CR** and **SR** includes a reflector made of $YZrO$ acting as driver material. The cladding is made of 15-15Ti SS and both the bundles are cooled by LBE and inserted in T91 guide tubes of different thickness. The **CR** are positioned at the boundary of the fissile zone in order to allow the slightly increasing of the flux in the centre and to limit the perturbation of its spatial distribution since, most of the time, they are partially inserted during operation; moreover, it has to be noticed that more than 1 **FA** separates the **CR** for avoiding common mode failures in case of a significant core deformation. The **SR** system is, instead, positioned in the middle zone of the core.

Dummies are of two types: LBE- or BeO-filled. The major role of both is to reflect the incoming neutrons to reduce the leakages and, in particular, the BeO-filled dummies act as lateral reflector to shield the core barrel (reflection due to BeO is more accentuated than to LBE). The number of the assemblies varies according to the configurations of the core (sub-critical, critical, all **IPS** in, etc.), but, due to their function, they always surround the active core.

The In-Pile Sections occupy in the configuration presented above ten channels: four of these are located in the central part of the core and the remaining out of the active core. Since the reactor does not work in **ADS** mode, the central channel is free to host an **IPS** instead of the accelerator. Due to the high energy of the neutrons in the centre of the core, the inner **IPS** are used mostly for the material testing under high

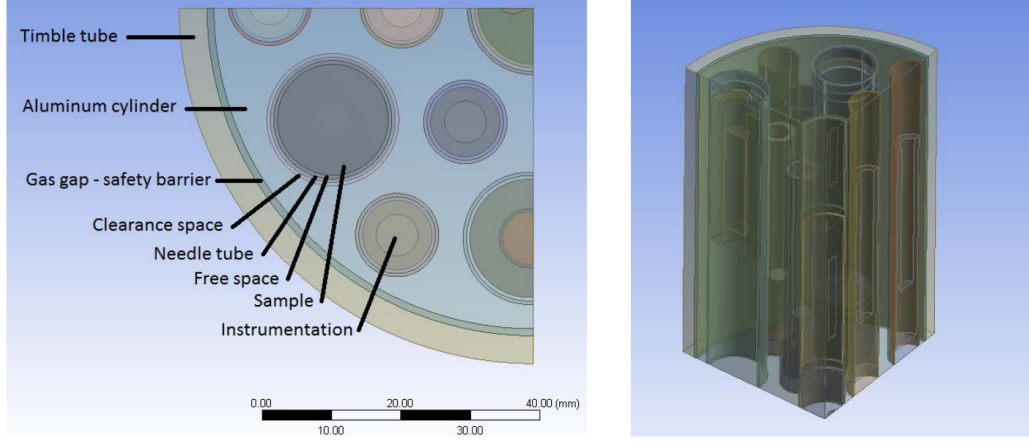


Figure 3.3: Top and 3D views of an IPS assembly for Material Testing. (Figure from [37])

neutron irradiation. For this purpose, seven sub-channels per IPS are designed to allocate the samples. Cooling has to be guaranteed and is provided by Helium. Figure 3.3 shows the design of these assemblies where it is possible to distinguish the holes of the sub-channels from those of the instrumentations. The other IPS are used mainly for radioisotope production. These are placed farther from the centre because most of the reaction involved in the production processes needs thermal neutrons to occur and in those positions the incoming neutrons are slower, so, it is easier to moderate them. For slowing down the incoming neutrons, Beryllium baskets are foreseen for allocating the targets. Beryllium is a good moderator, especially because of its low mass, but it is costly. A further moderation can be provided by light water, which flows in the basket itself for cooling down the target (refer to section 2.2.2). Light water has been chosen for this purpose because, from a safety point of view, the possible interactions among it and the LBE in the reactor are not dangerous. As well known, light water is a very good moderator, having a high neutron scattering cross section, even if its absorption cross section is not optimal. Moreover, it is easily available at reasonable costs. In the studied configurations, it is injected from the top; then, removing heat from the target, it becomes hotter and goes up again; the two flows (up and

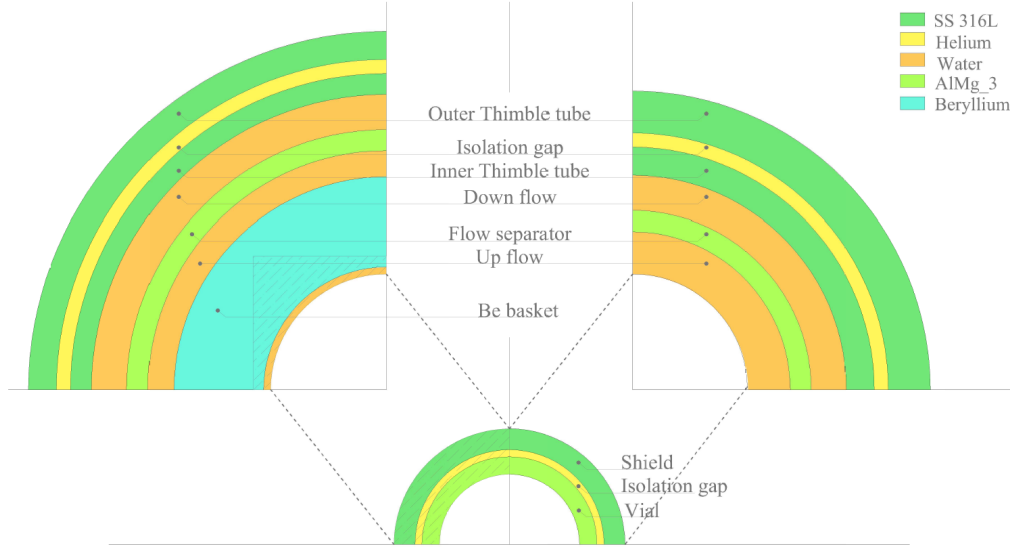


Figure 3.4: IPS design.

down) are separated by an AlMg₃ layer. All is contained in two concentric thimble tubes made of SS 316L separated by a layer of Helium. So far, the production of $^{227}\text{Ac}/^{228}\text{Th}$ and ^{99}Mo has been investigated in detail by the MYRRHA team. The configuration of the relative irradiation rigs is shown in Figure 3.4. In the first case, the Radium target for the production of ^{227}Ac and ^{228}Th is inserted in a vial separated by a layer of helium 2 mm thick from the capsule where it is inserted. This one is represented in the middle of the figure. Two configurations are under design, namely with (on the left) and without (on the right) a Beryllium basket. Of course, the one without the basket is more compact and presents a thicker layer of water surrounding the capsule and thicker thimble tubes. The configuration with the Be basket is dashed in order to show on the same figure the configuration for ^{99}Mo production. In that case, the basket presents a squared hole in which the HEU target plates are inserted thanks to suitable AlMg₃ spacers that allow water to flow through to cool them down. In Figure 3.5, the MYRRHA core configuration for the production of these radioisotopes is shown: four IPS are dedicated to the production

of ^{99}Mo and so are loaded with UAl_x targets in Beryllium baskets; the other two are loaded with the RaCO_3 targets but only in one of these the targets are placed in Be baskets.

