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Measurement of Gas and Volatile Elements Production Rates in Molten Lead Bismuth Target

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ABSTRACT

Production rates of gas and volatile elements produced following spallation reaction of 1.4 GeV protons on a liquid LBE target have been measured. The experiment was performed at the ISOLDE facility at CERN. These data are of great interest for the developments of targets for accelerator driven systems such as MEGAPIE. Calculations were performed using the Monte Carlo code FLUKA coupled with an evolution code such as ORIHET3. Preliminary results show good comparison with MC data for Hg and for noble gases. For other elements such as I results indicate that only a fraction of the produced isotopes are released.

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1 INTRODUCTION

Some concerns about criticality safety of fast reactors and the fast development of high-intensity proton accelerators has increased the interest in using sub-critical systems, fed by neutrons which are produced in a spallation process maintained by an external proton beam. These ideas have led to the proposal of Accelerator Driven Systems (ADS) (see Appendix B for a summary of abbreviations); in Europe first proposed by C. Rubbia, followed by other innovative concepts [1]. The aim with the ADS is to process high-level waste from nuclear power plants and decommissioned nuclear weapons by partitioning and transmutation of the waste. To make the concept technologically attractive a reduction of the high-level waste radiotoxicity, caused by actinides and long-lived fission products, by a factor of at least one hundred is desirable [2]. This means a reduction in waste storage from the order of 100 000 years to the less than 1000 years [3]. Even if these ideas are realized it will not totally solve the storage problem of nuclear waste, but it will definitely change the perspectives on the nuclear waste management and perhaps also the attitude towards nuclear power plants.

Scientists are today working in different programs to make these concepts work. Paul Scherrer Institut (PSI), in Villigen, Switzerland is one of the institutes participating within the Fifth European Community Framework Program, working on a project called MEGAPIE. The goal of the MEGAPIE project is to validate the target system, therefore playing a key role in the roadmap. [1] The MEGAPIE (MEGAwatt PIlot Experiment) [4] project was launched by the Commisariat à l'Energie Atomique France, Forschungszentrum Karlsruhe Germany, KAERI Korea, DOE United States and the Paul Scherrer Institut. Today six European research institutes and JAERI Japan are taking part of this project. The experiments are planned to start in year 2006 and last for about one year. The purpose is to validate the LBE target and demonstrate its functioning and safe disposal and thus enable for granting of a license. [5]

A Lead Bismuth Eutectic-target (LBE), that is liquid under operation conditions because of its low melting point, is under consideration for ADS. Since a neutron spallation source in ADS must have high intensity, the realistic testing of the target demands a high proton beam power, which in Europe is only possible with the ring cyclotron at PSI. In the neutron spallation source at PSI, called SINQ the liquid Lead Bismuth target will eventually be placed and remain for up to a year of experiments. There the target will be exposed to an intense high-energy proton beam, inducing spallation reactions and releasing of high flux spallation neutrons. There are still issues to be solved, among other things how the "un-wanted" spallation products, having consequences for the neutron balance, the material properties, the final disposal etc, are to be taken care of. [1] Up till a few years ago no experiments were done at all on liquid targets when exposed to high intensity proton beams. [6]

As a result of the spallation reaction in the liquid PbBi target, not only neutrons are produced, but many different nuclides, called residuals, some stable and some radioactive with different radiotoxicity. Many of them are highly volatile and in gaseous form and will diffuse out of the LBE and be collected on the top of the target inside the target

container. The cross sections for many of these materials are unknown, or can be predicted with great uncertainty [7]. Elements important to study for the MEGAPIE project are the highly-volatiles; H, He, N, O, Ne, Cl, Ar, Br, Kr, I, Xe and Hg. Other less volatile elements of interest are Na, P, S, K, Zn, As, Se, Rb, Cd, Te, Cs and Po. There are two main reasons to evaluate these elements. First it is of interest to know the amount of elements produced to be able to handle the gases during operation. Second, it is of great importance to be able to predict the activity induced in the target, due to the radioactive elements produced in the spallation reaction, so safe operation and disposal of the target is possible.

The aim of this project is to calculate the amounts of the most hazardous/problematic volatiles produced in the PbBi-target. The experimental part of the work has been performed at the ISOLDE facility at CERN. The production rate of several gaseous spallation products can be measured with mass spectrometric analysis, available at the ISOLDE facility. The calculated production rates will then be compared with the Monte Carlo code FLUKA.

The method described in this report and used for identifying the volatile isotopes was the off-line γ spectroscopy. During the experiment, measurements were also done with a Faraday Cup and with on-line β spectroscopy. These two methods will just be briefly discussed.

This first experiment and its analysis is an important step for continuing the research on the volatiles in the LBE target. A follow-up experiment will be conducted in fall 2004, where new isotopes will be measured at different beam energy. These results will not be included in the report, but the planning of the follow-up experiment will be commented.

2 THEORY

Spallation is a two-step process where an element is transformed into new lighter fragments. In a spallation target like the MEGAPIE target the activity will change over time, since the proton beam hitting the target is inducing new spallation reactions and the radioactive spallation products will decay through different modes. This will be discussed in the following chapters and finished by introducing the mathematical way of simulating the spallation reaction, using the Monte Carlo method.

2.1 SPALLATION

The most important step to understand what production rates of different radioactive isotope produced in the LBE target are expected is to understand the process of spallation. Spallation is a process where an element is turned into another element by hitting the nucleus with light particles, like protons or neutrons. Important products of the spallation reaction are the neutrons and therefore spallation is used as a neutron source for widely different applications; material science, basic research with neutrons and nuclear waste transmutation in ADS.

To induce the spallation reaction it is necessary to use highly energetic charged particles, like the protons. [9] The incoming protons induce a cascade process where among others nucleons, mesons (1 quark and 1 antiquark) and γ-rays are emitted. The target nucleus is excited and releases a high number of neutrons dependent of the incoming particle energy and type and target material.

In the fission reaction an excess amount of energy of about 200 MeV per usable fission neutron is released and transformed into thermal energy. In the spallation reaction this amount is much smaller, typically about 25 MeV per neutron for a lead target bombarded with a 1 GeV proton beam. The values for spallation vary depending on target and size of the target. Even though the thermal energy is much lower than in a fission reaction, the target still has to be cooled. In addition, in a spallation source like SINQ most of the kinetic energy of the neutrons is imparted to heavy water moderator that slows the neutrons down to thermal energies.

2.1.1 A two-step reaction

The spallation process can be divided into a two-step reaction [1]. In the first step the nucleus is hit by the incoming proton beam, a particle cascade inside the target starts and among others neutrons are emitted. The remaining target nucleus is left in an excited state and start to evaporate different particles or if possible fissions.

The first step, called the Intra-Nuclear Cascade (INC), can be considered as a hard ball collision between incoming proton and a nucleon in a target nucleus. See FIGURE 2.1. An incident proton of 1 GeV has a wavelength of about 1 fm ($\lambda = h/p$), which is equal to the distance between the nucleons in the target nuclei. This means that the proton sees the substructure of the target nuclei and can interact with the individual nucleons, creating a high energy particle cascade inside the nuclei of the target material.
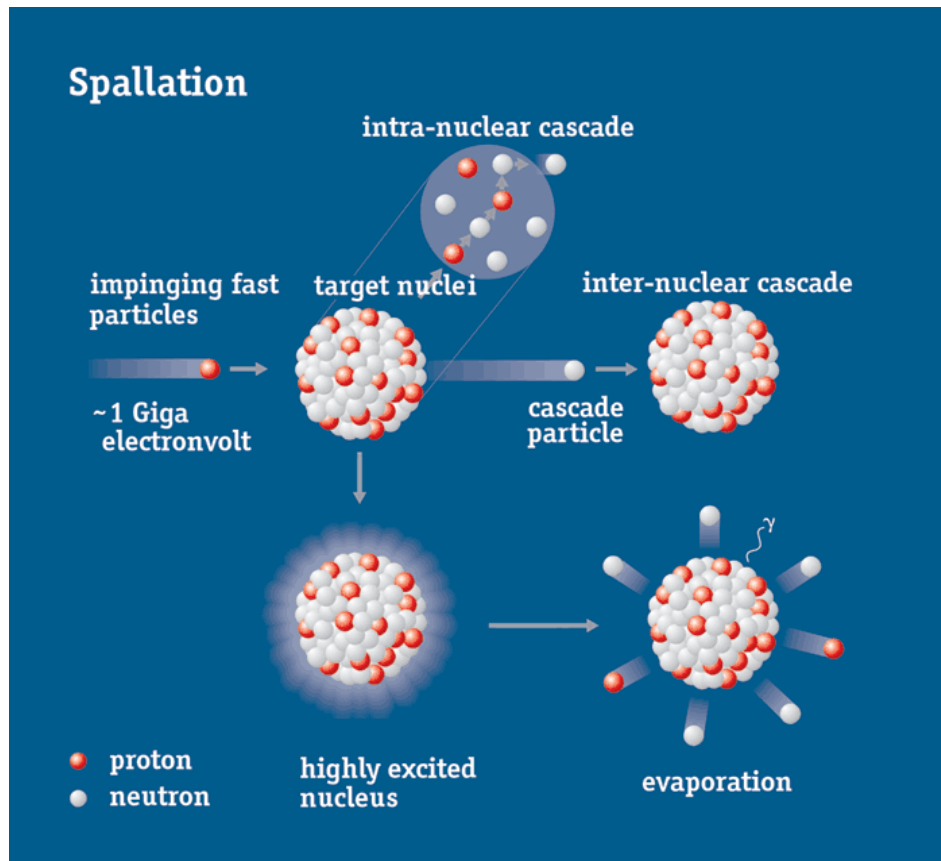
FIGURE 2.1 Spallation reaction showing the two steps, INC and evaporation

Some of the cascade particles, see FIGURE 2.1, leave the target with high energy (> 20MeV), sometimes up to the incident beam energy, or with low energy (< 20MeV) [10]. This inter-nuclear cascade of particles inside the target nuclei consists of neutrons, protons and also pions if the incident beam energy is higher than 400 MeV. [11]

The inter-nuclear cascade particles, see FIGURE 2.1 in the target induces secondary reactions in other nuclei of the target material, such as the (n, γ) and (n, xn) reactions [12]. Also scattered protons, with energies up to 50 MeV can induce secondary reactions. The secondary reactions can create elements with higher Z than the target element, if for example a proton or an α-particle is captured in a target nuclei.
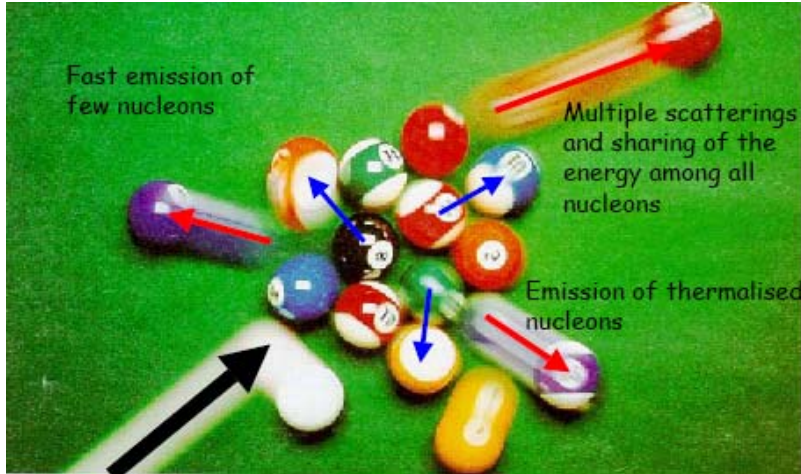
FIGURE 2.2 The intra-nuclear cascade; the first step in the spallation reaction

The second step of the spallation reactions occurs as the nucleons have been emitted and the remaining residual is left in an excited state. The kinetic energy in the excited nucleus is statistically evenly distributed over all nucleons. The de-excitation of the excited residual nuclei progresses in two ways, either through evaporation of nucleons or through fission. Both steps occur more or less immediately (in less than 10^{-16} s).
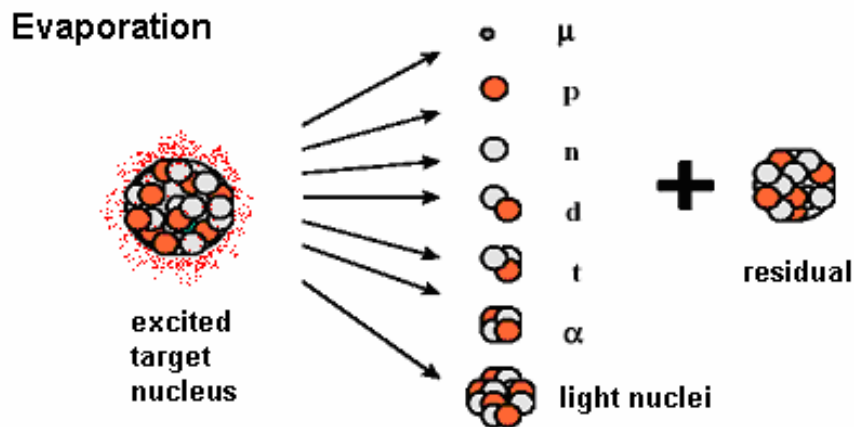
FIGURE 2.3 The evaporation mode; the second step in the spallation reaction [14]

For light nuclei, that are non-fissile or hardly fissile, the favored de-excitation mode is the evaporation. The primarily ejected particles are the evaporation neutrons of 2-3 MeV, but also protons. When the excitation energy is higher than required for the ejection of just one nucleon, not only nucleons but also light particles and nuclides like muons, deuterium, tritium, α-particles, ^{3}He, Li and Be-particles have a probability to be emitted. See FIGURE 2.3. Evaporation of for example an α-particle costs about 15 MeV. [13]

The relaxation of the excited target nuclei through fission is for medium mass nuclei and heavy nuclei, such as lead, also a possibility. The excited target nucleus fissions into two fragments of different masses.

During the evaporation and fission de-excitation of the target nuclei, excessive energy can be emitted as γ-rays. The spallation products are often radioactive after the de-excitation phase, and continue decaying through α, β and γ-emission, until the daughters nuclei have reached a stable state. The radiotoxicity of the remnant nuclei differ, depending on the initial target material, type and energy of the incident particle beam.

2.1.2 The spallation products

The distribution of the produced residual nuclei in a lead bismuth target hit by proton beam of 1.4 GeV energy can be seen in FIGURE 2.4.
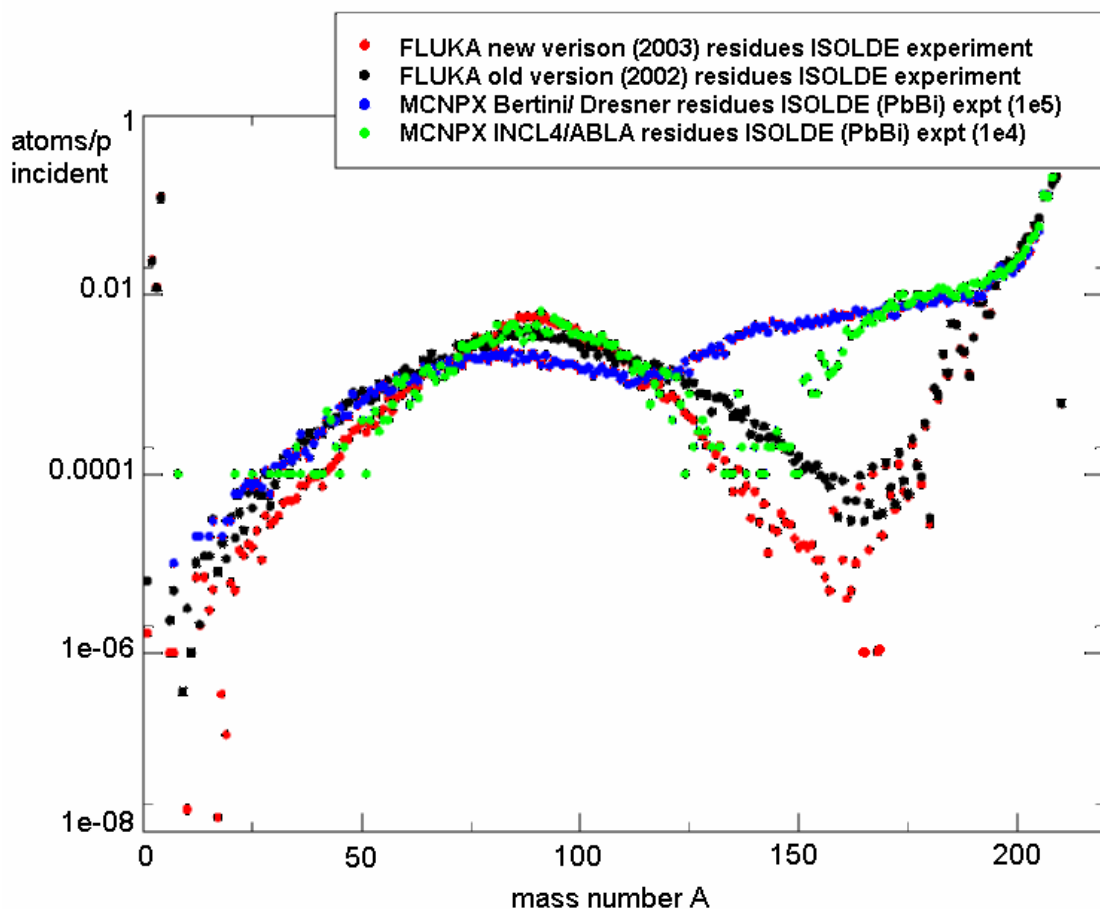
FIGURE 2.4 Distribution of the spallation products in a thick target with ISOLDE geometry, calculated with FLUKA and MCNPX code [15]

Such a high energy proton beam tends to favor spallation and fragmentation, which give

neutron deficient products. These can be found on the left side of the stability line in the chart of nuclides.