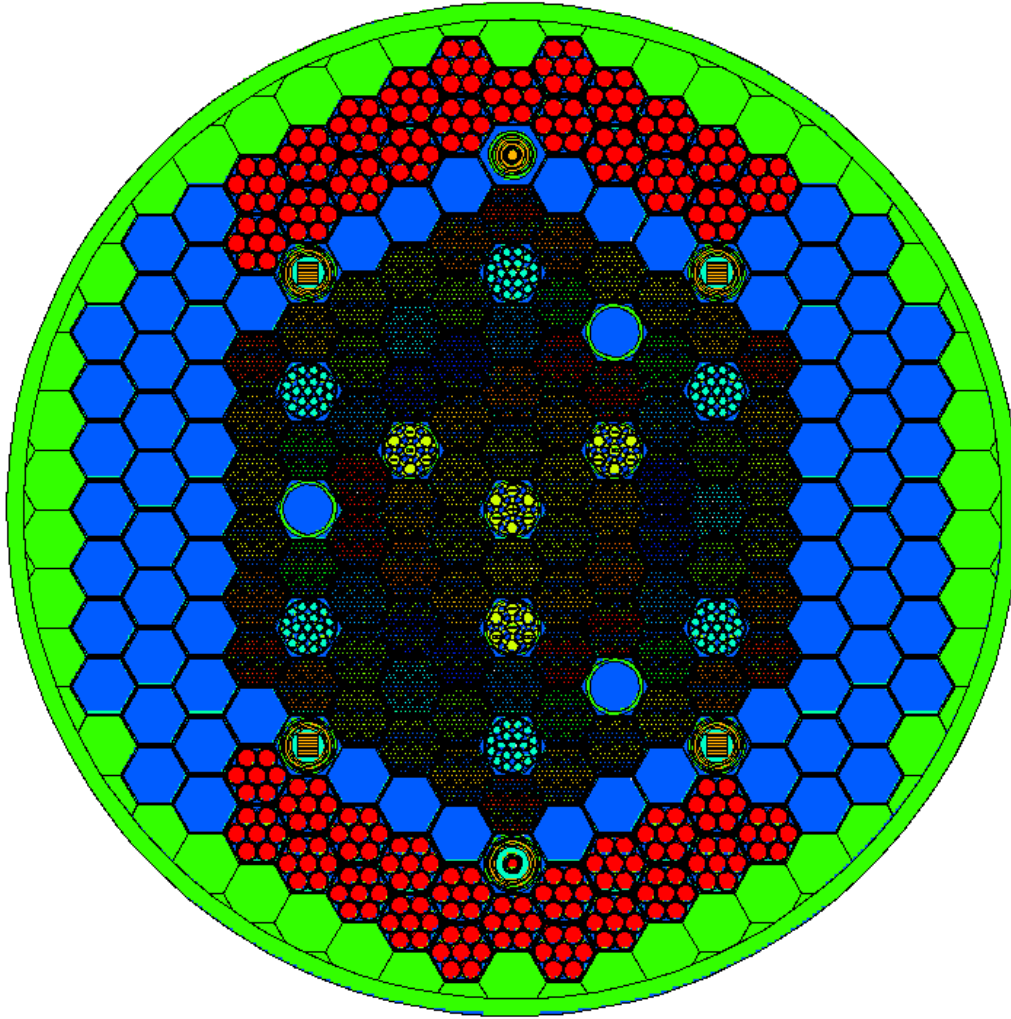


Figure 3.5: Generic version of MYRRHA Core Layout.

Chapter 4

MYRRHA radioisotopes production capabilities

The objective of this thesis is to prove the effective potentiality of MYRRHA in producing a certain number of radioisotopes used in industrial and medical field. In the following pages, the feasibility study is presented as it has been carried out. Thus, in the first section, the selection of the radioisotopes to be produced, needed before running the simulations, is explained and the reactions, with which we want to produce them, together with the targets, to be located in the suitable irradiation rigs, are listed; then, the second section shows the main part of the work in which the resulting activities per gram of target from the simulation are compared, if possible, with the values of the activities produced currently in BR2. Since it has been decided to perform even some reactions occurring with the capture of fast neutrons, those results are presented without comparison with other terms but just reported at the level of the saturation.

4.1 The Selected Radioisotopes

We could clearly produce in the reactor all those radioisotopes the creation of which is related to neutron captures but, inevitably, this study is focused

on the production of those that are related already to a specific economic market. The following list contains just a part of those radionuclides produced in BR2. For each of them, the peculiarities are presented together with the investigated routes of production and the possible uses. For some of these nuclides, different routes of production are studied. Moreover, since MYRRHA is a fast reactor, it will be possible to consider nuclear reactions, such as (n,n') , (n,p) and (n,α) , the cross-section of which presents a threshold in correspondence of high neutron energies. This is made because, since the usual (n,γ) reactions deal with target nuclides of the same species of those to be produced, the treatment for separating them at the End of Irradiation (EOI) are not so easy to perform, relying on the *isotope effect*; therefore, sometimes, it is more convenient to produce them starting from a nuclide of a different species because having simpler and more efficient separation methods allows to reach higher yields at the end of all the process of production. Eventually, it will be possible to establish if these alternative routes can be exploitable too.

The selected radioisotopes are now presented with their routes of production, characteristics (refer to [20]) and uses.

- ^{32}P , with a half-life of 14.3 days, is a pure β^- -emitter. It decays in the stable ^{32}S and the maximum energy that the emitted β^- can have is 1.7 MeV. It is produced by a (n,γ) reaction from a target of natural Phosphorus (^{31}P). It is investigated, moreover, the possibility of producing it from a target of ^{32}S by a (n,p) fast neutron reaction. It finds many applications in the medicine:
 - identification of malignant tumours thanks to the natural tendency of cancerous cells of accumulating more phosphate than normal ones;
 - Polycythaemia Rubra Vera (blood cell disease) and Leukaemia treatment;
 - Bone diseases diagnosis and treatment;
 - SPECT imaging of tumours;

- Pancreatic cancer treatment;
 - Radiolabeling;
 - possible chemotherapy agent to treat disseminated ovarian cancer.
- ^{79}Kr is a positron emitter with a maximum energy of 604 keV (6.8% of intensity). It decays with a half life of 34.92 h in ^{79}Br (stable) and is produced according to the thermal neutron capture of ^{78}Kr . The target cannot be natural Krypton, since the natural abundance of the mother isotope is 0.325 wt % (refer to [40]); for that, the target must be enriched. In the case of study, the enrichment is 99 wt%. ^{79}Kr is used mainly in PET exams for assessing cardiovascular and lung function.
 - ^{89}Sr is a pure β^- emitter for which the maximum energy is 15 MeV. It decays in ^{89}Y (stable) with a half-life of 50.5 days and is produced mainly by (n, γ) reaction. Other possible reactions for producing it are fission of ^{235}U or fast neutron capture of ^{89}Y ((n,p) reaction). It is used in both industrial and medical applications:
 - industrial gauging;
 - bone cancer pain palliation;
 - cellular dosimetry;
 - treatment of prostate cancer;
 - treatment of multiple myeloma;
 - osteoblastic therapy;
 - potential agent for treatment of bone metastases from prostate and breast cancer.
 - ^{90}Y is a pure β^- emitter with maximum energy of 2.28 MeV. It decays in ^{90}Zr (stable) with a $T_{1/2}$ of 64.00 hours. It is produced mainly by thermal neutron capture, but it can be produced also by fast neutron capture from ^{90}Zr ((n,p) reaction) and, sometimes, it can result useful producing it in form of radionuclide generator by fission of ^{235}U ($^{90}\text{Sr}/^{90}\text{Y}$ generator). It is used mainly in medical applications:

- internal radiation therapy of liver cancer;
 - monoclonal antibodies;
 - Hodgkins disease and hepatoma;
 - cellular dosimetry;
 - treating rheumatoid arthritis;
 - treating breast cancer;
 - treatment of gastrointestinal adenocarcinomas.
- ^{117m}Sn , being a radionuclide in metastable state, decays by isomeric transition ($T_{1/2}=13.6$ days) emitting gammas with energy of 158.56 keV (86.4% intensity). It is produced by Inelastic Neutron Scattering reaction. It is most used in medicine for bone cancer pain relief, treatment of finger joints inflammation and rheumatoid arthritis.
 - ^{131}I emits β^- with a maximum energy of 606.3 keV (89.6% intensity). To its decay in ^{131}Xe (stable), the emission of several gamma rays is associated; the most probable (81.5% intensity) has a maximum energy of 364.5 keV. It has a half-life of 8.0252 days. It is produced by both fission of ^{235}U and by thermal neutron capture in Tellurium ($^{130}\text{Te}(n,\gamma)^{131}\text{Te} \rightarrow ^{131}\text{I}$). The applications in which it is used are various:
 - in isotope hydrology and leak detection as tracer;
 - lymphoid tissue tumour/hyperthyroidism treatment;
 - treatment of carcinoma of the thyroid;
 - antibody labelling;
 - brain biochemistry in mental illness;
 - kidney agent;
 - [SPECT](#) imaging;
 - cellular dosimetry;
 - treatment of prostate cancer;