The nuclides of lower mass number are the fragments ejected from the target nucleus during the intranuclear cascade and during the evaporation process. The nuclides of mass number around 100 are mainly fission products. Stable lead isotopes have mass numbers ranging from A=204 to A=208. The residual nuclei of mass close to the target mass are the remnant from the spallation evaporation process. As one can see in FIGURE 2.4, the different Monte Carlo codes are predicting diverse distributions. This is a result of the fact that the codes are using slightly different physical models to simulate the spallation process.

2.2 THE CHART OF NUCLIDES

The chart of nuclides is a summary of all known isotopes of the elements and some of their properties; half-lives, decay modes and energies of the most common radiation modes.
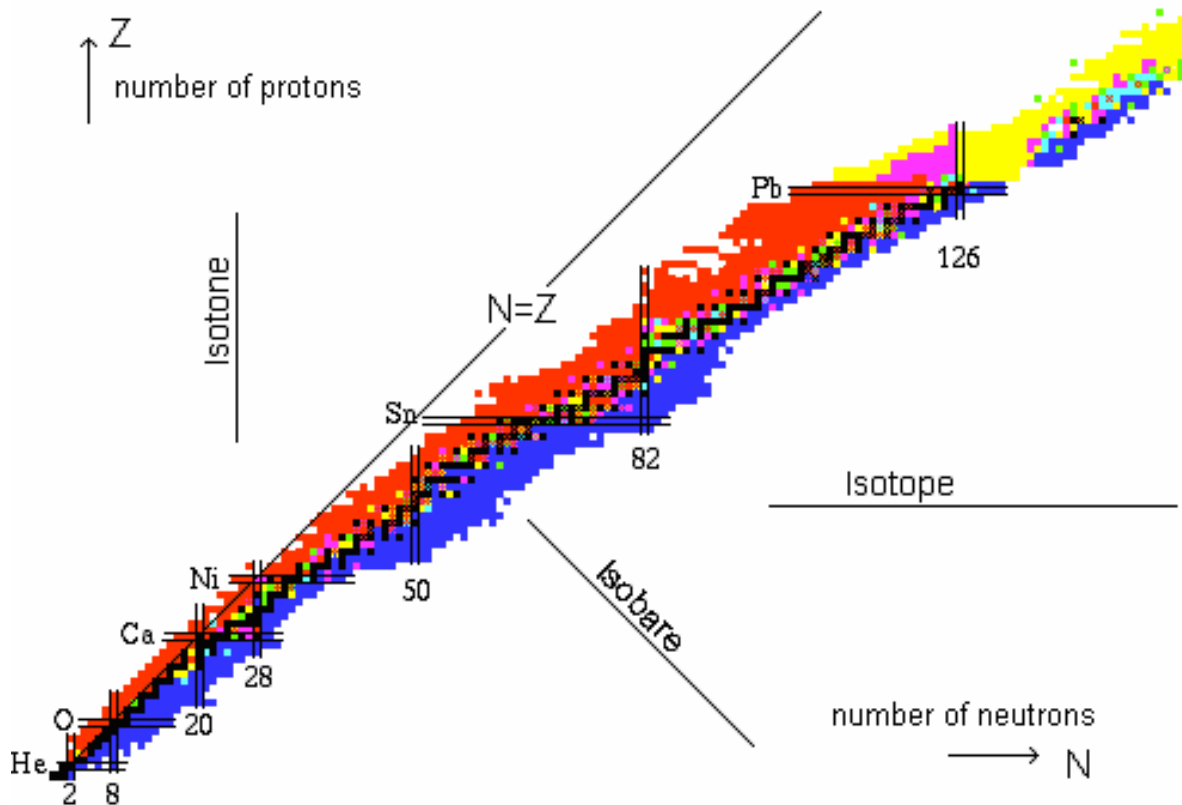
FIGURE 2.5 The chart of nuclides [16]

Most of the nuclides are not present in nature today but can be synthetically produced. All elements from Z=1 to Z=83 (except Tc Z=43 and Pm, Z=61) have at least one or

more stable isotopes and a number of radioactive isotopes. In the chart of nuclides the isotopes are arranged with the number of protons (Z) as a function of the number of neutrons (N). This result in the nuclide chart with the so-called stability line (black) in the middle, see FIGURE 2.5, tilted towards the neutron rich side of the chart. This can be explained by investigating the stability of the nucleus. To compensate the repulsive Coulomb force of the protons in the nucleus an abundance of neutrons is needed to maintain the stability for high Z elements.

Independently of the number of protons or neutrons in a nucleus, a nucleon has a mean binding energy of roughly 8 MeV. The nucleon-nucleon force seems to be almost independent on whether the nucleon is a proton or a neutron, called the independence of the strong force. To separate one neutron from the nucleus energy about 8-10 MeV is required.

On the right side of the stability line (marked in blue) the β⁻ decay is the favored, while on the left side (marked in red) the β⁺ decay is most common. For elements heavier than Z=83 the decay mode is mainly dominated by α decay (marked in yellow).

2.3 DECAY MODES

2.3.1 The β decay and electron capture

The β decay is a radioactive decay where a β particle, an electron or a positron, is emitted. In the β⁺ decay a proton is converted into a neutron through the weak nuclear force and a positron and a neutrino are emitted. In the β decay a neutron is converted into a proton and an electron and an antineutrino are emitted. The lower left hand side of the stability line up to lead is, because of its neutron deficiency, mainly dominated by β⁺ decay (2.1) and electron capture (2.2). In electron capture an electron of an atom's inner shell is drawn into the nucleus where it combines with a proton, forming a neutron and a neutrino. Sometimes the process is called K-capture, since the captured electron usually comes from the atoms K-shell. EC is favored when there is too little energy to emit a positron. [17] [18] [19]

$$\beta^+ \text{ decay:} \qquad {}^{A}_{Z}X_N \rightarrow {}^{A}_{Z-1}Y_{N+1} + e^+ + \nu_e \qquad\qquad (2.1)$$

$$\text{EC, } \epsilon \text{ decay:} \qquad {}^{A}_{Z}X_N + e^- \rightarrow {}^{A}_{Z-1}Y_{N+1} + \nu_e \qquad\qquad (2.2)$$
(K-capture)
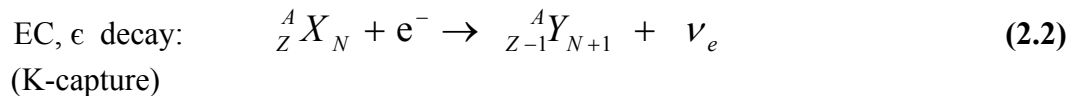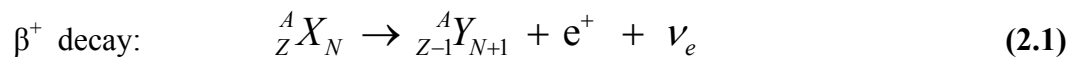
The right hand side of the stability line is, because it is neutron rich, dominated by β⁻ decay (2.3).

β⁻ decay: $$_{Z}^{A}X_{N} \;\rightarrow\; _{Z+1}^{A}Y_{N-1} \;+\mathrm{e}^{-}+\; \overline{\nu}_{e} \tag{2.3}$$
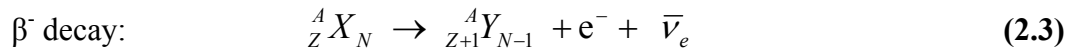
2.3.2 Internal transition

Electron capture and β decay, with some exceptions, result in emission of a specific γ spectrum. In most cases are the γ emitted directly (normally in less than 10^{-9} s [20]) as the isotope decays and falls into ground state, which is stable or also radioactive. Often these decays lead to an excited state of the daughter nuclide, with a certain half-life, which decay through internal transition (2.4), also called isomeric transition (IT). The excited daughter nuclide emits γ down to ground state. These isomeric states or also called metastable state, are denoted with a superscript m, e.g. ^{197m}Hg. Another way of denoting an excited nucleus is to add an asterisk*.

IT, 1γ $$_{Z-1}^{A}Y^{*}_{N+1} \;\rightarrow\; _{Z-1}^{A}Y_{N+1} +\gamma \tag{2.4}$$
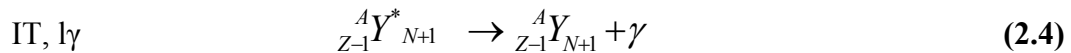
Sometimes, the only decay mode for a metastable state, is the internal transition, other times the metastable state decays partly through internal transition and partly through β decay and/or electron capture or even α decay.

2.3.3 Conversion electrons

A competing decay process to the γ-ray emission is the internal conversion. The excitation energy in the atom is used to emit one of the electrons from the atom. This electron is called a conversion electron. The vacancy is filled with an electron from higher level and at the same time the energy difference of the two levels is emitted as an X-ray. X-rays are distinguished from γ-rays by their origin. X-rays are produced by energy transition due to accelerating electrons while γ-rays are produced by nuclear transition. Sometimes the electron transitions have higher energy than nuclear transition, resulting in an overlap between low energy γ-rays and high energy X-rays. This causes a problem for the γ spectroscopy, since the γ-rays and the X-rays are not to be distinguished for the lower part of the γ spectrum.

The internal conversion coefficient is defined as the ratio of internal conversion decay probability to γ decay probability.

2.3.4 Example of a decay scheme

An example of a decay scheme is presented in FIGURE 2.6. The isomer ^{197m}Hg has a half-

life of 23.8 hours and decays in two ways; through internal transition (IT) and electron capture (ϵ). With the probability of 91.4 percent ^{197m}Hg is decaying through internal transition by emitting two γ-rays of 164.97 keV and 133.98 keV. The second mode of decay, with a probability of 8.6 percent, is to the daughter nuclide ^{197}Au. The γ-rays emitted are all coming from excited states in the ^{197}Au isotope. The most abundant of these γ-rays is the 279 keV. In the chart of nuclides this γ-ray "belongs to" the isomer of ^{197}Au, i.e. ^{197m}Au. The end product is the stable isotope ^{197}Au. When ^{197m}Hg is decaying to ^{197}Au there is an energy difference, denoted as the Q-value. For every possible level of decay the spin is indicated.
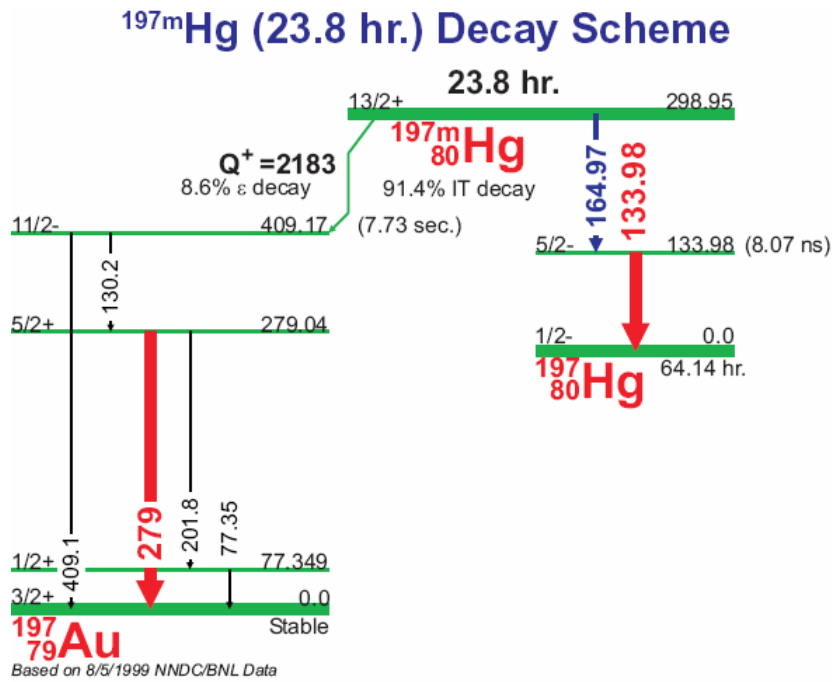
FIGURE 2.6 Decay scheme of ^{197m}Hg

2.4 ACTIVITY

When a target is hit by the continued or pulsed proton beam, the target is activated through the spallation reaction. This means that the activity of the target is changing over time, since new radioisotopes are produced and the already existing ones are decaying with different half-lives to stable elements or to other radioisotopes.

2.4.1 Production and decay

The production rate (R) of a radioisotope can be approximated by Eq (2.5). [20]

$$R = N_0 \sigma I_p \qquad \text{(2.5)}$$

The reaction cross section is denoted σ and I_p is the flux or current of the incident particle (here the proton). It is assumed that the number (N_0) of initial target nuclei is much bigger than the produced number of radioisotopes, which makes it possible to describe the production rate independent of how the initial target material is changing.

The number of radioactive isotopes is varying over time. If the number of radioisotopes are denoted N_1 one can describe the increase of N_1 according to how the production rate is varying over time, the decrease of N_1 is on the other hand described by the decay law, with decay constant λ_1 (2.6). It has not been taking into account that daughter nuclei also decay with a certain activity.

$$dN_1 = R dt - \lambda_1 N_1 dt \qquad \text{(2.6)}$$

From this relation (2.6), one can calculate the activity (2.7)

$$A(t) = \lambda_1 N_1(t) = R\left(1 - e^{-\lambda_1 t}\right) \qquad \text{(2.7)}$$

2.4.2 Radioactive parents

The isotopes produced in a spallation reaction are mainly radioactive and have also radioactive parents and daughters. A more generalized form of Eq. (2.5) is

$$dN_i = R_i dt + \lambda_{i-1} N_{i-1} dt - \lambda_i N_i dt \qquad \text{(2.8)}$$

where the number of daughter nuclei N_i depends on the production rate R_i induced by the incoming proton, the decay of the parent nuclei and its half-life and the decay of the isotope itself.

2.4.3 The Bateman equations

If one considers succeeding generations of radioactive nuclei, where all are decaying with its own specific decay constant λ_n this can be described by the Bateman equations. It is assumed that there are no daughter-products present at time zero, and the direct production of the isotope of interest is not taken into consideration. [21] The following relation (2.9) takes into account how the parent (*i*) and daughter relation (*i-1*), with different decay constants, influence the number of nuclei N_i.

$$dN_i = \lambda_{i-1} N_{i-1} dt - \lambda_i N_i dt \tag{2.9}$$

The Bateman equations (2.10), where the (1) symbolizes the first parent, (2) the daughter and (3) the granddaughter until the final daughter *n*, are presented below.

$$(1) \rightarrow (2) \rightarrow (3) \rightarrow \ldots \rightarrow (n)$$

$$N_n = C_1 e^{-\lambda_1 t} + C_2 e^{-\lambda_2 t} + C_3 e^{-\lambda_3 t} + \ldots + C_n e^{-\lambda_n t}$$

$$C_m = \frac{\lambda_1 \lambda_2 \cdots \lambda_{m-1}}{(\lambda_1 - \lambda_m)(\lambda_2 - \lambda_m) \cdots (\lambda_{m-1} - \lambda_m)} N_1^0 \tag{2.10}$$

$$m = 1 \ldots n$$

N_n denotes the number of nuclei of nuclide *n* into which the number of initial isotope nuclei N_1^0 has decayed to via daughters and granddaughters during time *t*.

2.5 MONTE CARLO CODES

Monte Carlo codes (MCC) are based on stochastic methods, simulating a particle cascade generated by the interaction of the proton beam. The MCC calculates the production rate of every specific isotope in the spallation process using the intranuclear cascade model described in chapter 2.1. To calculate the actual activity in a thick target like the MEGAPIE or ISOLDE target, where a proton beam, pulsed or continuous, is inducing new spallation reactions, an additional code has to be added to simulate the time dependence.

At the end of the input file the number of simulated protons has to be defined. In most runs of the program 1 or 2 millions of protons were chosen. The number of events was chosen to give a satisfactory statistical accuracy. For every run of the input file with

chosen parameters, the MCC makes a succession of calculations, giving the possibility to check whether the results are statistically significant.