- internal radiation therapy;
- ...
- ^{133}Xe decays in ^{133}Cs with a half-life of 5.2475 days emitting β^- with maximum energy of 346.4 keV. Several gammas are associated to it, and the most probable (intensity of 38%) has a maximum energy of 80.997 keV. It is an indirect product of fission of ^{235}U , since it results from the decay of ^{133}I . Its applications are mainly related with medicine:
 - lung imaging;
 - regional cerebral blood flow;
 - liver imaging;
 - [SPECT](#) imaging of brain;
 - lung scanning;
 - lesion detection.
- ^{140}La decays in ^{140}Ce (stable) with a half-life of 40.29 hours emitting several β^- of which three are the most probable: maximum energy of 1.35 MeV (43.9% intensity), 1.68 MeV (20.2%) and 1.24 MeV (11.05%). Many γ are related with its decay too; the most probable of those is the one with maximum energy equal to 1.60 MeV (95.4%). It is produced mainly by thermal capture, but it is also possible to separating it from the ^{140}Ba produced by fission of ^{235}U (in this case, we have a radionuclide generator). It is used mainly as radiotracer: in blast furnaces for determining the resident times and to quantify yields to measure the furnace performance.
- ^{153}Sm decays in ^{153}Eu (stable) emitting several β^- and γ , of which the most probable have a maximum energy of: $\beta_{max} = 704.4$ keV (49.4%) and 807.6 keV (18.4%); $\gamma_{max} = 103$ keV (29.25%) and 69.67 keV (4.73%). It has a half-life of 46.27 hours. It is produced mainly by thermal neutron reactions but alternative routes can be (n,p) and (n, α) fast neutron reactions. Its uses are purely medical:

- cancer diagnostic and treatment;
 - monoclonal antibodies;
 - bone cancer pain relief;
 - treatment of leukaemia.
- ^{177}Lu decays in ^{177}Hf (stable) emitting a β^- of maximum energy of 498.3 keV (79.4%) and several γ of which the most probable have maximum energy of 208 keV (10.36%) and 133 keV (6.17%). Its $T_{1/2}$ is of 6.647 days. Its production is carried out mainly by thermal capture reaction. It is used mainly in the medicine for heart disease treatment (restenosis therapy) and cancer therapy.
 - $^{188}\text{W}/^{188}\text{Re}$ is a generator produced by a double thermal capture reaction starting from a target made of Tungsten highly enriched in the isotope ^{186}W . As in case of $^{99}\text{Mo}/^{99m}\text{Tc}$ generator, the product of interest is the daughter nuclide that can be stored in an alumina-based system for being eluted from it when necessary. ^{188}Re decays in ^{188}Os (stable) with a half-life of 17.00 hours, emitting several β^- and γ , the maximum energy of which is: β_{max}^- is equal to 2.12 MeV (70%) and 1.97 MeV (26.3%) and γ_{max} to 155.04 keV (15.61%). Its uses are:
 - bone pain palliation (in form of ^{188}Re -HEDP and ^{188}Re -DMSA);
 - endovascular radiation therapy (in form of ^{188}Re -perrhenate and ^{188}Re -MAG3);
 - tumour therapy (^{188}Re -peptides);
 - Endoradiotherapy of tumours;
 - marrow ablation prior to stem cell rescue.
 - ^{192}Ir emits different β^- and γ in decaying ($T_{1/2} = 73.82$ d) in ^{192}Pt (stable), the maximum energy of which are: β_{max}^- is equal to 675.1 keV (48%) and 538.8 keV (41.43%) and γ is 316.51 keV (82.71%) and 468.07 keV (47.81%). It is produced by (n,γ) reaction. It find applications in both industrial and medical field:

- in gamma-radiography to locate flaws in metal components;
- treatment of prostate, brain, breast, gynaecological cancers.
- ^{203}Hg , in decaying in ^{203}Tl (stable) with $T_{1/2}$ of 46.59 days, emits β^- of maximum energy of 212.9 keV and γ of maximum energy of 279.2 keV (81.56%). It is produced by thermal neutron capture. Its major use is as radiotracer in industry and environment.

Table 4.1 lists all the radioisotopes mentioned above together with the route of production and the target used in BR2:

Table 4.1: Radioisotopes produced, nowadays, in BR2. For the data refer to [31] and only for those marked with * to [32].

Radioisotope	Reaction	Used Target
^{32}P	(n, γ)	20 mg of ^{31}P
$^{79}\text{Kr}^*$	(n, γ)	5 ml of Kr (enr.: 99%)
^{89}Sr	(n, γ)	6 g of SrCO_3
^{90}Y	(n, γ)	15 mg of ^{89}Y
^{117m}Sn	(n,n' γ)	260.30 g of ^{117}Sn (enr.: 92%)
^{131}Ir	(n,f)	4 g of ^{235}U
^{133}Xe	(n,f)	4 g of ^{235}U
$^{140}\text{La}^*$	(n, γ)	21.2 mg of La_2O_3 1 g of $\text{La}(\text{C}_2\text{H}_3\text{O}_2)_3$
^{153}Sm	(n, γ)	3 mg of Sm_2O_3
^{177}Lu	(n, γ)	300 μg of ^{176}Lu (enr.: 98%)
$^{188}\text{W}/^{188}\text{Re}$	double (n, γ)	8 g of W (enr.: 97.5%)
^{192}Ir	(n, γ)	5 g of Ir
^{203}Hg	(n, γ)	5.30 g of Hg

Furthermore, the production of other three radioisotopes has been chosen to be evaluated:

- ^{24}Na that decays β^- in ^{24}Mg (stable) with a half-life of 14.96 hours. It emits β^- with maximum energy of 1.39 MeV and γ of maximum energy of 2.75 MeV. Its production by (n, α) fast neutron reaction, starting from aluminium (^{27}Al) target, is evaluated. This radionuclide is used mainly as radiotracer in industry.
- ^{95}Zr , that can be recovered from the same target used for ^{99}Mo production as well as ^{140}Ba . It has a $T_{1/2}$ of 64.03 days and emits two betas and two gammas, which namely have maximum energy of 366.9 (54.46%), 399.4 (44.34%) keV and 756.73 (54.38%), 724.19 (44.27%) keV. This nuclide and its radioactive daughter ^{95}Nb find several applications in medicine.
- $^{191}\text{Os}/^{191m}\text{Ir}$ generator. ^{191m}Ir decays by isomeric transition emitting a gamma of 129.43 keV with a very short half-life, 4.90 s. That makes of it an important nuclide for medicine, in which it is used especially for angiocardiology. Of course, this cannot be produced except that in generator form. The route of production of ^{191}Os is thermal neutron capture. Then, it is loaded on an anion-exchange column and ^{191m}Ir is eluted with normal saline.

Eventually, the complete list of the radioisotopes considered in the calculations together with the routes of production is given in Table 4.2. The amount of target materials are considered: equal to those used in BR2 for the same reactions (look Table 4.1); equal or proportional to values provided by [38] (values marked with ¹ in the table); equal or proportional to the mass occupying the considered volume in standard conditions (values with ²).

Table 4.2: List of the Radioisotopes to be produced from the reported targets according to the specified routes.