2.5.1 FLUKA and ORIHET3

The FLUKA code was mainly developed at CERN and INFN (Instituto Nazionale di Fisica Nucleare). The intranuclear cascade is described by the PEANUT model. PEANUT (PreEquilibrium Approach to NUclear Thermalization) is a model working with energies up to 5 GeV and including pion reactions. FLUKA calculates the independent production rates of every nuclide created in the spallation reaction.

An additional code called the ORIHET3 can be added to the FLUKA code. [22] ORIHET3 is an adaptation from ORIGEN (Oak Ridge Isotope GENeration code). [1] ORIHET3 calculates how the parents and grandparents of a specific isotope are contributing to the production of that isotope, i.e. the build–up and decay of a system.

The ORIHET3 is based on the Bateman equations, see chapter 2.4.3, and applying that there is a continuous production rate of all spallation products as long as the target is being irradiated. The code takes the branching ratio of decay into account. Sometimes a parent decays through different modes, for example β and α decay. Only the part decaying to the isotope of interest is included. The ORIHET3 gives an evolution in time, showing how the activity of the parents is increasing and after some time (depending on the half-life of the parent) is reaching activity equilibrium, where production and decay of the parent is constant. When all the parents have reached equilibrium the cumulative amount of the isotope of interest can be calculated. In the production chain of n radioactive nuclei, the cumulative cross section σ_n^{cum} (proportional to the production rate) can be described as the sum of the independent cross section σ_n^{ind} and the parent contributing part

$$\sigma_n^{cum} = \sigma_n^{ind} + \sum_{i=1}^{n-1} \left(\sigma_i^{ind} \prod_{j=i}^{n-1} v_j \right) \tag{2.11}$$

where v_j is the branching ratio of the respective parent nuclides [23].

This means for example that the amount of a specific xenon isotope, e.g. ^{127}Xe, produced in the spallation target is a sum of decay products from its parent and grandparents and the direct production of ^{127}Xe (the independent production), i.e. the cumulative yield equals (^{127}Xe+^{127}Cs+^{127}Ba+^{127}La+...). In FIGURE 2.7 the build-up of ^{127}Cs is shown. If the parents have much shorter half-lives than the time of irradiation of the target the production and decay of the parents have reached the equilibrium state. For isotopes with longer-lived parents, that is parents with half-lives comparable with or longer than the time of irradiation, one only get a "partly cumulative yield", which is somewhere between the independent yield and the cumulative.
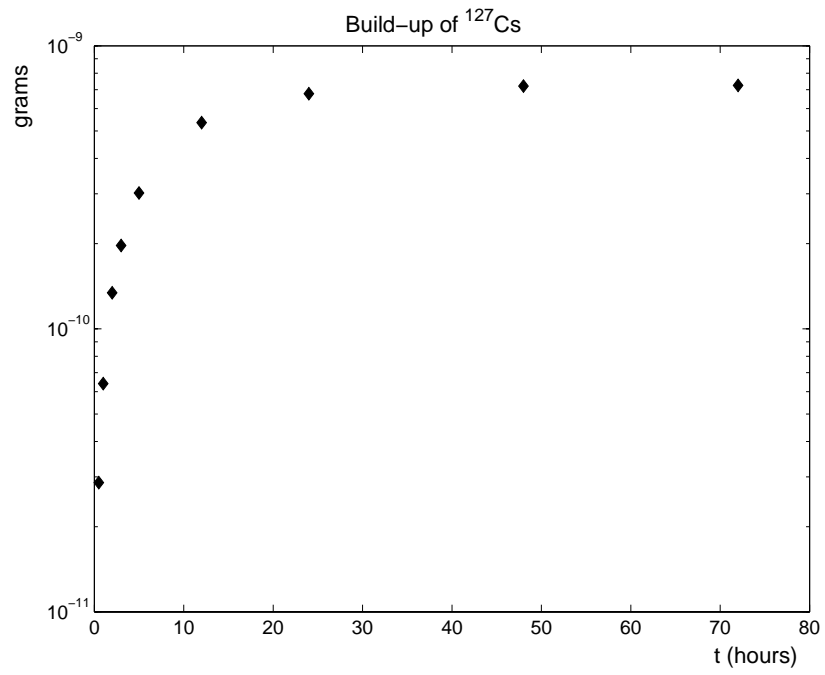
FIGURE 2.7 Build-up of ^{127}Cs to equilibrium, calculated by FLUKA coupled with ORIHET3

3 THE EXPERIMENT

3.1 THE ISOLDE FACILITY

The ISOLDE facility at CERN (FIGURE 3.1), Geneva, is one of the world's leading facilities of on-line separation of radioactive ion beams (RIB) [24], [25]. GANIL in France, GSI in Germany as well as ISAC/TRIUMF in Canada are other facilities using ISOL (ISotope On-Line separation) as a method for the RIB production, where a target in rest is bombarded with incident particles from a primary source or an accelerator.

The ISOLDE facility is used for studying problems in nuclear, atomic, surface and solid state physics, nuclear medicine and astrophysics. More than thirty years of experiments at ISOLDE, makes it the longest living experiment at CERN.
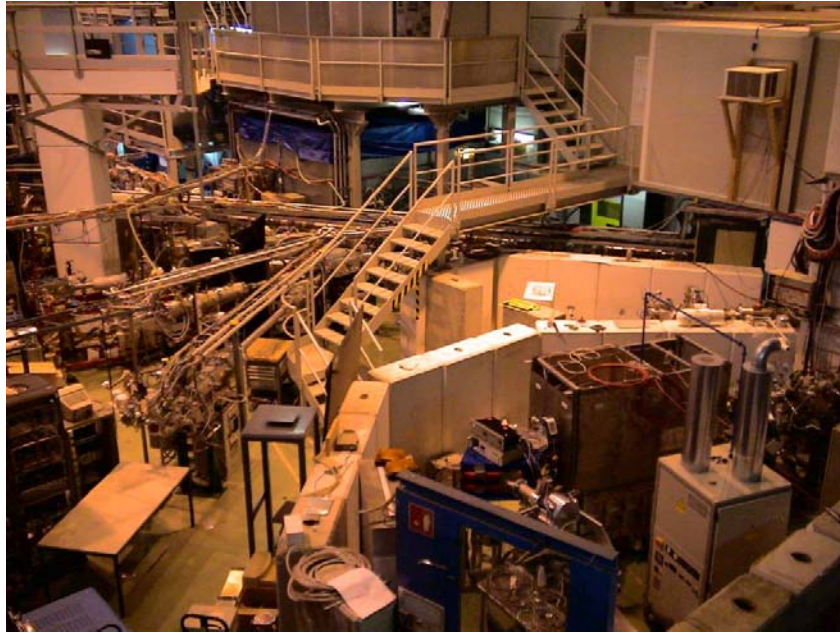
FIGURE 3.1 Picture of ISOLDE Experimental Hall

With the ISOL -technique, it has been possible to study 600 isotopes of 60 different elements (Z=2 to 88) at the ISOLDE facility. Radioactive nuclides are produced in thick high-temperature targets via spallation, fission or fragmentation reactions. Because of the thickness of the target the reaction products are stopped and sometimes trapped inside the target. For most elements the target has to be heated to make diffusion out of the target possible. The release if the reaction products vary in efficiency and rapidity depending on target material, target temperature and product itself [26]. The target and ion-source convert the released nuclear reaction products into a radioactive ion beam through chemical reactions. An electric field accelerates the ions, which are mass separated and steered to the experiments.

The facility is connected to the PS Booster (PSB), which is made up of four small synchrotrons. Protons from a linac (linear accelerator) are pre-accelerated to 1 GeV or 1.4 GeV, before being injected to the 27 GeV Proton Synchrotron. The PSB gives one pulse of $3.2 \cdot 10^{13}$ protons every 1.2 seconds. Up to half of the pulses in the 12 pulses long super cycle to the PSB is brought to bombard the ISOLDE target, which is placed in the external proton beam of the PSB. The high intensity proton beam of about 2 μA, is pulsed with a low repetition rate. This kind of pulsed beam favors the production of very short–lived isotopes, but increases the stress on the target. The pulsed proton beam is also causing a major technological problem; the extremely varying load on the acceleration voltage. It is caused by an intense ionization of the air induced by the beam in front and behind the target.

The protons from the PS Booster are delivered to the targets via an about 100 m long underground transfer. The proton beam bombards either the HRS target or the GPS target. The HRS target is connected to the High Resolution Separator and the GPS target is connected to the General Purpose Separator. See FIGURE 3.2.
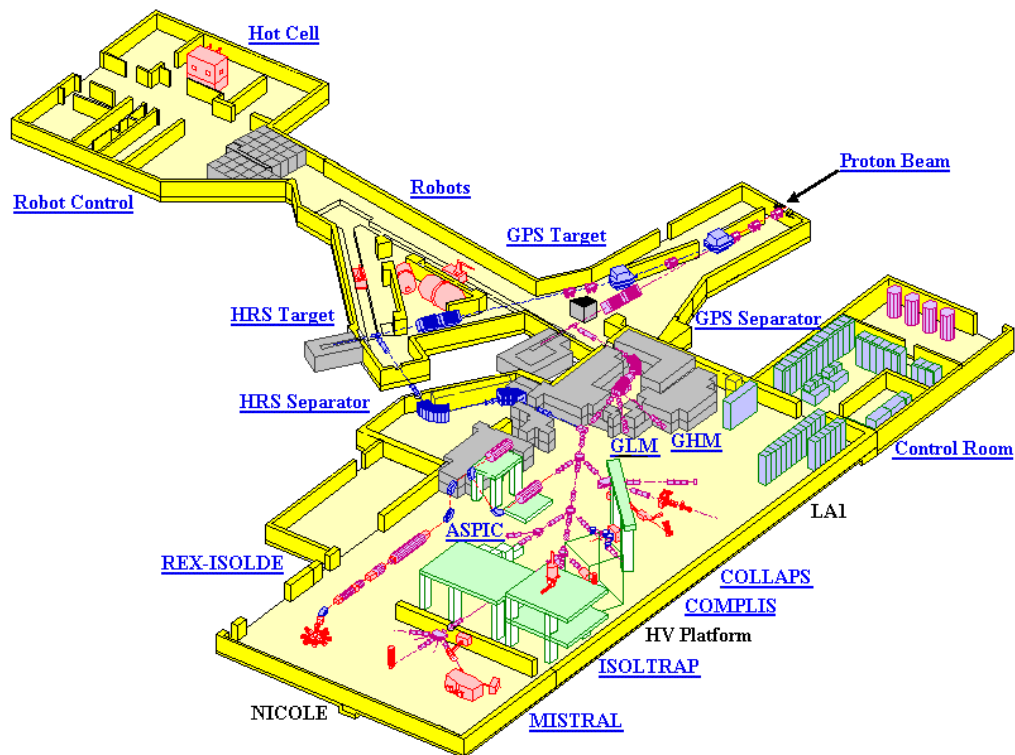
FIGURE 3.2 ISOLDE hall, schematic view

The ISOLDE area, except the experimental hall, is buried under 8 m of earth for radiation protection purposes. The handling of the radioactive target and ion source units are done by robots moving on rails. All radioactive material stays in the zone until time for disposal. The main experimental area, to which the physicists have access, is completely

separated from the radioactive handling bay, to which access is through a special entrance. The experimental hall, with its 900 m^2 is housing seven permanent experiments and four beam line ports for short term experiments.

3.2 PROCEDURE OVERVIEW OF THE EXPERIMENT

About five meters away from the target it is possible for the physicists to modify and do adjustments of the experiment. Important parts to adjust are the magnets in the proton beam transport line, before the beam hits the LBE target.

When the nuclides have effused out of the target, they are transferred through the copper transfer line and then ionized by means of an MK3 plasma ion source. The MK3 ion source is usually used for liquid targets to limit the vapor pressure in the ion source and avoid that isobars with low volatility are passing through. [27] To find out the ionization efficiency of some specific gases, a mixture of the gases of interest with known leak rate is connected to the ion source.

With a high voltage the ions are accelerated and gain a velocity given by $\frac{1}{2}mv^2 = qV$, where V is the potential difference used. The high voltage used during the experiment was 60 kV. The ions were transported through the General Purpose Separator. See FIGURE 3.2. In the separation chamber the charged ions are influenced by a centripetal force, given by $mv^2/r = qvB$, where B is the magnetic field. Only the isotopes with the right mass are able to go through the separation chamber [28].
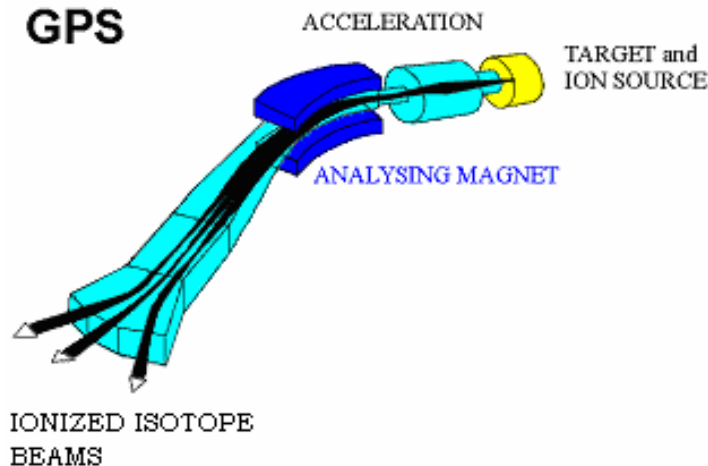
FIGURE 3.3 General Purpose Separator (GPS)

The GPS (FIGURE 3.3) is designed to select three ion beams with a mass range of ± 15%. The magnet is a double focusing H-magnet (an H-shaped iron magnet) with a bending angle of 70° and a mean bending radius of 1.5 m. The mass resolving power is 2400 and

is defined as R= m/Δm, where Δm is the mass difference of two neighboring masses m and (m+Δm) with equal intensity and overlap of 10%.

The selection of which ions that are allowed to go through the GPS is done by adjusting the magnets in the GPS by entering the wanted isotope mass number in the mass control window program placed in the control room, see FIGURE 3.2. Adjustments were possible for the ion beam, to make sure that the beam was well centered. This was done by using the mass calibration calculator.

In the GLM area, see FIGURE 3.2, a ladder of aluminum foil (ca 1x10 cm) of 0.5 mm thickness was inserted. Every aluminum ladder contained 8 samples of different γ-emitting radioisotopes. To be sure that the isotope ions really were hitting the aluminum foil, the ion current was measured with an ammeter. Every 16.8 second a proton beam pulse was hitting the LBE target and inducing a spallation reaction. For most isotopes one pulse was enough to get an ion beam of the wanted isotope of sufficient intensity. For less commonly produced isotopes two or more pulses of proton beam were needed to get a sufficient ion beam.

After irradiation the ladder of aluminum foil was taken out of the GLM and transported to the lab for evaluation with a germanium detector. Since many of the isotopes collected are rather short-lived (half-lives from 6-7 min up till half-lives of some days), it was of importance to start the γ detection as fast as possible, to avoid the decay of isotopes of interest.

FIGURE 3.4 shows a procedure time overview and FIGURE 3.5 summarizes the different collection steps.

Start of irradiation	⇨	Start of collection	⇨	Start of measurement with γ-detector
	(2-3 d)		(15 – 60 min)	and acquisition program
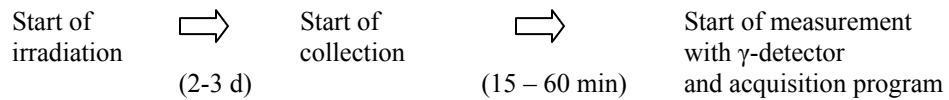
FIGURE 3.4 Procedure time overview

Isotope diffusion/ effusion from target	⇨	Isotope transportation through cupper transfer line	⇨	Isotope ionization in MK3 ion source	⇨	Ion separation in GPS	⇨	Ion transm. to exp. setup	⇨	Ion collection on Al foils
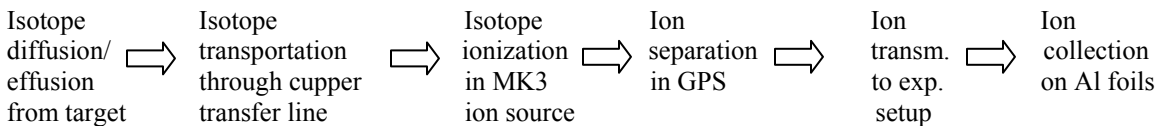
FIGURE 3.5 Collection overview

3.2.1 Efficiencies

To determine the production rate of a specific isotope produced during the spallation process in the LBE target one has to take into consideration that the final radioactive decay observed in the γ offline spectrometer was affected by a number of parameters. These parameters are:

- *The release efficiency* (ε_{rel}), i.e. the fraction of the isotope that is released from the LBE target after the spallation reaction. See chapter about the Faraday Cup

- *The ionization efficiency* (ε_{ion}), i.e. the fraction of the released isotopes that are really ionized, in the ion source. The ionization efficiency is really a product of the three different efficiencies: the ionization efficiency, the extraction efficiency and the separator efficiency, but is in this experiment considered as one unit.