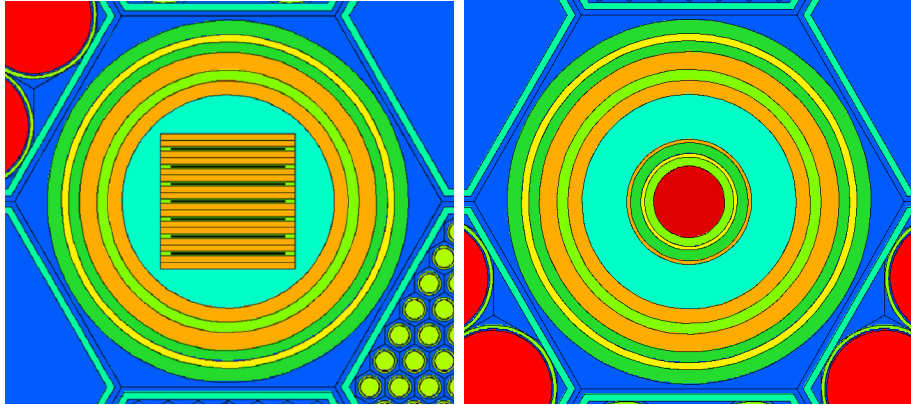
RADIO-ISOTOPE	ROUTE OF PRODUCTION	USED TARGET
^{24}Na	$^{27}\text{Al}(\text{n},\alpha)^{24}\text{Na}$	95.43 g of Al ²
^{32}P	$^{31}\text{P}(\text{n},\gamma)^{32}\text{P}$	20 mg of P
	$^{32}\text{S}(\text{n},\text{p})^{32}\text{P}$	126 g of S ¹
^{79}Kr	$^{78}\text{Kr}(\text{n},\gamma)^{79}\text{Kr}$	34.5 mg of Kr (enr.: 99%)
^{89}Sr	$^{235}\text{U}(\text{n},\text{f})^{89}\text{Sr}$	6.28 g of UAl _x (enr.: 90%)
	$^{88}\text{Sr}(\text{n},\gamma)^{89}\text{Sr}$	6 g of SrCO ₃
	$^{89}\text{Y}(\text{n},\text{p})^{89}\text{Sr}$	147.03 g of Y ₂ O ₃ ¹
^{90}Y	$^{235}\text{U}(\text{n},\text{f})^{90}\text{Sr}/^{90}\text{Y}$	6.28 g of UAl _x (enr.: 90%)
	$^{89}\text{Y}(\text{n},\gamma)^{90}\text{Y}$	15 mg of Y
	$^{90}\text{Zr}(\text{n},\text{p})^{90}\text{Y}$	200.75 g of ZrO ₂ ²
^{95}Zr	$^{235}\text{U}(\text{n},\text{f})^{95}\text{Zr}$	6.28 g of UAl _x (enr.: 90%)
$^{117\text{m}}\text{Sn}$	$^{117}\text{Sn}(\text{n},\text{n}')^{117\text{m}}\text{Sn}$	260.30 g of Sn (enr.: 92%) ²
^{131}I	$^{235}\text{U}(\text{n},\text{f})^{131}\text{I}$	6.28 g of UAl _x (enr.: 90%)
	$^{130}\text{Te}(\text{n}, \gamma)^{131}\text{Te} \rightarrow ^{131}\text{I}$	45.09 g of Te ¹
	$^{130}\text{Te}(\text{n}, \gamma)^{131}\text{Te} \rightarrow ^{131}\text{I}$	37.14 g of TeO ₂ ¹
^{133}Xe	$^{235}\text{U}(\text{n},\text{f})^{133}\text{I} \rightarrow ^{133}\text{Xe}$	6.28 g of UAl _x (enr.: 90%)
^{140}La	$^{235}\text{U}(\text{n},\text{f})^{140}\text{Ba}/^{140}\text{La}$	6.28 g of UAl _x (enr.: 90%)
	$^{139}\text{La}(\text{n},\gamma)^{140}\text{La}$	21.2 mg of La ₂ O ₃
	$^{139}\text{La}(\text{n},\gamma)^{140}\text{La}$	1 g of La(C ₂ H ₃ O ₂) ₃
^{153}Sm	$^{152}\text{Sm}(\text{n},\gamma)^{153}\text{Sm}$	3 mg of Sm ₂ O ₃
	$^{153}\text{Eu}(\text{n},\text{p})^{153}\text{Sm}$	262.24 g of Eu ₂ O ₂ ²
	$^{156}\text{Gd}(\text{n},\alpha)^{153}\text{Sm}$	279.2 g of Gd ²
^{177}Lu	$^{176}\text{Lu}(\text{n},\gamma)^{177}\text{Lu}$	300 mg of Lu (enr.:98%)
^{188}Re	$^{186}\text{W}(\text{n},\gamma)^{187}\text{W}(\text{n},\gamma)^{188}\text{W}/^{188}\text{Re}$	8 g of W (enr.: 97.5 %)
$^{191\text{m}}\text{Ir}$	$^{190}\text{Os}(\text{n},\gamma)^{191}\text{Os}/^{191\text{m}}\text{Ir}$	30 mg of Os (enr.: 96%) ¹
^{192}Ir	$^{191}\text{Ir}(\text{n},\gamma)^{192}\text{Ir}$	2.5 g of Ir
	$^{191}\text{Ir}(\text{n},\gamma)^{192}\text{Ir}$	2.5 g of Ir (enr.: 80%)
^{203}Hg	$^{202}\text{Hg}(\text{n},\gamma)^{203}\text{Hg}$	5.30 g of Hg

4.2 The evaluated Performances

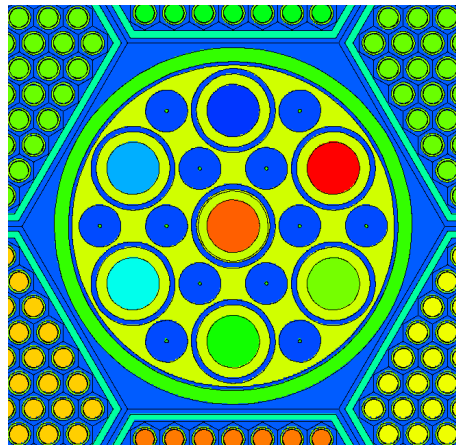
In Section 3.5 we have already discussed about the MYRRHA irradiation rigs; three of them are considered in this work for the radioisotopes production. Figure 4.1 and 4.2 depict the cross-sections of those rigs; in particular Figure 4.1(c) and 4.2(c) show the cross sections of an inner IPS already loaded with targets for radioisotope production. With the exception of the one used for the Molybdenum production by fission of ^{235}U , the other two rigs are loaded with different targets, necessary for the desired reactions to occur. Irradiating the targets, we see the building up of the amount of the radioisotopes until saturation is reached. Saturation represents the maximum amount achievable from a certain target: in the evolution of the radioisotope, we see that it proceeds asymptotically after that a maximum value is reached. This trend is kept until when the decay starts to become predominant in comparison with the production. The reason of this phenomenon is that all the target nuclei have been transmuted, thus there are no more available for the reaction. This means that it results useless irradiating the targets for very long time: each irradiation cycle can be calibrated on the used target. Usually, the irradiation is not longer than three half-lives, for which it is reached a value of about 88% of the saturation. Considering the case of Molybdenum-99 production, a cycle of 150 hours is foreseen in the BR2 and in MYRRHA of 168 hours is studied. The trend of its evolution in time is shown in Figure 4.3: as it is easy to see, irradiation is stopped before saturation occurs.

As already specified in the previous chapter, we run calculations using a probabilistic code; in order to have good statistics, we have to specify an appropriate number of both histories to follow and cycles to run, even more so because we deal with a criticality problem. In the studied simulations these options are defined in the *KCODE card*:

- the number of histories, that is the number of neutrons to follow, is set equal to 1.0E+05, to not have too long computation times.
- the initial k_{Eff} guess, set equal to 1.
- the total number of cycles is chosen to be equal to 300, of which the

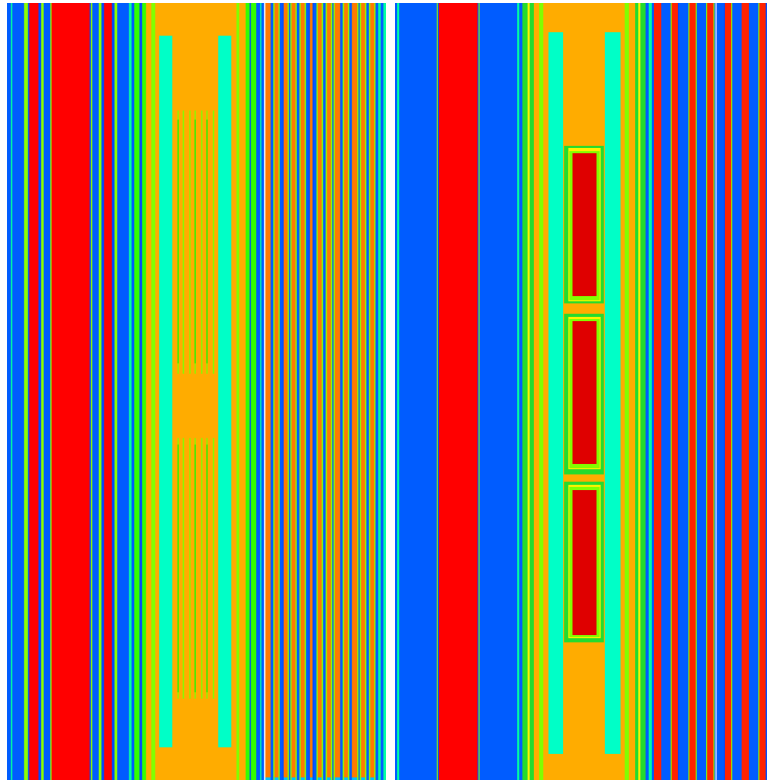


(a) Used rig in production of ^{99}Mo . (b) Used rig in performing radiative capture reactions.

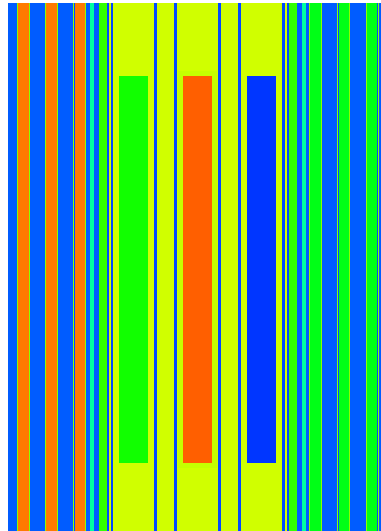


(c) Inner IPS rig used for performing fast reactions.

Figure 4.1: MYRRHA Irradiation Rigs: cross-section.



(a) Used rig in production of ^{99}Mo . (b) Used rig in performing radiative capture reactions.



(c) Inner IPS rig used for performing fast reactions.

Figure 4.2: MYRRHA Irradiation Rigs: transversal cross-section.

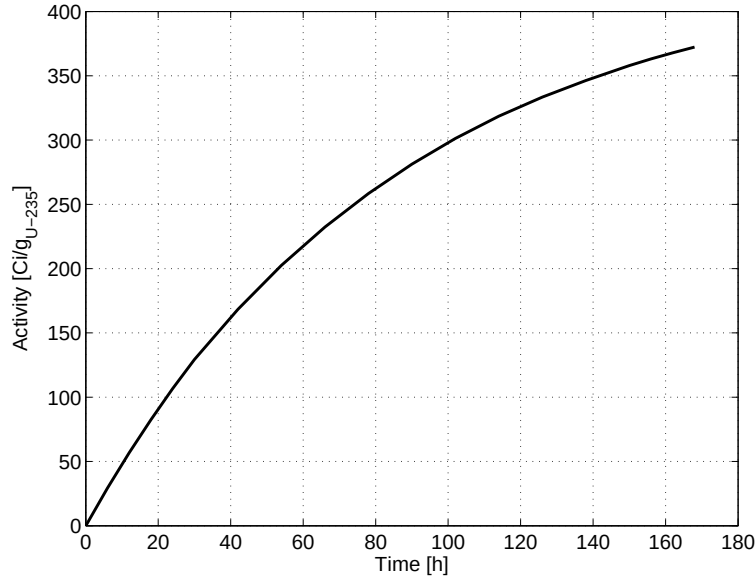


Figure 4.3: ^{99}Mo production with an Irradiation Cycle of 168 hours in MYRRHA.

first 50 will be skipped to let the neutrons spread across the core and to avoid the recording of useless data related to localized interactions, and the other 250 will constitute the active cycles.

All the neutrons, prompt and delayed, are considered too. That presented here is the neutronics modelled for all the simulations except the one used for the production of ^{99}Mo by fission of ^{235}U . In this case the number of histories is taken equal to $1.0\text{E}+06$ and also the number of active cycles is increased of 50 (300 active cycles). Such simulation can be run in reasonable times only by the use of many CPUs: in deed, it was run using 64 CPUs (4 nodes in the cluster) while the others using only 16 CPUs (1 node). At last, the temperature at which the data, used for running the calculations, are taken from the libraries is 300 K (0.0253 eV).

In the following, MYRRHA production capabilities are presented considering the reactions listed in Table 4.2.

4.2.1 Fission Products

The irradiation rig for ^{99}Mo production is, nowadays, under design. It foresees an *IPS* loaded with a Beryllium basket containing two sets of 7 plates of UAl_x each, highly enriched (90%) in ^{235}U . The plate contains about 4 g of ^{235}U , for a total of 56 g in 14 plates. The irradiation cycle, as previously said, is of 168 hours (7 days) after that the targets are unloaded from the *IPS* and stored for up to 24 hours for allowing the decay in gamma heating and short-lived fission products. Figure 3.5 shows four *IPS* dedicated to the Molybdenum production in the periphery of the core. The simulation is run in this configuration but the presented results are related just to one of them, taking in consideration the symmetry of the reactor and so of the problem itself. For the simulation the reactor is considered at the startup with a critical fresh core and is operated at nominal power, 100 MW. For the ALEPH simulation, it is decided to perform a constant flux irradiation (Irradiation at constant Flux (*IRF*)) with the steady-state source strength, S , equal to $8.64\text{E}+18$.

With the designed configuration described in the previous chapter, it is possible to have in the desired location a flux of about $1.3\text{E}+15 \text{ n}/(\text{cm}^2\text{s})$ and a spectrum-averaged cross section of 88.5919 b . Even if this last value is an order of magnitude smaller than the value of the fission cross-section related to a Maxwellian distribution (570.79 b [39]), the obtained results are really positive. For an irradiation of 168 hours it is possible to achieve about 365 Ci per gram of ^{235}U and if we refer to an irradiation cycle of 150 hours as the one performed in BR2, it is possible to compare this result with the amount nowadays produced: the simulation shows that about $350 \text{ Ci/g}_{^{235}\text{U}}$ can be produced in MYRRHA while in BR2, from the same targets they obtain $250 \text{ Ci/g}_{^{235}\text{U}}$. This means that the moderation provided by the designed system to the fast neutron in the reactor is good for the production of this radioisotope.

As well known, the fission of Uranium does not correspond to only one product; there is a long list of products from this reaction, each of them created with a certain probability that is represented by the so-called *Inde-*

pendent Fission Yield. This probability is very small because the number of fission products is huge; but, since many of these radioisotopes have a very short half-life (of the order of milliseconds or less), it is possible to refer to the so-called *Cumulative Fission Yield*: it is given by the sum of all the independent fission yields of all the precursors of one considered isotope. For example, the independent fission yield of ^{99}Mo is 1.79E-05, and the cumulative, given by the sum of all the independent fission yields of those created radioisotopes with $A=99$ and $Z<42$, is 6.15%. So, since from the same Uranium target is possible to obtain many different radioelements and the chemical processes to separate them are easier than of those used for separating a radioisotope from its stable isotope (as in the case of (n,γ) reactions), it is investigated which is the amount of certain radioisotopes created as byproducts in the process of production of Molybdenum. The selected radioisotopes in this case are: ^{89}Sr , ^{90}Sr (as parent nuclide of ^{90}Y), ^{95}Zr , ^{131}I , ^{133}Xe and ^{140}Ba (as parent nuclide of ^{140}La). In Table 4.3, the Independent and Cumulative Fission yields for these radioisotopes, evaluated with ALEPH2 for the simulation in exam, are listed.

Table 4.3: Independent and Cumulative Fission Yields of ^{89}Sr , ^{90}Sr (as parent nuclide of ^{90}Y), ^{95}Zr , ^{131}I , ^{133}Xe and ^{140}Ba (as parent nuclide of ^{140}La) evaluated with ALEPH2.