- *The transmission efficiency* (ε_{transm}), i.e. the fraction of the released and ionized isotopes that are transported to the GLM and hitting the aluminum foil.

- *The detector efficiency* (ε_{det}), i.e. the fraction of the emitting γ-rays from the isotopes on the aluminum foil is really detected by the germanium detector. See chapter about the off-line γ spectroscopy

The rate of independent production **r** can be described by the following relation [26]

$$r = \Phi \cdot \sigma_{ind} \cdot N \cdot \varepsilon_{rel} \cdot \varepsilon_{ion} \cdot \varepsilon_{transm} \cdot \varepsilon_{det}$$

where the **Φ** is the flux of the incident particles, the protons, σ_{ind} the cross-section of producing a given isotope in the target and **N** the number of target atoms exposed to the incident beam. Decay losses are included in all the efficiencies, and are only relevant for short-lived isotopes. One should note that the individual efficiencies correlate. For example if the efficiency of an ion source is improved, the emittance may be worsen, thus reducing the transmission efficiency. [26]

When taken all these efficiencies into consideration, one can calculate from the γ-rays detected in the off line γ-spectrometer, how much of a specific isotope was produced in the spallation target.

3.3 TARGET AND BEAM

3.3.1 The MEGAPIE target

The target proposed for the MEGAPIE project, and investigated in the ISOLDE experiment, is an eutectic mixture of lead and bismuth with the composition 45 % lead and 55 % bismuth. Natural lead is planned to be used, including the isotopes [204]Pb with natural abundance 1.4%, [206]Pb (24.1%), [207]Pb (22.1%) and [208]Pb (52.4%). The isotope [209]Bi, which is the only stable isotope of bismuth, makes up the bismuth part of the target. The size of the cylindrical target in the MEGAPIE experiment will have a total length of 4 m and a diameter of 20cm in the bottom part and of 40 cm in the top part.

Properties	Pb	Bi	LBE	Hg
Composition	elem.	elem.	*Pb 45% Bi 55%*	elem.
Atomic number, Z	82	83	-	80
Atomic mass A (g/mole)	207.2	209	*208.2*	200.6
Density (g/cm3) solid	1135	9.75	*10.5*	-
liquid	10.7	10.07	*10.57*	13.55
Thermal expansion ($10^{-5}K^{-1}$)	2.91	1.75	-	6.1
Contraction upon solidification (%)	3.32	-3.35	*0*	-
Melting point (ºC)	327.5	271.3	*125*	-38.87
Boiling point at 1 atm (ºC)	1740	1560	*1670*	356.58
Specific heat capacity (J/gK)	0.14	0.15	*0.15*	0.12
Thermal neutron absorption (barn)	0.17	0.034	*0.11*	389
Fast neutron absorption	0.002		*0.0025*	0.1

TABLE 3.1 Relevant properties for liquid metal targets [6] [29]

Relevant properties of different liquid metal targets are shown in TABLE 3.1. The LBE has a low melting point of 125 ºC (lead alone has a melting point of 327 ºC) and a high boiling temperature of 1670 ºC. The low LBE melting point in comparison with lead makes it a favorable as target in the MEGAPIE experiment.

Most countries involved in the ADS development favor LBE as spallation target [1]. The liquid metal target has several advantages [6]. First of all a liquid target has a larger flexibility than a solid target, since it is not susceptible to structural radiation damage and the solid parts can be chosen as convenient as possible. A solid target brings more difficulties in handling the heat removal, whereas in a liquid target a much higher power density and direct heat removal are possible. A liquid target minimizes the amount of water needed to cool the target, reducing the radioactivity levels in the cooling water and leads to less exposure for the maintenance personnel. The liquid LBE will circulate inside its container, for cooling purposes.

The planned operation temperature of the LBE target at the SINQ in the MEGAPIE project varies mainly between 230 ºC and 360 ºC, depending on the location in the target. The temperature in the LBE has a peak value of 400 ºC.

The liquid LBE is contained in a containment, consisting of martensitic and stainless steel. The container is enclosed in a safety hull that is filled with an isolating gas. Both are closed during the irradiation. The expansion tank above the liquid LBE, where the volatiles accumulate during irradiation, will however be drained from time to time, depending on the gas pressure rise. The isolating gas serves as a thermal insulation and through pressure observation it can be used as detection system for leakage. The isolation gas will be a mixture of argon and helium.

3.3.2 The ISOLDE target

The same composition of LBE and a tantalum container are used in the ISOLDE experiment. Tantalum is a heavy, hard and ductile metal most unlikely to react with most elements and with a high boiling point. [30] The mass of the LBE target, used in the ISOLDE experiment, is 547 grams and the target has a cylindrical shape with a length of 20 cm and radius of 1 cm.

For the ISOLDE experiment, the LBE target was inserted in the GPS in the ISOLDE facility [31], [32]. The target was first held at a temperature of 600 ºC with the help of a current of 280 A. A few measurements were also done at 400 ºC. There is a maximum operation temperature for the target, determined by the vapor pressure. For lead the maximum operation temperature is 800 ºC and for bismuth 600 ºC. The tube through which the volatile ions are transported away from the target is also heated to a temperature of 300 ºC.

3.3.3 The MEGAPIE beam

The accelerator at PSI produces a proton beam, at a power level of 1 MW, with proton energy of 575 MeV and continuous current of 1.8 mA. Most of the current is transported to the neutron spallation source SINQ, where the LBE target will be placed. During the MEGAPIE experiment the current will be 1.4 mA.

3.3.4 The ISOLDE beam

The beam used in the ISOLDE experiment has the energy of 1.4 GeV and arrives from the PSB as a pulsed beam. The fact that the proton energy of the beam used in the ISOLDE experiment differs from the beam to be used in the MEGAPIE will not change much of the physics investigated in the ISOLDE experiment. The neutron yield will be lower but the volatiles will still be produced in both experiments. The pion production has, as mentioned, a threshold production level of 400 MeV and will be produced in the

MEGAPIE experiment as well as the ISOLDE experiment, i.e. isotopes produced by a pion induced reaction will occur in both experiments.

When the experiment was carried out at ISOLDE one pulse per super cycle was used, meaning one proton pulse every 16.8 seconds. Most isotopes were irradiated for just one super cycle, but some long lived isotopes were irradiated on the aluminum foil for ten super cycles i.e. ten pulses during 168 s or even longer.

The beam hitting the LBE target has a proton intensity of about 10^{13} protons per pulse. If the proton beam intensity is increased there is a risk that the target will suffer from shockwave effects.

The energy loss in a thick target ($>>1$g/cm^2 [26]) is given by the stopping power of $\int_0^L \frac{dE}{dx} dx$. When the beam leaves the target of the ISOLDE experiment it has the energy of about 800 MeV. This is a typical thick target experiment and chosen for the ISOLDE experiment to resemble the MEGAPIE target. Nevertheless in the MEGAPIE the beam is slowed down completely. The effect of the small target diameter will be taken into consideration in the Monte Carlo simulation.

In a thick target experiment the proton beam energy changes as it moves through the target, while in a thin target experiment it remains more or less the same. A disadvantage of a bigger target is that it is more difficult to get good release efficiency. For isotopes close to the lead and bismuth mass, which are conducted to secondary reactions, the thick target experiment is more reliable than the thin target experiment. Though, the farther away one goes from target mass, the better a thin target experiment would be to predict the independent production rate of a specific isotope.

3.3.5 Summary of beam and target parameters

The following table, TABLE 3.2, presents the parameters of the beam and target for the ISOLDE experiment and the MEGAPIE experiment.

Parameter	ISOLDE experiment	MEGAPIE experiment
Beam energy, incoming	1.4 GeV	575 MeV
Beam pulse length	2.4 μs	Continuous beam
Beam repetition rate	16.8 s	Continuous beam
Beam current (mean value)	$0.96 \cdot 10^{-4}$ mA	1.4 mA
Beam profile (Gaussian)	$\sigma_x = \sigma_y = 3.5$ mm, FWHM=8.24mm	$\sigma_x = 3.31$ cm, $\sigma_y = 1.9$ cm, [29] $FWHM_x$=7.78, $FWHM_y$=4.47
LBE mass	547 g	800 kg
Target size (cylindrical)	L = 20 cm, r = 1 cm	L = 4 m, d = 20-40 cm
Target temperature at experiment	400 ºC, 600 ºC	230-360 ºC, with peak temp of 400 ºC
Transfer line temperature	300 ºC	300-350 ºC

TABLE 3.2 Summary of beam and target parameters used in the
ISOLDE experiment and MEGAPIE experiment

3.4 MEASURED VOLATILES

As a result of the spallation reaction in the liquid LBE target, as mentioned not only the wanted neutrons are produced, but many different nuclides, stable or radioactive with different radiotoxicity. Many of them are highly volatile and in gaseous form and will diffuse out of the target. [7] In the case of MEGAPIE the volatiles will be collected at the top of the LBE target and will have to be drained from time to time during operation of the experiment.

3.4.1 Volatility

When a liquid evaporates a certain amount of pressure is exerted by the vapor, the so called vapor pressure. If an element has a large vapor pressure it is normally a volatile and the volatility increases with increasing temperature. An exception is when the element reacts with other materials in the target or its container. Then the element can not effuse out of the target container freely and can not be considered as volatile [27], unless it forms a volatile compound. There is an equilibrium vapor pressure when the rate of evaporation of the liquid is equal to the condensation rate of the vapor. [33]

One way of classify the volatile isotopes are on the basis of the temperature at which the vapor pressure reaches 0.01 mbar. [34] Elements important to study for the MEGAPIE project are mainly the highly volatiles; H, He, N, O, F, Ne, Cl, Ar, Br, Kr, I, Xe, At and Hg. They all reach the 0.01 mbar level at a temperature lower than 100 ºC. Other less volatile elements of interest are Na, P, S, K, Zn, As, Se, Rb, Cd, Te, Cs and Po, which all reach the vapor pressure level of 0.01 mbar at a temperature lower than 400 ºC. Other elements need a higher temperature to reach the same vapour pressure. See FIGURE 3.6

Group	1	2		3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
Period																			
1	1 H																		2 He
2	3 Li	4 Be												5 B	6 C	7 N	8 O	9 F	10 Ne
3	11 Na	12 Mg												13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
4	19 K	20 Ca		21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
5	37 Rb	38 Sr		39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
6	55 Cs	56 Ba	*	71 Lu	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	* *	103 Lr	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Uub	113 Uut	114 Uuq	115 Uup	116 Uuh	117 Uus	118 Uuo

*Lanthanoids	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb
**Actinoids	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No

Yellow high lights the highly volatile elements
Green high lights the less volatile elements
Purple represents LBE target material

FIGURE 3.6 Periodic table showing the most volatile elements

3.4.2 Volatiles from irradiated LBE target

There are two main reasons to evaluate the volatile elements produced in the MEGAPIE experiment. Firstly it is of interest to know the amount of stable and radioactive elements produced to be able to handle the gases during operation. The stable volatiles are produced in large quantities, in particular ^{4}He and H, which are expected to be produced at a rate of 1 liter per month. In the MEGAPIE target a gas plenum of 2 liters volume will be inserted to handle this amount of gas. These production rates must be experimentally verified.

If there is a leakage in the target, it is of great importance to avoid that the radioactive volatiles come out. If the real amount of volatiles is known, protection systems in the form of chemical and physical traps can be constructed to trap the volatiles.

The following considerations of production rates and releases are about the ISOLDE target and the volatiles produced in the ISOLDE experiment, if nothing else is mentioned. Most of the considerations can directly be applied to the MEGAPIE target.

The goal with the ISOLDE experiment was to measure all isotopes that come out of the LBE target. By choosing some specific isotopes of interesting elements, prediction of

other isotopes can be done. For example all noble gases are highly volatile and are of interest to measure, since they are easily released from the target. As mentioned, most of the elements where measured at the temperature of 600 ºC, but a few were also measured at the temperature of 400 ºC.

3.5 OFF LINE GAMMA SPECTROSCOPY

3.5.1 Measured isotopes

In the off line γ spectroscopy there are some things to consider before one can do some reliable measurements. The half-life of the isotopes is the first and most important parameter. To be able to get reliable results, the half-life has to have a lower range limit of 10-15 minutes (the time it takes to get the sample to the germanium detector and starting the analysis with the acquisition program). Theoretically there is no upper limit in half-life, but to get good statistics isotopes with half-life less than some days or weeks were chosen.

The low energy γ-rays coincide with the X-rays, making it difficult to identify the low energy γ-lines of some isotopes. Therefore isotopes with only low energy γ-rays (in the region of about 10 to 100 keV) should be excluded.

The following table, TABLE 3.3, shows the isotopes measured with off-line γ spectroscopy. They have all medium half-lives, to be able to measure them off-line.

Elem.	Mass number (A)
Hg	189, 189m, 190, 191, 192, 193, 193m, 195, 195m, 197, 197m 199m, 203
Xe	120, 121, 122, 123, 125, 127, 129m, 131m, 133, 133m
Kr	74, 76, 77, 78, 79, 85, 88
Br	78, 80, 84 (nothing observed at 600 ºC)
Ne	24 (not observed at 600 ºC)
Ar	41
I	120, 120m, 121, 123, 124, 126, 128, 130, 130m, 132, 132m
Po	202, 203, 204, 205, 206, 207, 208, 209, 210
At	203, 204, 205, 206, 207, 208, 209, 210

TABLE 3.3 Isotopes measured with the off-line γ spectroscopy

3.5.2 The germanium detector

The high purity Germanium detector used to detect the γ-rays emitted from the decaying isotope samples is a high resolution detector, able to detect γ-rays with about 2 keV resolution at 1.4 MeV. The Germanium detector is a semiconductor detector with n- and p-type material. [35] The γ-ray interacting with the germanium produces electron-hole pairs, moving in the material due to the electric field in the detector. γ-rays moving through medium interacts in the three ways, depending on the incident energy: Compton scattering, photoelectric absorption and pair production processes. In Compton scattering, the γ-ray interacts with an electron and is scattered without loosing all of its energy. When the γ is not totally absorbed it contribute to a background in the γ spectrum, known as the Compton continuum. This can be suppressed by using an inorganic scintillation detector surrounding the germanium detector. The photoelectric absorption is the most favorable process for a γ spectroscopy and is most likely to occur for γ-rays with low and intermediate energies. If the incident γ energy is greater than 1.022 MeV (twice the electron rest mass) in the presence of an atomic nucleus an electron/positron pair can be produced. When the positron is slowed down to thermal energy through interaction with the absorption material it can annihilate with an electron producing two γ-rays of energy 511 keV. If the γ escapes the detector, the escape peak at 511 keV can be observed in the γ spectra.

Radiation decay in the human body is also detected by the sensitive germanium detector. The following table, TABLE 3.4, shows the most abundant radioactive isotopes in our body. The only isotope that clearly can be seen in the γ spectra is the ^{40}K and because of its low activity, it first becomes visible after about half an hour detection time.

Isotope	Half-life	% of radiation from our body	Decay mode	γ energy	Notes
^{40}K	$1.28 \cdot 10^9$ a	~50 %	β⁻ β⁺ EC	1461 keV	Can be seen in spectra
^{210}Pb	22.3 a	~40 %	β⁻ α	47 keV	Disappears in X-ray background
^{14}C	5730 a	~3 %	β⁻	no γ	Will not influence spectra

TABLE 3.4 Characteristics of the naturally abundant radioisotopes in our body

3.5.3 Data acquisition

The acquisition program GENIE (FIGURE 3.8) was used for identification of the γ spectra for each individual nuclide. The aluminium foils from the collection, containing the eight different radioactive samples each, were cut into eight pieces and taped onto carton for individual analysis. The aluminum foils irradiated by the radioactive ion beams were inserted into the germanium detector. One could choose three different positions for the sample, depending on how active the sample was. The higher the activity the farther away from the detector one should place the sample. In some cases spectra were measured more than once at different times to assess the isotope identification by the spectra attenuation.
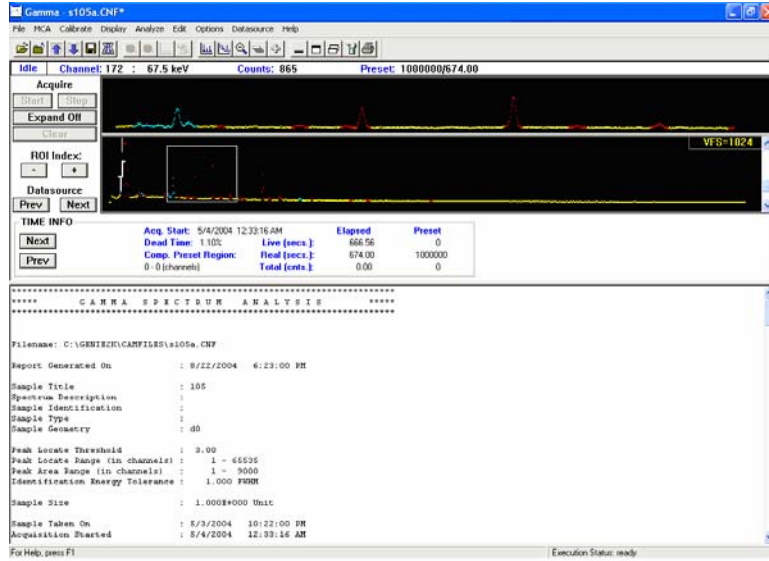
FIGURE 3.7 GENIE acquisition program

The detector energy and efficiency have to be determined, using some specific isotopes and their well known γ-lines. All three sample positions were calibrated. ^{60}Co, ^{137}Cs, ^{57}Co, ^{241}Am and ^{109}Cd and their γ-lines used for efficiency calibration can be seen in TABLE 3.5. The γ energies from the selected isotopes represent the energy span of interest.