RADIOISOTOPE	INDEPENDENT FISSION YIELD	CUMULATIVE FISSION YIELD [%]
^{89}Sr	2.19E-05	4.76
$^{90}\text{Sr}/^{90}\text{Y}$	3.13E-04	5.81
^{95}Zr	3.52E-04	6.53
^{131}I	1.48E-05	2.86
$^{133}\text{I} \rightarrow ^{133}\text{Xe}$	1.54E-03	6.49
$^{140}\text{Ba}/^{140}\text{La}$	2.93E-03	6.31

Referring, as before, to an irradiation cycle of 150 hours, the produced activities by fission of Uranium-235 are listed in Table 4.4. As expected, the activity of ^{133}Xe is high and comparable with that of ^{99}Mo ; the activities of ^{131}I and ^{140}Ba are high too, thus, it can be profitable to extract them from the target; the activities of ^{89}Sr and ^{95}Zr are smaller but, for the simplicity

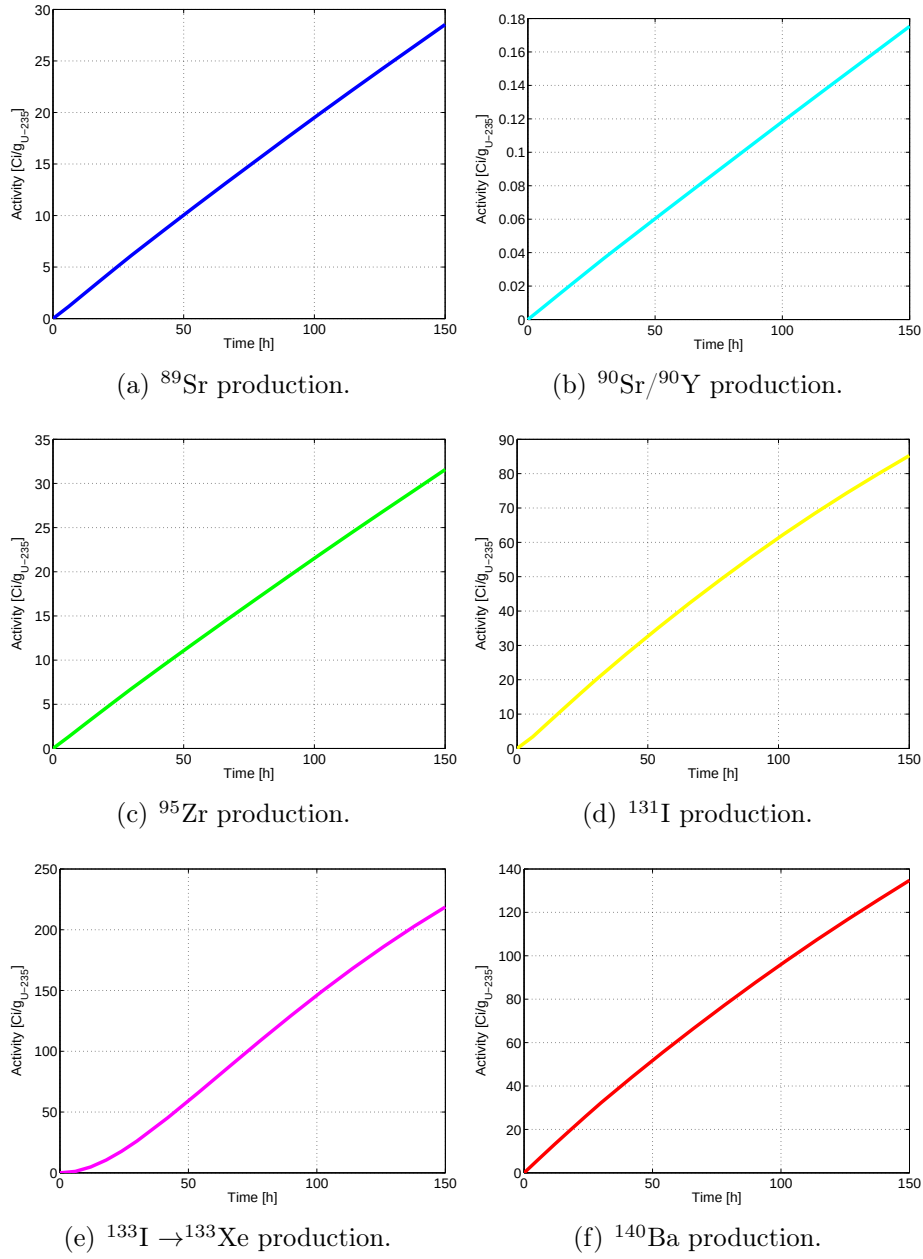


Figure 4.4: Evolution of the considered fission products for an irradiation time of 150 h.

Table 4.4: Produced Activities of ^{89}Sr , ^{90}Sr (as parent nuclide of ^{90}Y), ^{95}Zr , ^{131}I , ^{133}Xe and ^{140}Ba (as parent nuclide of ^{140}La).

RADIOISOTOPE	ACTIVITY [Ci]	ACTIVITY [Ci/g ^{235}U]
^{89}Sr	1597.813	28.532
$^{90}\text{Sr}/^{90}\text{Y}$	9.819	0.175
^{95}Zr	1767.665	31.565
^{131}I	4773.118	85.234
$^{133}\text{I} \rightarrow ^{133}\text{Xe}$	12248.233	218.719
$^{140}\text{Ba}/^{140}\text{La}$	7546.525	134.760

in separating them, their recovery from the target can be considered too; the activity of the ^{90}Sr , instead, results too low. The processes for extracting Iodine and Xenon from the target are well known and already performed, as mentioned in Section 2.2.3. Finally, Figure 4.4 shows the evolution during the irradiation of these radioisotopes. As we can see in Figure 4.4(e), the trend of the evolution of Xenon is not equal to the others showing at the beginning a change in the curvature; this is due to the fact that, contrarily to the other nuclides, it is produced indirectly by fission, that is, it results from the decay of the ^{133}I ($T_{1/2} = 20.8$ h). Inevitably, since the cycle is calibrated on the production of the Molybdenum, we are not at the same grade of saturation but, nevertheless, it is possible to extract these radionuclides from the target instead of treating them as waste: in this way it is possible to reduce the amount of waste and to take advantage of the produced amounts for other purposes.

4.2.2 Radiative Capture Reactions

The irradiation rig designed for the production of ^{227}Ac and ^{228}Th from targets of ^{226}Ra is used in this work for the production of several others radioisotopes. The moderation provided by the Be and the water guarantees the occurrence of the reactions with good yields. The considered thermal reactions, as previously mentioned, are those already performed in the BR2 for the production of ^{32}P , ^{79}Kr , ^{89}Sr , ^{90}Y , ^{140}La , ^{153}Sm , ^{177}Lu , $^{188}\text{W}/^{188}\text{Re}$, ^{192}Ir and ^{203}Hg ; moreover, the production of ^{131}I , from two targets of Tellurium in

natural and oxide form, and $^{191}\text{Os}/^{191m}\text{Ir}$ are investigated. As already specified, the used targets are either the same of those used nowadays in BR2 or equivalent or proportional to those provided by different documents (refer to Table 4.2). The targets are inserted in AlMg_3 vials or in Quartz (SiO_2) ampoules, which, in turn, are loaded inside the Beryllium basket inside the *IPS*. As well as in the BR2, the quartz ampoules are used for the ^{89}Sr , ^{152}Sm and ^{202}Hg targets. In Figure 4.2(b), we have seen that in the Be basket, three vials can be hosted. So, taking advantage of this for avoiding a big number of simulations to be run, three different targets are inserted inside the mentioned vials; moreover, the other *IPS*, which is used for the production of Ac and Th without the use of a Be basket, can be loaded in the same way of the one just said in order to have the possibility of running only a simulation for 6 different reactions. The neutron flux in the cells is affected by the different positions, but not in a marked manner for our purposes. Only in the cases of $^{188}\text{W}/^{188}\text{Re}$ and ^{192}Ir production, the targets are modelled as rings piled in an AlMg_3 capsule around a cylinder for keeping them in position. Since we have to perform the production of ^{192}Ir from both natural and enriched targets, in order to reduce the computations, the two rings to insert in the capsule are different: one is composed by natural Iridium and the other by ^{191}Ir enriched at 80%. In this case, each *IPS* is loaded with a capsule filled with one of these targets. The results, in the case of W, are presented as the sum of the activities produced per each ring. Figure 4.5 shows how the capsule for W production is made.



Figure 4.5: AlMg_3 capsule loaded with 5 rings.

In the simulation, MYRRHA is considered to be at the startup with critical fresh core and operated at 88 MW. Even in this case, we set a constant

flux irradiation in the simulation with S equal to $7.60\text{E}+18$, value determined again with Eq.3.2 for 88 MW. In the following tables, the comparison between the irradiation time needed in BR2 (operated at 55-60 MW) and in MYRRHA for obtaining a certain Targeted Activity in terms of Curie per gram of target isotope are shown. Table 4.5 shows that in producing the listed radioisotopes, MYRRHA is more efficient than BR2 requiring less time for achieving the targeted activity. In Table 4.6, it is shown that, in performing the listed reactions, MYRRHA achieves the results of the BR2 spending longer times. In Table 4.7, the values of the activities achieved in MYRRHA for the listed reactions are presented: in this case the targeted activities are not reached. Of course, the reason is due to the fact that the considered reactions involve thermal neutrons; MYRRHA, unlike BR2, is a fast spectrum reactor: for this reason we have to provide inside the IPS moderation in order to allow reactions to occur, otherwise their yields would be really low. Comparing the values of the spectrum average neutron cross sections for the considered reactions in case of Maxwellian neutron distribution and in our case, it is possible to better understand the differences in the results. This comparison is meant as a comparison among neutron distributions to understand how far we are in MYRRHA from having a thermal (Maxwellian) flux, that is how good are the designed systems in thermalizing neutrons. Table 4.8 shows that the moderation system allows to obtain for certain reactions values of the cross-sections comparable with those related to the Maxwellian distribution: thus, the obtained activities are comparable with those produced in BR2, which is characterized by a thermal spectrum. For the last reactions listed in the table, we see that the average cross sections are one order of magnitude smaller than those related to the Maxwellian: this is the reason why we cannot reach the desired results.

Other thermal reactions, not performed in BR2, have been investigated: Table 4.9 resumes the obtained results. These results can be compared with those presented in the relative sections in [38], from which the specification

¹referred to natural Ir.

²referred to highly enriched Ir.

³referred to La_2O_3 target.

⁴referred to $\text{La}(\text{C}_2\text{H}_3\text{O}_2)_3$ target.

Table 4.5: Comparison between the irradiation time needed in BR2 and MYRRHA for producing a certain Targeted Activity: smaller times are required in MYRRHA.

REACTION	TARGETED ACTIVITY [Ci/g]	IRRADIATION TIME BR2	IRRADIATION TIME MYRRHA
$^{152}\text{Sm}(n,\gamma)^{153}\text{Sm}$	~ 4500	4 d	1.39 d
$^{186}\text{W double}(n,\gamma)^{188}\text{W}$	~ 1.1	21 d	19 d
$^{191}\text{Ir}(n,\gamma)^{192}\text{Ir}$	~ 750 ¹	21 d	5.17 d
	~ 1500 ²	21 d	17.06 d
$^{202}\text{Hg}(n,\gamma)^{203}\text{Hg}$	~ 0.131	7 d	5.28 h

Table 4.6: Comparison between the irradiation time needed in BR2 and MYRRHA for producing a certain Targeted Activity: longer times are required in MYRRHA.

REACTION	TARGETED ACTIVITY [Ci/g]	IRRADIATION TIME BR2	IRRADIATION TIME MYRRHA
$^{78}\text{Kr}(n,\gamma)^{79}\text{Kr}$	~ 58.54	8.5 h	15.38 h
$^{88}\text{Sr}(n,\gamma)^{89}\text{Sr}$	~ 0.3	21 d	38 d
$^{139}\text{La}(n,\gamma)^{140}\text{La}$	~ 27.683 ³	6.2 h	12.18 d
	~ 4.554 ⁴	0.8 h	1.85 h

Table 4.7: Comparison between the irradiation time needed in BR2 and MYRRHA for producing a certain Targeted Activity: targeted activity not achieved in MYRRHA. Last column presents the values of Activities reached in MYRRHA for the same irradiation times of BR2.

REACTION	TARGETED ACTIVITY [Ci/g]	IRRADIATION TIME BR2	ACTIVITY [Ci/g] MYRRHA
$^{31}\text{P}(n,\gamma)^{32}\text{P}$	~ 50	21 d	~ 5.81
$^{89}\text{Y}(n,\gamma)^{90}\text{Y}$	~ 60	9 d	~ 2.31
$^{176}\text{Lu}(n,\gamma)^{177}\text{Lu}$	~ 28000	7 d	~ 17921

Table 4.8: Comparison between the neutron cross sections averaged over a Maxwellian Spectrum ([39]) and those averaged over the spectrum evaluated with ALEPH2 in the considered cell in the simulation. In the last column, even the cell averaged total neutron flux is provided.

REACTION	MAXWELLIAN CROSS SECTION [b]	CELL AVERAGED CROSS SECTION [b]	CELL AVERAGED TOTAL FLUX [n/(cm ² s)]
¹⁵² Sm(n,γ) ¹⁵³ Sm	206.6	138.797	6.22E+14
¹⁸⁶ W(n,γ) ¹⁸⁷ W	39.499	16.726	7.24E+14
¹⁹¹ Ir(n,γ) ¹⁹² Ir	950.72	188.475	6.81E+14
²⁰² Hg(n,γ) ²⁰³ Hg	4.9531	0.7767	6.28E+14
⁷⁸ Kr(n,γ) ⁷⁹ Kr	4.8444	1.7193	6.20E+14
⁸⁸ Sr(n,γ) ⁸⁹ Sr	0.0058	0.0014	7.05E+14
¹³⁹ La(n,γ) ¹⁴⁰ La	9.0336	1.7984	6.50E+14
³¹ P(n,γ) ³² P	0.1662	0.0284	6.47E+14
⁸⁹ Y(n,γ) ⁹⁰ Y	1.278	0.2294	6.16E+14
¹⁷⁶ Lu(n,γ) ¹⁷⁷ Lu	3593.7	707.607	6.66E+14

Table 4.9: Achieved activities performing the reactions listed in the first column in the irradiation rig of MYRRHA foreseen for the production of ²²⁷Ac and ²²⁸Th.

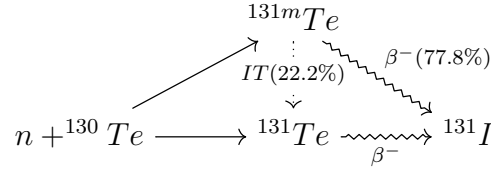
REACTION	IRRADIATION TIME [d]	ACTIVITY [Ci]	ACTIVITY [Ci/g]
¹³⁰ Te(n,γ) ¹³¹ Te (→ ¹³¹ I)	5	~1.468 ⁵ ~1.191 ⁶	~4.259 ~4.320
¹⁹⁰ Os(n,γ) ¹⁹¹ Os (/ ^{191m} Ir)	22	~2.435	~84.532

about the target compositions and the route of production have been taken.

The production of Iodine-131 from Tellurium-130 is a good alternative to the fission of U-235 usually performed. In limiting the uses of HEU, the probabilities of nuclear proliferation are reduced too: thus, even in terms of safeguards, performing this route results very important, being ¹³¹I one of the most used radioisotope in medicine. In this case, radioiodine is not directly produced because it is the result of the decay of the ¹³¹Te and its isomer; the one produced cannot be considered a radionuclide generator since the ¹³¹Te and ^{131m}Te half-lives, namely 25 minutes and 33.25 hours, are shorter than that of the Iodine (8.0252 days). Inevitably, the produced activities are given in terms of ¹³¹Te and not ¹³¹I.

⁵referred to natural Te target.

⁶referred to TeO₂ target.



Regarding the case of production from targets of natural Tellurium, the cited Manual [38] reports as typical yield 300 mCi/g_{130Te} after 3 weeks of irradiation in the Dhruva reactor in BARC complex in Mumbai (India) at a flux of 10^{14} n/(cm² s). The obtained result for the simulation in exam gives a positive confirmation: even if the moderation provided to the neutron is not enough to have values of the cross-section averaged over the spectrum comparable to the case of Maxwellian flux, that is, the value of σ_c that we have in MYRRHA (0.0523 b) is one order less than that averaged over the Maxwellian one (0.28998 b), thanks to the total flux of $6.80 \cdot 10^{14}$ n/(cm² s), it is possible to produce satisfying amounts. Even in case of using targets made of TeO₂, the comparison of the obtained result with the one presented in the manual is positive: there, for an irradiation cycle of 6 days with a flux of $1.1 \cdot 10^{14}$ n/(cm² s) they obtain 589.6 mCi/g_{130Te}. The values of the cross-section and of the flux in this case are similar to the previous case since the targets are loaded close to each other in the Be basket; we have 0.0486 b and $6.41 \cdot 10^{14}$ n/(cm² s). The two different ways of production are investigated since, dealing with two different targets, especially chemically speaking, the separation treatments are different as well as the final products at the end of the processes. Of course, at EOI, we obtain similar results in terms of activity per gram of target atom of ¹³⁰Te but we have to consider, moreover, that the produced activity is different because the amount of ¹³⁰Te in the target is different: 66.2057 Ci with natural Tellurium and 44.2266 Ci with Tellurium oxide.