Isotope	γ energy (keV)
^{241}Am	59.54
^{109}Cd	88.03
^{57}Co	122.05
^{137}Cs	661.62
^{60}Co	1173.23
^{60}Co	1332.51

TABLE 3.5 Calibration energies for GENIE [36]

The calibration files were done for the three positions and by placing the carton in forward and backward position. This was done to estimate the systematic error, due to bending of the sample carton. The middle value could be used for the calibration. Following FIGURE 3.8 shows the linear relation between energy and channel, calculated in GENIE with the help of the above listed isotope energies.
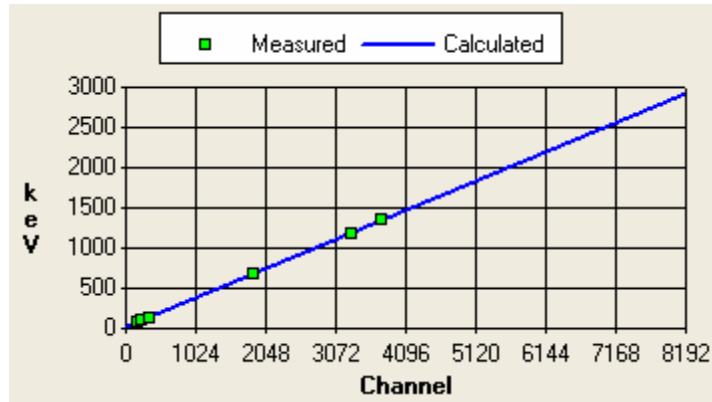
FIGURE 3.8 Energy calibration in GENIE

Using the same known radioactive isotopes above, one can with the help of GENIE calculate the detector efficiency. The following FIGURE 3.9 shows how the efficiency varies for different energies/channels, with the highest efficiency for lower γ energies. Though, for low γ energies it is most difficult to estimate the efficiency, because the steep slope of the peak.
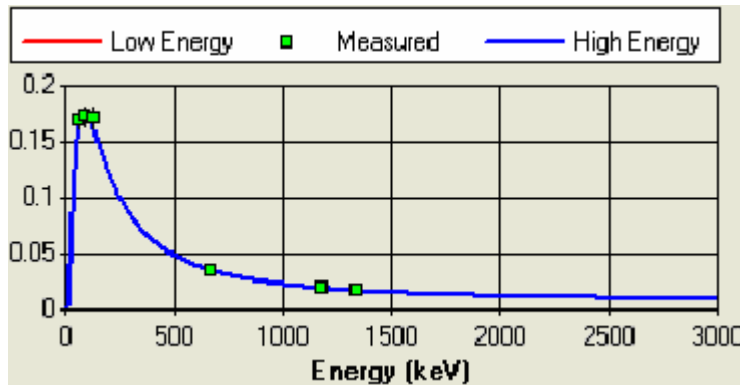
FIGURE 3.9 Efficiency calibration curve for the germanium detector

During acquisition a slight shift in the peaks in GENIE was noted after calibration of the detector. This can be due to liquid nitrogen (used for cooling the detector) influencing the amplifier. Also temperature differences can cause a peak shift, since measurements were done during daytime and night time. By adjusting the tolerance for identification this small problem can be overcome.

In the γ spectra one can identify not only the isotopes of interest but sometimes also isobars with the same mass, since they may have been collected on the aluminium foil. In FIGURE 3.10, the γ spectrum from ^{126}I can be seen. The half-life of ^{126}I is 13.11 days and a long detection time is needed to be able to see the γ lines. The detection was running for 19 hours, making it also possible to see the annihilation energy of 511 keV and the γ line at 1461 keV for ^{40}K.
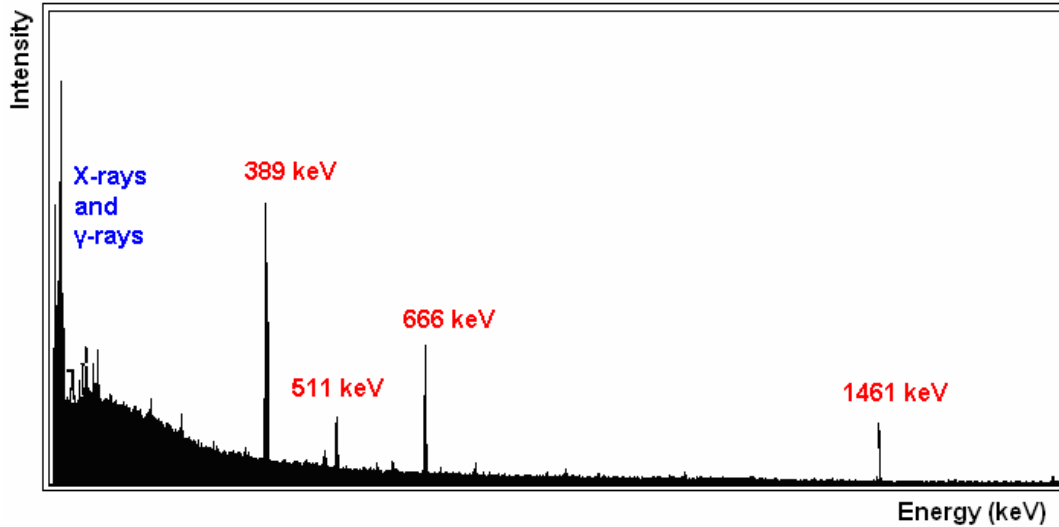
FIGURE 3.10 A γ spectra of isotope ^{126}I with γ energies of 389 keV and 666 keV, as well as the annihilation energy of 511 keV and the ^{40}K γ peak at 1461 keV

When running the acquisition program it is of great importance to record the sample position and the time of collection. After the acquisition the analysis is implemented with the help of GENIE. To run analysis with GENIE a library with name of nuclide, half-life, γ energy and γ abundances is necessary. This was prepared in advance and assumes that one knows which isotopes one is looking for. An analysis report is generated on the basis of the nuclide library and the data from the acquisition. In the report information about the γ peaks, detection counts and statistical error, collection time, measurement time, sample position are listed and suggestion to which isotope they might belong to, as well as the activity of the sample calculated by GENIE. The acquisition program uses two different times, the real time corresponding to the normal clock time and the live time, the actual time when the program is counting the γ-rays from the radioactive sample. The activity calculated from GENIE takes the total decay time into consideration from the collection of the sample on the aluminum foil to the end of the acquisition.

To calculate the number of nuclides per µC produced in the target, one has to take decay constant, ionization efficiency and transmission efficiency for each specific isotope, as well as number of protons per µC into consideration.

3.6 FARADAY CUP AND TAPE STATION

Other methods used to calculate the isotope production rate in the LBE target are the Faraday Cup (FC) and the tape station (TS), also called on-line β spectroscopy. The Faraday Cup is most useful for measuring stable isotopes or long-lived radioisotopes. The on-line β spectroscopy has the advantage over the off-line γ spectroscopy, that very short-lived radioisotopes can be measured, since it is done immediately. In Faraday Cup and tape station measurements the so called release function plays an important role.

3.6.1 The Faraday Cup

The Faraday Cup method uses ion currents ranging from pA to nA to calculate production rates of different isotopes. The release function of the specific isotopes is measured for 6 seconds after beam interaction. The data acquisition system for the Faraday Cup has been, differently from the off-line γ spectroscopy, developed at the Paul Scherrer Institut. He, Hg, Kr and Xe were some of the elements measured with the FC. Since the method is not γ dependent it has the possibility to detect stable isotopes.

3.6.2 The tape station

The short-lived radioisotopes beams are deposited in a tape and the decay is detected online for a given time. The advantage of this method in comparison with the off-line γ spectroscopy is that isotopes of very short half-lives can be measured, since the measurement is done directly. Release function can be measured as well as integrated number of counts from which the production rate can be deduced. The measurements done by the tape station are absolute measurements and sensitive to very low currents of radioactive ion beams.

3.6.3 Diffusion

Volatile species are emitted not instantaneously but through diffusion out of the target and then through effusion out of the container. When the isotope has diffused out of the target material it has to effuse out of the container to the transfer line so it can be ionized and transported to the detector. The effusion is depending on the target geometry which influences number of possible wall collisions and probability of the isotope to get stuck in the container material. Following figure, FIGURE 3.11, shows schematically how the reaction products are diffusing out of the LBE and the effusing out of the target container. The two effects, diffusion and effusion, are summarized in the release function.
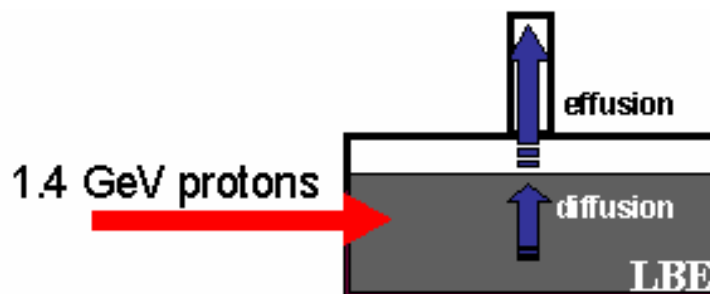

FIGURE 3.11 Target hit by 1.4 GeV protons

Sometimes the isotope produced through the spallation or fission reaction in the target or

its parents, does not diffuse/effuse out of the target before it has decayed. This means that the measured value of the production rate is lower than the actual production of the isotope in the target.

Fick's second law (3.1) describes mathematically the concentration gradient $\nabla\rho(\vec{x},t)$ in solid matrices.

$$\frac{\partial}{\partial t}\rho(\vec{x},t) = D\cdot\nabla^2\rho(\vec{x},t) = D\cdot\Delta\rho(\vec{x},t) \qquad\qquad \textbf{(3.1)}$$

with $\rho(x, t)$ the local density of the diffusing element, D the diffusion coefficient, ∇ the gradient operator and Δ the Laplace operator.

Since the isotopes diffuse out of the target one can use the diffusion law (3.2) to explain the high increase in diffused isotopes at a temperature level of 200 ºC higher. The relationship between the rate $k(T)$ a reaction proceeds and its temperature T [K] is determined by the Arrhenius equation [37]

$$k(T) = A\cdot\exp\left(-\frac{E_a}{R\cdot T}\right) \qquad\qquad \textbf{(3.2)}$$

At a higher temperature the kinetic energy is higher, which has an effect on the activation energy, E_a [kJ/mol] of the reaction, i.e. how fast the reaction takes place. A is the diffusion coefficient at infinite temperature and R is the universal gas constant.

3.6.4 Release function

There is a function describing how the release of the different isotopes in the LBE target is proceeding during a short time range. The average release time from a liquid target is typically in the order of minutes, which decreases with higher temperature. Almost all elements in our experiment are released with a fast peak after about 6 seconds. The tail after the peak represents the slow component of the release function. This tail of the release function is not measured during one cycle of 16.8 seconds, the collection time for most off-line isotopes, but it accumulates as a background in the next measurement.

The following two figures, FIGURE 3.12 and FIGURE 3.13, show the release curves for ^{80}Kr and ^{196}Hg measured with the Faraday Cup. The fast part of the release curve is easily identified as the peaks in the beginning of the curve. The end of the tail from the previous isotope measured is seen in the beginning of the curves. The fluctuations in the peaks are less important for the calculation of the release efficiency, since a big part of the release function is in the tail. The small drop just before the big peak is a result of the fact that the high voltage shortly was switched of. After about 6 ms it was restored. This is done to avoid a breakdown of the high voltage at beam impact, induced by the intense ionization of the air surrounding the target. [25] The drop in the middle of ^{80}Kr release peak is due

to fluctuations in the plasma ionization source because of the pulsed beam. This is an effect that is dependent of the proton beam intensity. Also this is negligible because of the long tail, when integrating the whole release curve to get the release efficiency.
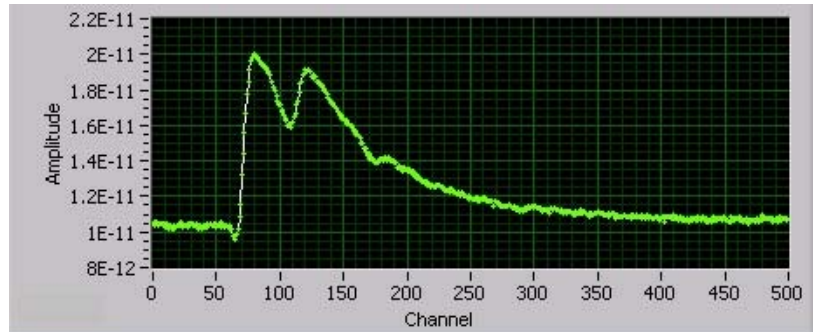
FIGURE 3.12 ^{80}Kr release function, during 6 seconds
from the arrival of the proton beam on target.
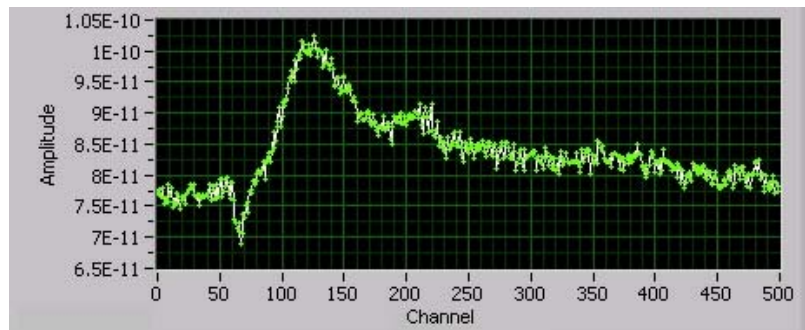
FIGURE 3.13 ^{196}Hg release function, during 6 seconds
from the arrival of the proton beam on target.

If the collection is done in equilibrium and for one cycle, the whole release of the isotope is collected. This results in a release function characteristic of each isotope, but with similar release function for isotopes of the same element.

The study of release functions is important for very short lived isotopes, like ^{6}He. By comparing the release functions of ^{6}He (measured with the tape station) and ^{4}He (measured with the Faraday Cup) the fraction of the decay of ^{6}He before detection can be calculated.

For the off-line γ spectroscopy the release function plays a less important role. For the off-line collection we were measuring for 16.8 s or multiples, this means that the complete release of the volatile was detected.

3.6.4 Release efficiency

The release efficiency is the fraction of created isotopes which are released before decaying. The release efficiency is the fraction between the integral of the release function folded with the exponential decay of the isotope and the integral of the release function without decay. [32]

For long lived isotopes that are 100 % released from the target, like expected from Hg and Xe, the release efficiency is equal to 1. The release efficiency is not known before calculating the production rate from the experimental data.

4 EXPERIMENTAL RESULTS

A spallation reaction in a thick target is a complex process. If one wants to extract a certain isotope produced in the target one has to consider different steps. The incoming beam in the thick target induces the first reactions with the target nuclei. But also secondary reactions have to be considered. One of these secondary processes is the cascade of hadrons, where among others neutrons are inducing reactions to form new isotopes. But also secondary products like pions and α-particles (^{4}He) can induce reactions and produce new elements.

Helium, which is not measured by the off-line γ spectroscopy, but only with the tape station and Faraday Cup, is produced in different ways in the target; fragmentation, fission and α decay from different isotopes (for example Hg, Au, Pb and Th). Fragmentation differs a little from spallation, since the nucleus is directly cut in two or more pieces, while the spallation is a two-step process, with heating and evaporation of particles and light nuclides.

Better predictions of independent production rates or cross sections of neon, argon and xenon may be given by a thin target experiment, but in the MEGAPIE experiment the production of these elements is a cumulative production, where the progenitors contribute to the production rates of given isotopes. This is better simulated in a thick target experiment like the ISOLDE experiment. Krypton and xenon are produced in the spallation reaction only at high proton beam energy. Exceptions are the neutron rich isotopes of krypton and xenon, which are also products of the fission reaction.

4.1 CONTRIBUTION OF PROGENITORS

Progenitors decay and contribute to the production rate of a specific isotope, see TABLE 4.1. The half-lives of the progenitors are normally shorter than for the isotope of interest. In the beginning of the irradiation there is an increase in concentration of a specific isotope, due to the decay of the parents. After some time the concentration of the parents reaches equilibrium, which means that decay equals the production of the parent. At equilibrium the parent contribute with a constant amount to the release of the volatile isotope. In the experiment it is important that equilibrium of the parents is reached. The measurements started 2-3 days (depending on element collected) after begin of irradiation.