For the production of ¹⁹¹Os, in the same Manual the calculated specific activity values at various thermal neutron flux are illustrated; we see that saturation is reached even in these cases for about 22 days of irradiation. Taking the case of irradiation at a thermal flux of $2.2 \cdot 10^{14}$ n/(cm² s), the yield is of about 140 Ci/g_{190Os}. This means that the result obtained in the simulation at a total flux of $6.47 \cdot 10^{14}$ n/(cm² s) and shown in the Table 4.9

is positive since the difference is small.

4.2.3 Fast Reactions

Since MYRRHA is a fast neutron spectrum reactor, it is possible to perform those reactions that present a threshold, in term of cross section, in correspondence of high energy of the neutrons (fast neutrons). The production by fast reactions of the already mentioned ^{32}P , ^{89}Sr , ^{90}Y and ^{153}Sm is investigated together with that of the ^{24}Na and ^{117m}Sn . The targets are loaded in the sub-channels of one of the three inner IPS as shown in Figure 4.1(c). With the only exception of the case of ^{117m}Sn , there are no irradiation cycles to which refer; so, in the simulation, these are taken equal to the operational cycle, which lasts 90 days. Then, the results are elaborated to see which is the appropriate irradiation time. The ^{117m}Sn production is performed in the BR2 and the target is loaded inside a quartz (SiO_2) ampoule instead of an AlMg_3 vial; in the mentioned figure we can notice the ampoule surrounding the target. In the simulation, the vials for the other targets are not distinct from the aluminium slots in which they are hosted. For the simulation, the reactor is considered to be operated in the same way of the previous case. Table 4.10 compare the cross-section values taken as reference from JEFF-3.1.2 library with those obtained in the simulation. As it can be noted, the cross section values for these fast reactions are lower than those of the thermal ones, even for neutrons of energy higher than 1 keV. Obviously, this affects the production of the radioisotopes leading to lower activities. In the BR2, nowadays, ^{117m}Sn is produced followed a cycle of 21 days of irradiation with a fast neutron flux of $6 \cdot 10^{14} \text{ n}/(\text{cm}^2 \text{ s})$; the produced activities go up to 10 Ci per gram of ^{117}Sn . The value of the cross section for neutron energy higher than 318 keV is 0.222 b in BR2. As reported in Table 4.11, in MYRRHA we are able to produce the same activity after only 3 days of irradiation thanks to the higher fast flux. In the same table, the obtained results for the other reactions are listed. As we can see, for some reactions, the results in term of Activity per gram of isotope target are not satisfying being the produced activity very low: this is the case of the production of ^{89}Sr , ^{90}Y and ^{153}Sm

Table 4.10: Comparison among the spectrum averaged cross sections in MYRRHA evaluated with ALEPH2 and those averaged over a fission spectrum (from 1 keV to 20 MeV) taken from [39].

REACTION	< σ > [b]	
	MYRRHA	JEFF-3.1.2
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	2.91E-05	8.9998E-04
$^{32}\text{S}(n,p)^{32}\text{P}$	0.0059	0.0692
$^{89}\text{Y}(n,p)^{89}\text{Sr}$	1.92E-05	2.9942E-4
$^{90}\text{Zr}(n,p)^{90}\text{Y}$	9.79E-06	2.1261E-4
$^{117}\text{Sn}(n,n')^{117m}\text{Sn}$	0.2521	1.2432
$^{153}\text{Eu}(n,p)^{153}\text{Sm}$	8.88E-07	1.5415E-5
$^{156}\text{Gd}(n,\alpha)^{153}\text{Sm}$	6.61E-07	6.2902E-6

from Gadolinium. The productions of ^{32}P and ^{153}Sm are enough satisfying, but still not competitive in terms of both activity and irradiation time with the thermal reactions presented above. The case of producing ^{24}Na from ^{27}Al could result interesting seen the discrete value of achieved activities and the relative short irradiation time. Of course, before to start producing it in this way, the results must be compared with the effective activity produced with the usual thermal reaction and then see if the simplification of the treatment at the end of irradiation justifies this process.

Table 4.11: Achieved activities performing fast reaction in the inner [IPS](#) within MYRRHA core.

R_{REACTION}	TARGET	FLUX [n/(cm² s)]	IRRADIATION TIME [d]	ACTIVITY [Ci]	ACTIVITY [Ci/g]
²⁷ Al(<i>n,α</i>) ²⁴ Na	95.43 g of Al	~2.38E+15	8	~6.763	~0.071
³² S(<i>n,p</i>) ³² P	126 g of S	~2.45E+15	76	~833.459	~6.981
⁸⁹ Y(<i>n,p</i>) ⁸⁹ Sr	147.03 g of Y ₂ O ₃	~2.49E+15	90	~0.720	~6.22E-03
⁹⁰ Zr(<i>n,p</i>) ⁹⁰ Y	200.75 g of ZrO ₂	~2.45E+15	16	~0.400	~5.31E-03
¹¹⁷ Sn(<i>n,n'</i>) ^{117m} Sn	260.30 g of Sn enr. at 92%	~2.36E+15	3	~2747	~10
¹⁵³ Eu(<i>n,p</i>) ¹⁵³ Sm	262.24 g of Eu ₂ O ₃	~2.25E+15	90	~29.918	~0.240
¹⁵⁶ Gd(<i>n,α</i>) ¹⁵³ Sm	279.21 g of Gd	~2.29E+15	32	~9.73E-03	~1.70E-04

Conclusions

The irradiation rigs for the production of ^{99}Mo by fission of ^{235}U and of ^{227}Ac and ^{228}Th from neutron captures of ^{226}Ra are already under design at SCK·CEN in MYRRHA project. In this work, the effective possibility of producing a certain number of radioisotopes taking advantage of the same irradiation rigs just mentioned is evaluated. The capabilities of the designed rigs for MYRRHA are good and shows satisfying results for many other reactions as discussed in Chapter 4.

The possibility of extracting from UAl_x plates other radionuclides as byproducts of the production of ^{99}Mo (^{131}I , ^{133}Xe , ^{140}Ba ...) results advantageous especially considering that otherwise these are treated as waste; since the production of Molybdenum as parent nuclide of ^{99m}Tc is high, in extracting them, the amount of waste and, with it, the impact on the environment can be reduced considerably. This can be seen as *recycling*, so it leads to a better use of the resources in our possession.

The designed systems works well even for a certain number of other reactions considered in this pages. As seen, it will be possible to produce most of the radionuclides nowadays produced by the BR2; for some of them, the obtained results are really positive leading to the possibility of reducing the irradiation time for targeted activity (^{153}Sm , ^{186}W , ^{192}Ir , ^{203}Hg). But not in all the cases we have a positive feedback: this is due to the fact that the involved reactions have higher yields with thermal neutrons; the moderation we provide to the fast neutron within MYRRHA core is not always enough to guarantee good results. In some cases, the high total flux can counterbalance the smaller cross section allowing to achieve rather satisfying results.

For what concerns the other (n,γ) reactions not performed in the BR2, the simulations show good results in comparison to what found in the reference.

Eventually, the possibility of producing radioisotopes using the irradiation rig designed for the material test under fast flux is evaluated. The considered reactions occur with fast neutrons and, in particular, they present a threshold in correspondence of high value of energy ($\sim$ MeV). Anyway, these reactions are characterized by very small cross section values; thus, the yield for the production of a same radionuclide results higher with thermal reactions. The reason why these reactions are investigated is that, using as target an isotope that is chemically different by the one to produce, the processes at the EOI are more efficient and allow to obtain carrier-free radioisotope, that is radioisotope essentially undiluted with its stable isotope. It is obtained one of the best result in this configuration for the production of ^{117m}Sn : thanks to the higher fast flux in comparison with the one of the BR2, even the yield is higher. Other reactions present enough satisfying results but not really competitive with the thermal reactions. Instead, for some of the reactions considered, the cross section values are too small to think about the possibility of using these routes for economical purposes.

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