4.2 EFFICIENCIES

Release efficiency is equal to one for the off-line results and plays only a role in the short-lived isotope measurements done by the Faraday Cup and tape station. All measured isotopes in the off-line measurements have half-lives long enough for not influencing the release efficiency.

Different transmission lines in the ISOLDE facility were used for the off-line and the on-line measurements. The *transmission efficiency* is about 90 -95 % for the off-line collection as well as for the FC and slightly lower, 88 %, for the tape station.

During the experiment two tests were done to measure the *ionization efficiency*. In the first test the ion source was injected with a mixture of 95% Ar and 5% Xe at 0.922 bar and in the second test it was filled with a mixture of 40% He, 20% Ne, 20% Kr and 20% Xe at 1.00 bar. A slight difference between the two tests indicates a probable small leakage of the ion source. Measured ionization efficiencies for xenon were 2.8-4%. The ionization efficiency for mercury was never measured but results in [38] show an ionization ratio of about a factor 1.5 for Hg+/ Xe+. Using this ratio, one can calculate the ionization efficiency to 4 % for Hg. Since mercury is a heavier element and has a lower ionization potential than Xe, higher ionization efficiency would be expected.

The *detector efficiency* is only of interest for the off-line γ spectroscopy and varies for different γ energies, see chapter about germanium detector.

4.3 RESULTS

The measured isotopes can be divided into different categories, depending on how they were produced, if the isotope has isomers, how the isotope decays and the half-life of the isotope. Some of the measured isotopes are "shielded", meaning that the production is not or almost not populated by parent decay, because the nearby isobars are stable or very long lived. Other isotopes measured were populated by parent decay that had already reached equilibrium and therefore contributing with a constant production rate. Measurements were done with Tl and Cs, parents to Hg and Xe, to be sure that the parents are not released from the target. The contributing parent decay is therefore inside the target. Isotopes with long-lived parents that have not yet reached equilibrium were avoided for measurements, since the production rate then would increase with time.

The following presented production rates in TABLE 4.1 are cumulative ones, except if they belong to the "shielded" group, where no parents can contribute to the production. The half-lives of the individual isomers are listed, as well as the half-lives of the parents contributing to the activity of the isotopes of interest. Ionization efficiencies, which are an important source of uncertainty in the calculations, are also listed below.

Some isotopes, such as Br, were not detected or had a low not measurable release. Data for Kr was not available, because of experimental problems during collection.

TABLE 4.1 Nuclides, half-lives and production rates from off-line γ spectroscopy in the ISOLDE experiment

Nuclide	Nuclide t$_{1/2}$ [39]		Parent	t$_{1/2}$	Nuclide Prod. Rate atoms/ μC	Stat. err.	Ioniz. Eff.

Target at 600 °C

^{190}Hg	20 m		^{190}Tl	2.6-3.7 m	**3.03E+10**	1.14E+09	4 %
^{192}Hg	4.9 h		^{192}Tl	9.6-10.8 m	**7.66E+10**	1.72E+09	4 %
^{193g}Hg	3.5 h		^{193}Tl	2.1-22.6 m	*3.13E+10*		
^{193m}Hg	11.1 h	IT^p		(2.1-22.6 m)*	*3.63E+10*		
^{193}Hg					**6.75E+10**	1.24E+09	4 %
^{195g}Hg	9.5 h		^{195}Tl	1.13 h	*5.03E+10*		
^{195m}Hg	40 h	IT^p		(1.13 h)	*2.54E+10*		
^{195}Hg					**7.57E+10**	8.45E+08	4 %
^{197g}Hg	64.1 h		^{197}Tl	2.84 h	*1.21E+11*		
^{197m}Hg	23.8 h	IT		(2.84 h)	*2.06E+10*		
^{197g}Hg					**1.42E+11**	9.14E+09	4 %
^{203}Hg	46.59 d		^{203}Au	1 m	**7.42E+09**	1.39E+08	4 %
^{120}Xe	40 m		^{120}Cs	57-64 s	**3.17E+08**	8.16E+06	2.8 %
^{121}Xe	38.8 m		^{121}Cs	2-2.6 m	**2.98E+08**	5.76E+06	2.8 %
^{122}Xe	20.1 h		^{122}Cs	21s -4.2 m	**7.42E+08**	2.89E+07	2.8 %
^{123}Xe	2.08 h		^{123}Cs	5.9 m	**6.82E+08**	1.50E+07	2.8 %
^{125g}Xe	16.9 h		^{125}Cs	45 m			
^{125m}Xe	57 s	IT		(45 m)			
^{125}Xe					**1.46E+09**	3.48E+07	2.8 %
^{127g}Xe	36.4 d		^{127}Cs	6.25 h			
^{127m}Xe	70 s	IT		(6.25 h)	*not measurable*		
^{127}Xe					**1.28E+09**	3.01E+07	2.8 %
^{133g}Xe	5.25 d		^{133}I	20.8 h	*6.32E+06*		
^{133m}Xe	2.19 d	IT		(20.8 h)	*1.14E+07*		
^{133}Xe					**1.77E+07**	1.12E+06	2.8 %
^{120g}I	1.35 h		^{120}Xe	40 m			
^{120m}I	53 m			(40 m)	*not observed*		
^{120}I					**1.49E+08**	4.77E+06	2.8 %
^{121}I	2.12 m		^{121}Xe	38.8 m	**2.04E+08**	7.64E+06	2.8 %
^{123}I	13.2 h		^{123}Xe	2.08 h	**2.01E+08**	8.49E+06	2.8 %

¹²⁴I	4.15 d	¹²⁴Xe	stable	**8.04E+07**	1.95E+06	2.8 %
¹²⁶I	13.11 d	¹²⁶Xe/¹²⁶Te	stable	**6.46E+07**	2.13E+06	2.8 %
⁴¹Ar	1.83 h	⁴¹Cl	38.4 s	**1.14E+09**	2.6E+07	0.33 %
²⁰⁵At	26.2 m	_indep. prod.**_		**1.25E+07**	6.88E+05	4 %
²⁰⁶At	29.4 m	"		**1.06E+07**	2.36E+05	4 %
²⁰⁷At	1.8 h	"		**3.59E+07**	6.54E+05	4 %

Target at 400 °C

²⁰⁶At	29.4 m	"	**1.37E+06**	3.13E+04	4 %
²⁰⁷At	1.8 h	"	**1.18E+07**	3.10E+05	4 %
²⁰⁸At	1.63 h	"	**8.99E+06**	2.16E+05	4 %
²⁰⁹At	5.4 h	"	**2.71E+07**	6.31E+05	4 %
²¹⁰At	8.3 h	"	**4.34E+07**	5.76E+05	4 %

_____

g = ground state, e.g. ^{193g}Hg
m = metastable state, e.g. ^{193m}Hg
IT: total internal transition of the metastable state of the isotope
IT^{p}: partly internal transition of the metastable state of the isotope
*($t_{1/2}$): maximum 5 % parent decay to nuclide isomer
** independent production, no population from parents

The production rate of ground state ^{g}X and metastable state ^{m}X often varies, as can be seen above. This is due to different half-lives and different population of the decaying parents. The total production rate (bolded) for a specific isotope is presented as the sum of ground state and metastable state production rates.

Here follows an individual discussion for the elements: mercury, xenon, iodine, astatine and polonium measured with the off-line gamma spectroscopy.

4.3.1 Mercury

Some mercury isotopes, [197]Hg, [197m] Hg, [203]Hg, are expected to contribute with a big part of the total radiotoxicity during operation of LBE target. [34] The mercury is produced in large quantities because it is a relatively close neighbor to lead and bismuth. To trap any released mercury in the MEGAPIE target a condensation trap is designed to and a chemical reaction trap with a solid element is under consideration.

Mercury can be a direct spallation remnant, if the target element has evaporated charged light nuclei, for example alpha particles or it is formed by the decay of a parent isotope of Tl. As mentioned, there was no release of thallium at 600 ºC in the ISOLDE experiment. In accordance with data, no thallium is released from a LBE target for temperatures under 1000 ºC. [40] Most thallium isotopes have half-lives in the range of minutes or some hours, meaning that they have already decayed into mercury isotopes, through β^+ decay or electron capture. An exception in decay mode is [203]Hg which is produced from [203]Au, through β decay. Data of release of mercury isotopes around mass number 190 have shown a release efficiency of about 99 %. [41] See FIGURE 4.1.
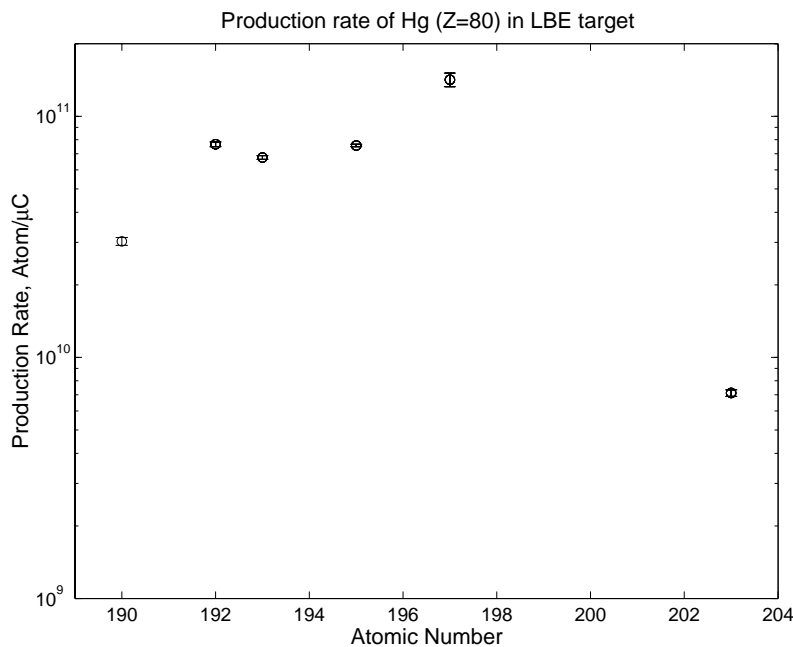

FIGURE 4.1 Measured production rates of Hg isotopes,
statistical errors plotted as bars

[194]Hg is not measured with the off-line gamma spectroscopy, because it is not a γ emitting isotope. The FC method was used instead. One can assume that the production rate of the isotope is in the same order as the neighboring isotope. [194]Hg has a long half-life (520 a) and decays into the hard γ ($E_\gamma > \sim 1$ MeV) emitting isotope [194]Au before it

decays to a stable isotope.

^{203}Hg is the only measured isotope on the right side of the stability line. Since the spallation reaction favors production of the neutron deficient isotopes it is in agreement with the lower production rate measured in the experiment and seen in FIGURE 4.1.

^{190}Hg has a half-life of 20 minutes, which is a relative short half-life for detection with the off-line γ spectroscopy and where there is a risk that some of the isotope nuclei has time to decay before it has diffused out of the target. This reduces the amount of ions transported through the mass spectrometer and finally bombarded on to the Aluminum foil. This would probably be a minor effect, since the release efficiency for mercury is in the order of some minutes.

For ^{197}Hg, ^{195}Hg and possibly ^{193}Hg, metastable states have rather long half-lives, see TABLE 4.1. Because of that, even if equilibrium is reached for the parents (Tl), the metastable and the ground state are not in equilibrium with each other, and this will somehow affect the measured production rate of the ground states. This effect is difficult to estimate since the Monte Carlo codes do not treat properly the formation of metastable states. One can compare the sum of the ground state + metastable state from the measurement and the Monte Carlo, and if the results are consistent with each other and in line with the other isotopes where this issue does not exist, it is safe to conclude that the measurements are reliable. Moreover, from the correct measurement of the production rate of the metastable state (which is in equilibrium), it is possible to correct the measured production rate of the ground state (not in equilibrium) and deduce the total cumulative production rates in equilibrium conditions. This correction is the subject of a further analysis and it is not presented in this thesis.

4.3.2 Xenon

Xenon as well as krypton is produced through fission by high energy neutrons. The xenon isotopes are either not or nearly not populated by parent decay or have short-lived parents that are not released from the target. Release of caesium would first be measurable at temperatures greater than the 600 ºC that was used during the experiment [41]. Xenon is expected to be totally released, being a noble gas. See FIGURE 4.2.

Three of the measured isotopes are nearly not populated by parent decay; ^{125m}Xe, ^{127m}Xe, and ^{133m}Xe. The two first metastable isotopes have very short half-lives of 57 s and 70 s both decaying through internal transition. The metastable states decay directly to the ground state. If the release function is instantaneous the contribution will not be constant, because of the radioactive decay of the parents. The release function is dominated by the long time component, making the release of the considered isotopes nearly constant, even with a pulsed beam. ^{133m}Xe is nearly not (< 5 %) populated by its parent. ^{133m}Xe is a metastable state that has a rather long half-life (2.19 d) and has during experiment not totally reached equilibrium and this will somehow affect the measured production rate of the ground states. This effect is difficult to estimate since the Monte Carlo codes do not

treat properly the formation of metastable states as discussed in chapter 4.3.1 about mercury.
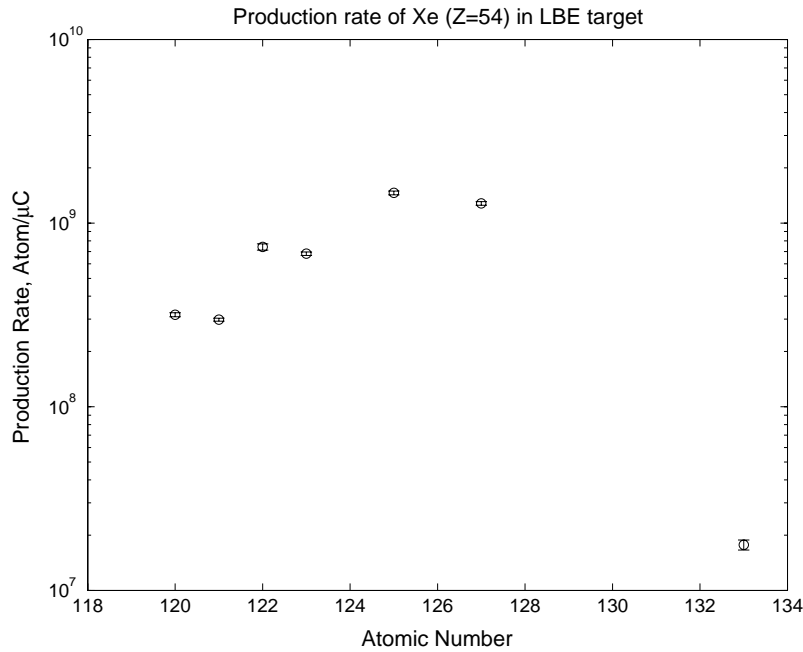

FIGURE 4.2 Measured production rates of Xe isotopes,
statistical errors plotted as bars

The other xenon isotopes have parents that are short-lived and not released. Cesium is the parent to xenon and was not released during test with masses of 130 and 132. That means that cesium will decay inside the target and be released as xenon.

4.3.3 Iodine

Iodine has two different parents, for iodine up to mass number 126 the parent is xenon. ^{124}I and ^{126}I both have stable parents, i.e. they do not have any parent decay contribution in the production. The contribution from decaying parent will be minimal, because the xenon is completely released from the target. However, for the isotopes ^{120}I, ^{121}I and ^{123}I a contribution from the decay of the corresponding xenon isotopes deposited onto the foil. ^{120}I is the only iodine isotope measured with an isomer. The abundance of the isomer is very low and its gamma lines difficult to measure with the acquisition program, i.e. measured production rate from ^{120}I is from ^{120g}I only.

Iodine is expected to partly be trapped inside the target, because it has a slightly lower volatility than xenon. See FIGURE 4.3.
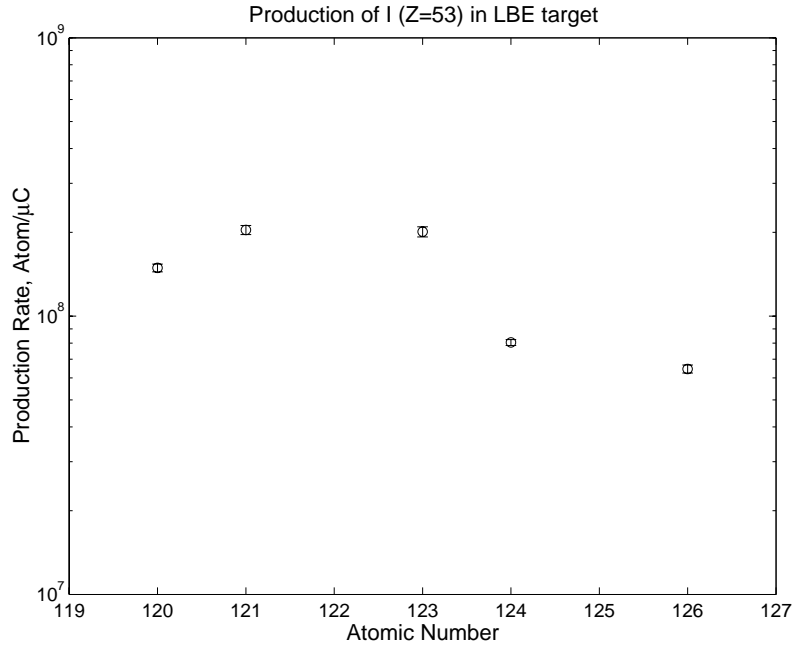

FIGURE 4.3 Measured production rates of I isotopes,
statistical errors plotted as bars

4.3.4 Astatine

Astatine is an element without any stable isotopes. The longest living isotope, ^{210}At, has a half-life of 8.3 h. Astatine is a result of secondary reactions in the spallation target, because of the presence of bismuth in the LBE target. There are a number of possible reactions that can lead to astatine production (4.1), (4.2), (4.3), (4.4) and (4.5). One of the most likely reaction modes is an α-particle interacting with a bismuth nucleus and emitting neutrons. [42] The α particle is produced through fragmentation or α decay from numerous elements, i.e. Hg, Au, Pb and Th.

$$^{209}\text{Bi} \, (^4\text{He, xn}) \, ^{213-x}\text{At} \tag{4.1}$$

Since also ^{3}He-production occurs in the spallation reaction, the following reaction could be possible:

$$^{209}\text{Bi} \, (^3\text{He, xn}) \, ^{212-x}\text{At} \tag{4.2}$$

Experiments have shown a peak cross section of $\prec$ 1barn for the above reactions in infinitely thick targets, where the production of secondary particles is high. [42]

As mentioned in the spallation chapter pions are produced in a thick target for proton beam energy higher than 400 MeV, leading to a possibility of following reactions:

$$^{209}\text{Bi} \,(\pi^+, \pi^- \, xn) \,^{209-x}\text{At} \qquad\qquad\qquad \textbf{(4.3)}$$

$$^{209}\text{Bi} \,(\pi^-, X) \, \text{At} \qquad\qquad\qquad\qquad\qquad \textbf{(4.4)}$$

$$^{209}\text{Bi} \,(p, \pi^- \, xn) \,^{210-x}\text{At} \qquad\qquad\qquad \textbf{(4.5)}$$

The latter reaction is likely to be the dominating one, given the high proton flux. This is confirmed by calculations performed with MCNPX [43]

Astatine will not be a problem for future disposal or for operation of the MEGAPIE, because of the low production rate. A problem with astatine production can rise from the decay into long-lived polonium isotopes.

The following two figures, FIGURE 4.4 and FIGURE 4.5 show the measured production rates for astatine at different target temperatures.
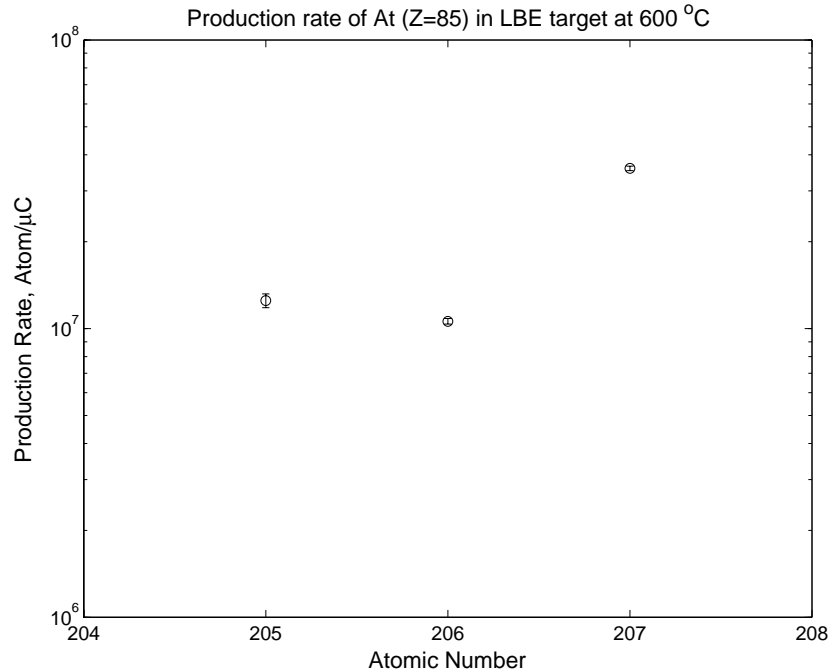

FIGURE 4.4 Measured production rates of At isotopes
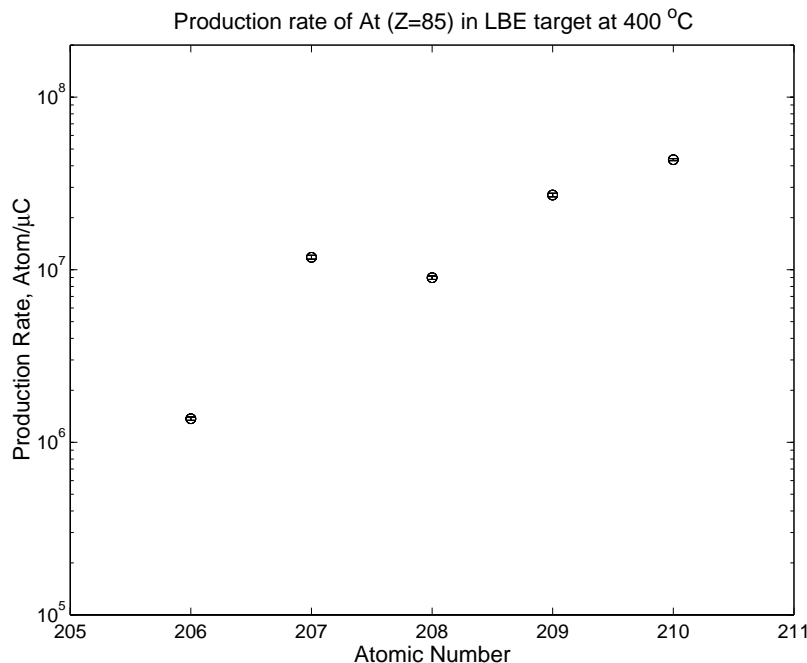at target temperature 600 °C

FIGURE 4.5 Measured production rates of At isotopes
at target temperature 400 °C

4.3.5 Polonium

In LBE target the buildup of ^{210}Po is an important issue to be considered in order to assess the polonium contamination in the target [1]. ^{210}Po has the highest radiotoxicity among the volatile elements, and is expected to be produced in the LBE reactions, by (p, γ) and (n, βγ) –reactions with ^{209}Bi and decay of astatine. Because of the presence of bismuth, the ^{210}Po production rate in the LBE target is 1000 higher than in a target consisting of only lead. Evaporation rates in LBE of Polonides are on the other hand much lower than in liquid lead targets. ^{209}Po and ^{208}Po are like ^{210}Po α-emitting isotopes with long half-lives and contribute strongly to the radiotoxicity of the target after one year of operation. Within one year of operation and employing a proton beam of 1 mA a total production of 2 grams Po will be produced in MEGAPIE. [44]

In MEGAPIE with its high thermal neutron flux, the production of ^{210}Po by neutron absorption in ^{209}Bi would, of course, be much more important than in ISOLDE.

Polonium is not as volatile as some of the noble gases and is predicted to form polonides and eventually escape the target as PbPo molecules. [34] This is difficult to detect, since the molecule breaks up into ions in the ion source.

The measured production rate of polonium changes for different measurement times. This indicates that the measured polonium mainly is a decay product of astatine, since astatine decays with half-lives up to 8.3 hours (^{210}At). Earlier experiments have shown that

polonium has a slow evaporation from a LBE target first at temperatures higher than 600 ᵒC. [44] This supports that the polonium released in the ISOLDE experiment is not the independently and directly produced polonium, but only the decay product of already released astatine.

This can also be verified by comparing polonium production rates at different times. The production rates for [207]Po were $7.1 \cdot 10^6$ atoms/μC after 1 hour and $8.9 \cdot 10^6$ atoms/μC after 8.6 hours. The corresponding production rate for astatine was $2 \cdot 10^6$ atoms/μC. By using following equation, Eq (4.6) [20], where N_0 is the astatine at collection time (assuming no initial polonium), the production rate of polonium N_2 can be calculated.

$$N_2(t) = N_0 \frac{\lambda_1}{\lambda_2 - \lambda_1}(e^{-\lambda_1 t} - e^{-\lambda_2 t})$$

$$N_2(0) = 0$$

(4.6)

From the equation one gets a production rate of polonium of $6.5 \cdot 10^6$ atoms/μC and $9.4 \cdot 10^6$ atoms/μC.

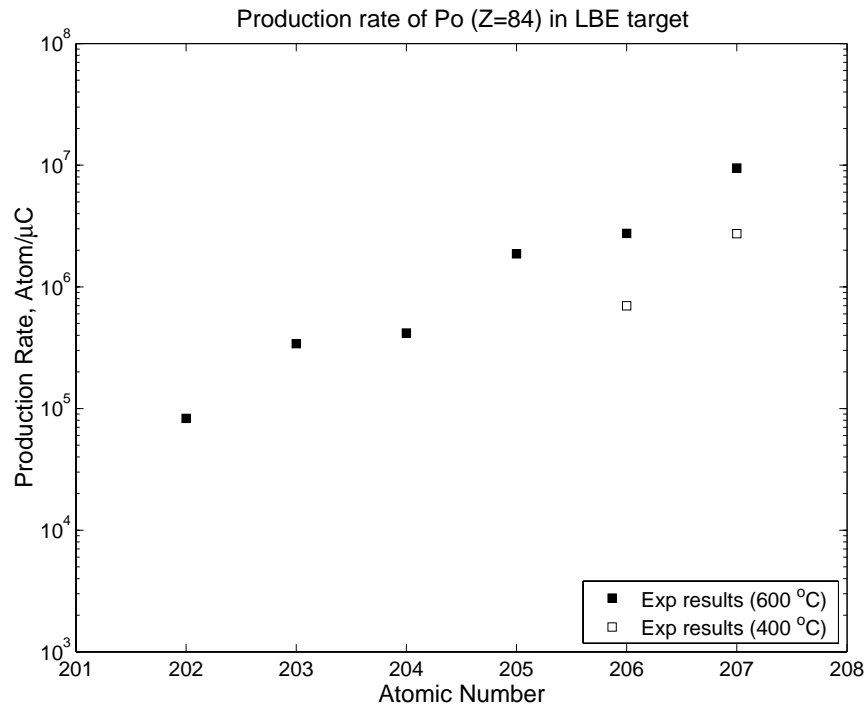The following figure, FIGURE 4.6, shows the measured polonium production rates at two different temperatures.

FIGURE 4.6 Measured production rates of Po isotopes
at target temperature 600 °C and 400 °C

4.3.6 Helium

Helium is one of the few elements presented from the FC measurements. The highest production rates in the LBE target, when irradiated with protons of 1.4 GeV, come from H and He atoms. Predictions with Monte Carlo code have shown production rates of 1.94 H atoms per proton and 0.208 He atoms per proton [34] In the ISOLDE experiment it is possible to measure the ^{4}He production. MCNPX code predicts $5.5 \cdot 10^{12}$ atoms/μC and the measured production rate in the ISOLDE target was $2.8 \cdot 10^{11}$ atoms/μC. When the experimental result is extrapolated to the MEGAPIE target, the result is a production of 1 liter per year of ^{4}He.

The measurement of hydrogen is more difficult because of the background of non-radiogenic rest gas in the target.

4.4 DISCUSSION

If there is no release of a specific nuclide at 600 ºC then one can assume to observe nothing at 400 ºC. A release of a certain isotope at 600 ºC motivates to do experiments at 400 ºC. At both temperatures the target states are liquid and therefore also have similar chemical properties.

There are some advantages of a thick target experiment like the one at ISOLDE in comparison with a thin target experiment like experiments done at GSI (research institute for heavy accelerated ions in Germany). All secondary reactions in the target are present, which one would not have been able to observe in a thin target experiment. In a thin target the secondary products escape the target directly, while in the thick target the secondary products are trapped and induce secondary reactions. In the GSI experiments they could not detect α-particles and also no isomers.

To simulate a long irradiation time of the MEGAPIE target, the temperature of 600 ºC were chosen. In this way the diffusion reaction is accelerated and reduces the risk that some isotopes, not seen at 400 ºC, are released from the target after a month or a year of operation. One can be sure that if there is no release of a specific element in an experiment with a much smaller target at a much higher temperature (600 ºC), there will not be any release of that element after a year of operation in the MEGAPIE target. If there had been a transition phase (for example when liquid becomes solid) one would not have been able to scale.

Since the size of the target in the MEGAPIE is much bigger, the results from the ISOLDE experiment has to be extrapolated to fit the MEGAPIE target. There might be a difference in the release function of some elements, since the target differ in size. Larger targets mean a smaller release of specific isotopes, because of smaller surface/volume ratio and longer transport times inside the target. [40] Therefore is it possible that some elements, which are easily released in the ISOLDE experiment, partly will stay inside the MEGAPIE target. The production rate is a product of the beam intensity, the cross

section and the effective target thickness, which makes it possible to re-scale the measured ISOLDE production rates to production rates in MEGAPIE target.

Calculated values of stable gases indicated a production of about 1 liter per month, mainly stable H and ^{4}He. The radioactive gas will be handled by a specially designed cover gas system.

A bigger concern is the release of the volatile astatine, which will decay to the very radiotoxic polonium. This is a topic for further investigations, to define how much of the polonium that is independently produced and how much is a decay product from astatine.

Bromine was not observed during measurements, concluding that bromine is trapped inside the LBE at a temperature of 600 °C. During measurements with krypton, which is like xenon produced through fission, some problems influenced the collection and no reliable results are presentable. Just one isotope for argon and neon respectively were measured, which is a too week basis for further discussion. Also this will be a subject for the follow-up experiment as well as further experiments with target temperature at 400 ºC.

4.4.1 Statistical uncertainties

The statistical errors are directly calculated by the acquisition program GENIE. In all plots the statistical errors are included. No systematical errors are included in the plots.

4.4.2 Systematic uncertainties

The biggest systematic errors come from the ionization efficiency. In our experiment, ionization efficiencies were measured by leaking known amount of He, Ne, Ar, Kr, and Xe gases. However, measurements preformed at different times showed variations in the efficiencies of the order of 30%. Therefore we assume these uncertainties in our results. This will be further investigated in the next experiment. Concerning heavier elements (At, Hg), the ionization efficiency could not be measured, but it was extrapolated from the ones measured for the noble gases, from data from the literature.

5 MONTE CARLO SIMULATIONS

5.1 CALCULATIONS

The Monte Carlo calculations were done using the code FLUKA. For the calculations a simple model of the LBE target was used, see FIGURE 5.1. A cylinder filled to 75 % with lead bismuth and bombarded by a 1.4 GeV proton beam was implemented. See also Beam and Target chapter for specific parameters. The isotope production rate in the target was calculated in atoms per protons and was converted into atoms per µC (1 µC= 6.24 · 10^{12} protons).
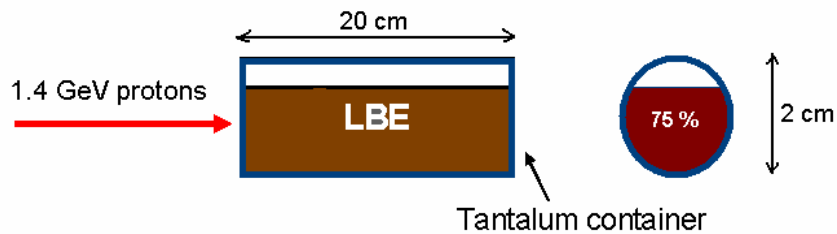

FIGURE 5.1 Calculation model used in the Monte Carlo programs

Different parameters have to be defined in the input file to FLUKA, among others choice of materials, type of particles and type of scoring. The input file is divided into different parts, were the beam parameters, the target geometry and respective material assigned to a specific region in the target, are defined. In the end of the input file a so-called tally card must be defined. The USRBIN scores a distribution in a regular spatial structure, also called a binning, independent of the geometry were the scoring is taking place.

At the end of the input file the number of simulated protons has to be defined. In most runs of the program 1 or 2 millions of protons were chosen. The number of events was chosen to give satisfactory statistical accuracy.

For every run of the input file with chosen parameters, FLUKA makes a succession of calculations, giving the possibility to check whether the calculations are statistically significant.

By using the additional code ORIHET3 the cumulative production rates were calculated for some selected elements.

5.2 RESULTS AND COMPARISON

Here follows a comparison of the measured experimental results and the calculated results from FLUKA. A comparison is made for mercury, xenon, iodine and astatine.

5.2.1 Mercury

Following figure, FIGURE 5.2, shows a comparison of experimental results and MC calculated results.
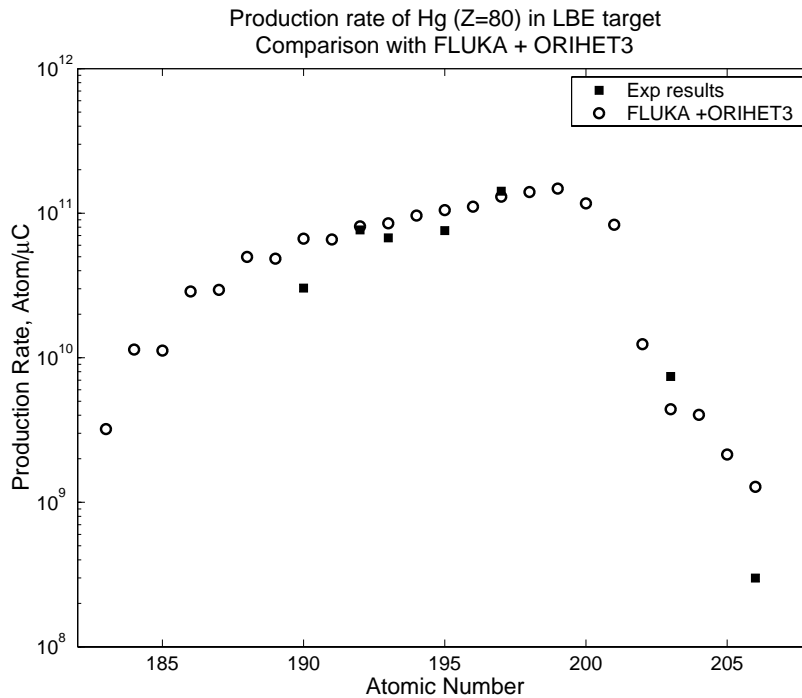

FIGURE 5.2 Comparison experimental results and MCC for Hg isotopes

The experimental production rates are all in good agreement with the calculated FLUKA results, which is of importance for FLUKA benchmark. A deviation (a factor of two) of [190]Hg from FLUKA results has already been discussed for the experimental results. As seen in the FIGURE 5.2, Hg has a high production rate in LBE, peaking at a value higher than 10^{11} atoms per μC. Hg is the most important radioisotope for MEGAPIE and contributes to a big part of the activity of LBE. The ionization efficiency used for Hg was 4 %, and one should note that it was not measured in the experiment.

5.2.2 Xenon

The following figure, FIGURE 5.3, shows the comparison of experimental results and MC calculated results for xenon.

Production rate of Xe (Z=54) in LBE target
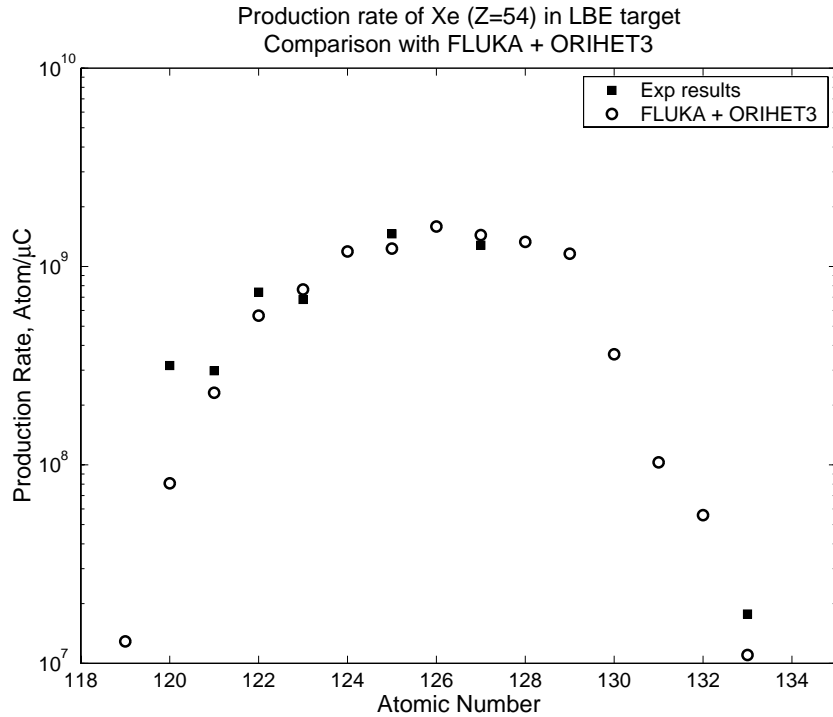Comparison with FLUKA + ORIHET3

FIGURE 5.3 Comparison experimental results and MCC for Xe isotopes

Also here are the experimental results in good agreement with the calculated FLUKA results. The difference in results for ^{120}Xe could be due to experimental factors, or even a wrong prediction from the FLUKA code.

An ionization efficiency of 2.8 % was used, though one should note that there were some uncertainties in that measurement.

Since xenon is a fission product (and not a direct spallation product like mercury) the production rate is much lower.

5.2.3 Iodine

The following figure, FIGURE 5.4, shows the comparison of experimental results and MC calculated results for iodine.
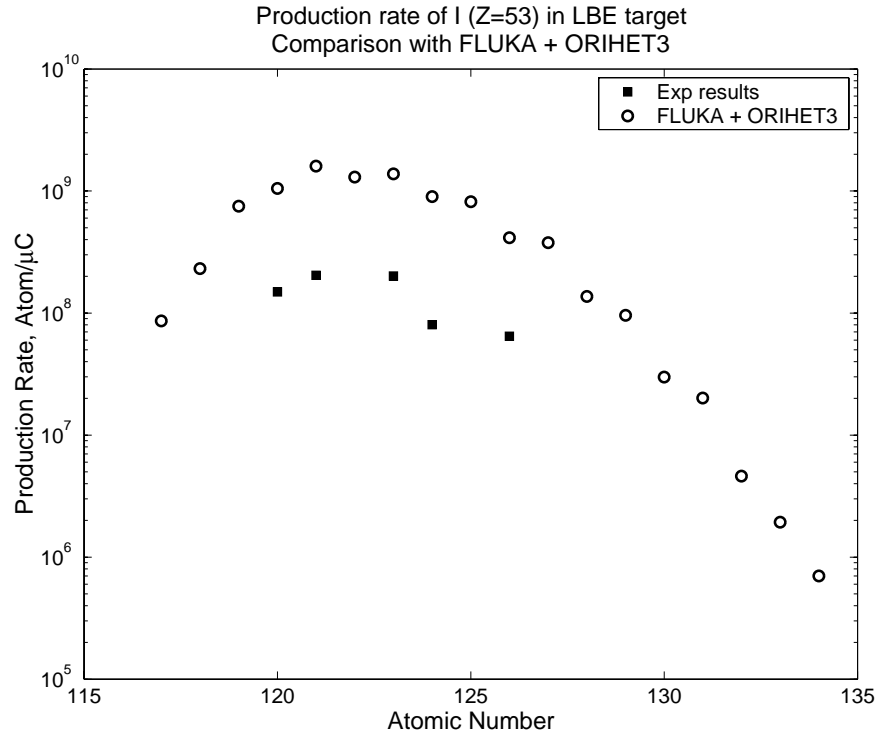

FIGURE 5.4 Comparison experimental results and MCC for I isotopes

The experimental results are here one order lower than the calculated FLUKA results. They are, however, following the same shape as the calculated ones. Iodine is, as mentioned before, slightly less volatile than xenon. The Monte Carlo code is smooth with no big changes between two close elements and since the release of xenon is 100 % a comparison can be made between the xenon and iodine. This leads to the conclusion that not all iodine is released from the target.

The FLUKA results is therefore a good estimation of the really production rate of iodine in LBE target. Of special concern are the two isotopes ^{125}I and ^{131}I, which are highly radiotoxic.

5.2.4 Astatine

The following two figures, FIGURE 5.5 and FIGURE 5.6, show the experimental and calculated astatine production rates with different target temperatures. Astatine was not expected at the experiment and resulted in some new ideas for future experiments.
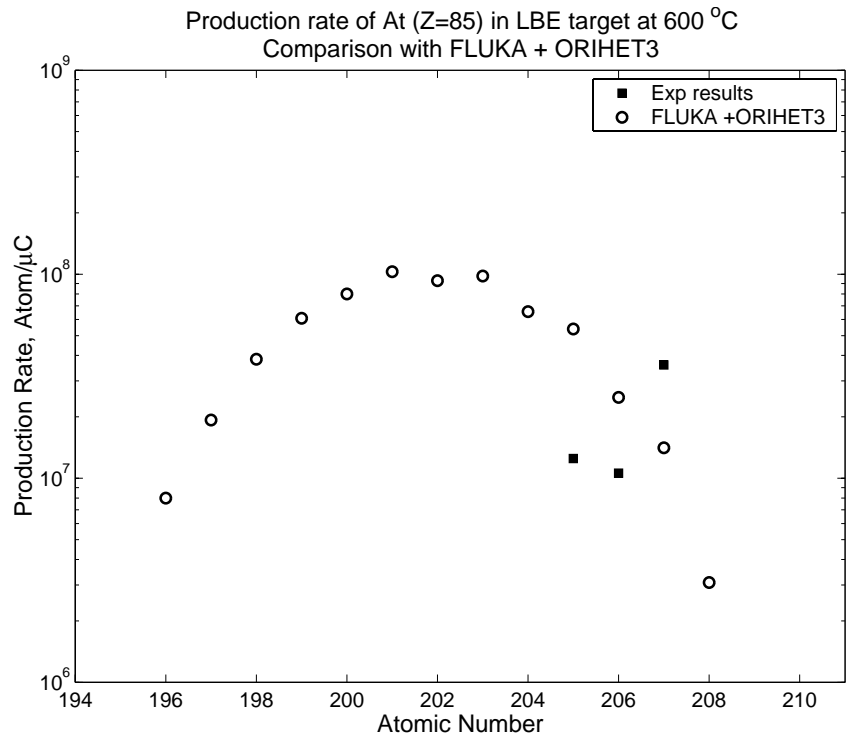
FIGURE 5.5 At production obtained from experiments at 600 °C, comparison with FLUKA
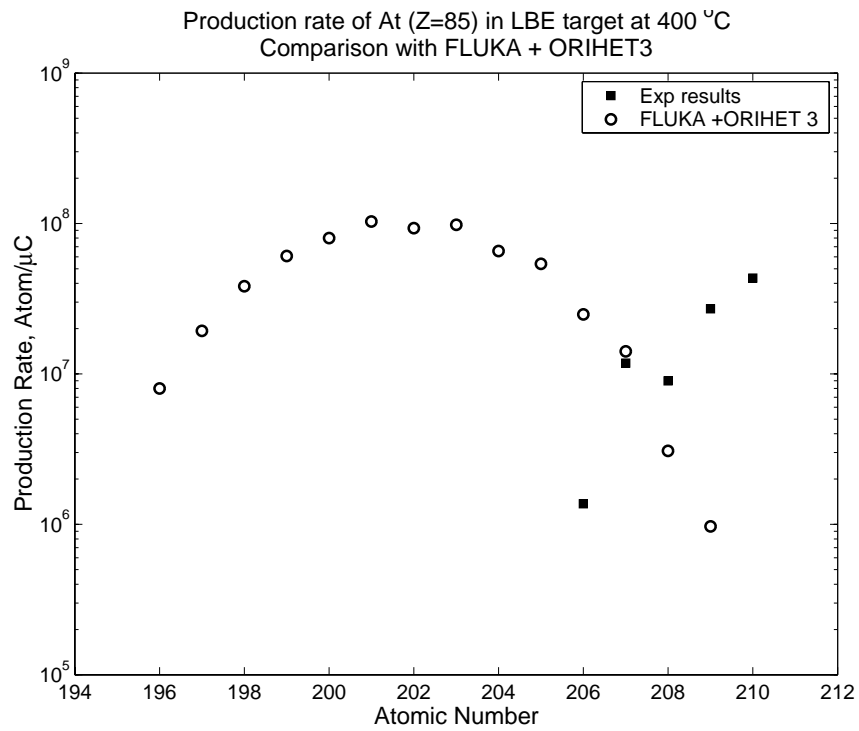

FIGURE 5.6 At prod obtained from experiments at 400 °C, comparison with FLUKA

Astatine is as mentioned before produced through four different mechanisms, which may all not be included in the Monte Carlo codes. The discrepancy from the calculations has to be further investigated. Astatine is not an issue for MEGAPIE, because of the low production rate.

5.2.5 Polonium

As shown before, the measure Po comes in fact from the At decay. Therefore a comparison with MC data does not make sense since in fact the observed Po is just a decay product from the At.

5.3 DISCUSSION

In this thesis results of relevance in the frame of the ADS development have been obtained. Here in the following are discussed:

Cumulative production rates of At, Hg, Xe, I isotopes, and of ^{4}He have been measured. The data show trends as a function of the atomic mass in line with the expectations from the physics of the isotope spallation production.

The measurement of Hg is of particular interest since it is a highly volatile elements and it is expected to contribute very significantly (10 to 15 %) to the total radionuclide inventory of the MEGAPIE target. When comparing the measured cumulative production rates to MC calculations, a good agreement is found. This result is of great importance since the neutronic design of the MEGAPIE, and in particular of the system to handle the production of Hg, was based on the Monte Carlo expectation values.

The result for Xe production is of interest for a different reason. While the radiotoxicity of the Xe isotopes is certainly lower than for the Hg, the results obtained allow for a test of the models used in the Monte Carlo simulations which calculate the production of the nuclei in the fast neutron fission region. Therefore these results will allow for a benchmark of the most currently used models in the nuclear calculations, such as FLUKA and MCNPX.

Some of the I isotopes have also high radiotoxicity, and the results obtained for the I release will give a precious indication on the level of the I release that can be expected in a target of the size of MEGAPIE.

Finally, the At production is a very interesting results, not only for MEGAPIE (it confirms that the expected release rates of At are very low, and not of concern). More importantly, this result is of interest for ISOLDE, as it is the first time that At beams are observed, and it is at present a subject of discussion if the production rate is high enough to allow for possible experiments with At beams.

The good agreement with Monte Carlo calculated results makes future calculations with FLUKA for the MEGAPIE project a reliable method to predict the volatiles produced in the LBE target.

6 FUTURE WORK

In the follow-up experiment, focus will be on making new measurements with some of the volatiles, at different beam energy (1 GeV instead of 1.4 GeV) and at the target temperature of 400 ºC. With a lower target temperature, one can be sure to avoid delayed isotopes, possibly influencing following isotope measurements. Since iodine was not fully released at target temperature 600 ºC, it is of great interest to investigate if iodine is totally trapped inside the target or what amount that will be released at 400 ºC. Short and long release components will be further investigated as well as ionization efficiencies. A small leakage of the ionization source was a probable explanation of the different ionization efficiencies measured for xenon. Measurements will be done to define the actual ionization efficiency for xenon.

A THE PERIODIC TABLE

The following periodic table, TABLE A, summarizes the detected isotopes from the ISOLDE experiment.

Group	1	2		3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
Period																			
1	1 H																		2 He
2	3 Li	4 Be												5 B	6 C	7 N	8 O	9 F	10 Ne
3	11 Na	12 Mg												13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
4	19 K	20 Ca		21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr
5	37 Rb	38 Sr		39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe
6	55 Cs	56 Ba	*	71 Lu	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	**	103 Lr	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg	112 Uub	113 Uut	114 Uuq	115 Uup	116 Uuh	117 Uus	118 Uuo
*Lanthanoids			*	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb		
**Actinoids			**	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No		

Red – target components
Blue – measured in the off-line γ spectroscopy, isotopes found
Yellow – measured in the off-line γ spectroscopy, isotopes not detected/ not released
Green – largest production rate of the volatiles from the LBE target

TABLE A Periodic table summarizing the detected elements in the ISOLDE experiment.

B ABBREVIATIONS

ADS   Accelerator Driven System
CE   Conversion Electrons
EC, ϵ Electron Capture
FC   Faraday Cup
FLUKA   Monte Carlo code used mainly at CERN, Geneva
GPS   General Purpose Separator
GSI   Gesellschaft für Schwerionenforschung mbH, Darmstadt
HRS   High Resolution Separator
INC   IntraNuclear Cascade, the first step of the spallation reaction
INFN   Instituto Nazionale di Fisica Nucleare
ISOL   ISotope On-Line separation
ISOLDE   Facility at CERN, where the ISOL-technique is used
IT   Isomeric Transition or Internal Transition
LANL   Los Alamos National Laboratory
LBE   Lead Bismuth Eutectic
MCC   Monte Carlo Code
MCNPX   Monte Carlo code, developed at LANL
MEGAPIE   MEGAwatt PIlot target Experiment
ORIHET   Complementary code to FLUKA, for calculating activity build-up
                    and decay
PSB   PS-Booster, a synchrotron at CERN
PSI   Paul Scherrer Institute
RIB   Radioactive Ion Beam
SINQ   Swiss Spallation Neutron Source
TS   Tape Station or off-line γ spectroscopy

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