

Maximizing the radionuclide collection efficiency at CERN-MEDICIS:

A case study on terbium collections and sputtering

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Preface

From the moment I decided to study physics, to this moment of completing my master's thesis, the journey has been incredible. It took five years of hard work and commitment, but here I am, proud to have completed a master's thesis that I genuinely enjoyed working on.

The primary objective of this thesis was to optimize future collections of terbium at CERN-MEDICIS (Switzerland). The aim was to comprehensively examine the production and implantation of terbium, aimed at understanding the processes and factors that influence the collection efficiency. However, writing this thesis would not have been possible without the invaluable contributions of numerous people, whom I would like to thank.

First and foremost, I wish to express my gratitude towards professor Thomas Cocolios for guiding me through this thesis and for providing me with the topic and main outline of this master's thesis. Furthermore, I am extremely grateful for the opportunity to visit CERN. Being able to be on-site and seeing the operations at the MEDICIS facility has provided me with invaluable insights and has increased my understanding of the collections. Moreover, thanks for letting me present my (preliminary) results at the LISA conference. The experience of attending the conference and engaging with fellow enthusiasts of nuclear physics was truly enjoyable. Additionally, thanks for sparking my interest for nuclear physics from the second bachelor onwards. Creating the movies for AN3 played a significant role in igniting my passion for nuclear physics and I am truly thankful for that experience.

Furthermore, I would like to express my sincere thanks to Michael and Wiktorina for guiding me through every step of this process, for guiding me in the right direction and for being there to answer all of my questions. Their advice and feedback helped me navigate through the challenges of writing a thesis, thank you for that. I have enjoyed the interesting discussions a lot! Moreover, I am very grateful for the pieces of code that I received from Michael. Without any doubt, I could not have wished for a better duo of mentors than the two of you.

Next, I want to thank Cyril Bernerd and Charlotte Duchemin for performing the experiments at MEDICIS and for providing me with the necessary data and information. Moreover, I am indebted to the whole research group of Professor Cocolios for creating a supportive and collaborative atmosphere. Special thanks goes to Jake for pointing out and helping with the exploration of recoil-sputtering.

I also want to thank my friends for supporting me throughout this journey, for being there to lend an ear during moments of frustration and for sharing memorable experiences with me! Especially, I would like to thank Hanne Baeyens and Marie Cloet for proofreading parts of my thesis. Als laatste wil ik graag mijn ouders en broer bedanken. Jullie hebben mij alle kansen gegeven om mijn dromen na te jagen en mij aangemoedigd om door te zetten. Dankjewel!

After having expressed these genuine acknowledgements, I have one final message to share with you, dear reader: Enjoy!

Marie, June 2023

Contribution statement

I would like to express my gratitude towards the people who carried out several of the measurements presented in this thesis. Their contributions were invaluable and are greatly appreciated. Therefore, I want to thank those people here for their contributions.

During the collections discussed in Part I, I had the opportunity to be present at CERN-MEDICIS for the collection of July 15. However, the measurements taken during the collections were performed by the team of researchers at MEDICIS. Especially, I would like to thank Cyril Bernerd for providing me with all necessary data to analyze these collections. Additionally, I would like to thank Charlotte Duchemin, who directed me towards additional data that proved to be valuable for the analysis of the collections, including the dose rate measurements and the irradiation conditions.

The activity was determined using the activity spectra acquired with the Kromek CZT detector. The energy calibration of the Kromek CZT detector was performed by Kaat Spoormans and the efficiency measurements of the Kromek CZT detector were performed by Pinelopi Christodoulou.

In Part II, the high-current implantations were performed by Bart Caerts. Moreover, the Rutherford backscattering spectrometry measurements were performed by Qiang Zhao.

In Part III, the α -spectroscopy measurements for the recoil-sputtering of ^{225}Ac were performed by Jake Johnson.

Scientific summary

This thesis is focussed on maximizing the efficiency of medical radionuclide collections at CERN-MEDICIS. More specifically, a case study on terbium was performed.

In particular, radionuclide collections begin with the production of the radionuclide. Here, a target is irradiated with a beam (often protons) in order to induce nuclear reactions. In that way, the radionuclide of interest is produced amongst others. Separation of the isotope of interest from other nuclei is required to ensure the purity of medical samples. Here, both elemental selection (with laser resonance ionization) and mass selection (by using a mass separator) are combined in order to obtain a high specific activity. Thereafter, the purified radionuclide beam is implanted in a foil to collect the radionuclides and to be able to transport them to a chemical facility or hospitals. During this implantation process, some isotopes are lost due to self-sputtering. All the above-mentioned steps contribute to the total efficiency of a collection, which should be maximized. Therefore, the main aim of this thesis was to investigate terbium collections and sputtering in order to maximize the collection efficiency.

The first crucial contribution is the efficiency of the mass separating method. To gain insight into this process, the terbium collections that were performed in 2022 at MEDICIS are analyzed. The targets for these collections were irradiated at an external facility and chemical separation was performed prior to mass separation to purify the samples. Using chemical separation, terbium can be separated from other elements, but no purification within the terbium isotopes can be performed. Hence, mass separation is needed and its efficiency should be maximized.

The next part focuses on implantation of the radionuclides in the collection foil, more specifically, on the self-sputtering that occurs during these implantations. In particular, self-sputtering takes place when the momentum of an impinging ion is transferred to nuclei in the collection foil, causing a nuclear collision cascade that can eject surface nuclei, i.e., these nuclei are sputtered out of the foil. As the collection proceeds, more foil material nuclei will be sputtered out of the foil, hence the original surface of the foil will approach the earlier implanted isotopes. Consequently, these earlier implanted isotopes eventually reach the surface, so that they can be *self*-sputtered from the foil. An analysis on the self-sputtering behavior has been performed by the use of TRIDYN, a Monte Carlo-based package for implantation simulations. By performing these simulations, insight in the self-sputtering process is obtained, which can, eventually, be used to reduce the amount of self-sputtering that occurs during a collection. Moreover, an experimental protocol is put forward to verify the reliability of the simulations.

The last part entails the additional time-aspect for α -decaying nuclei. In α -decaying nuclei there is an additional decay-related loss of isotopes, namely that after the decay, the daughter nucleus obtains a large recoil momentum. Hence, the daughter nucleus travels through the foil and can sputter away other implanted mother nuclei. A first approximation was obtained by combining TRIDYN and TRIM simulations.

Ultimately, this work aims to contribute to the improvement of radionuclide collections, which is crucial for the production of radiopharmaceuticals for diagnostic and therapeutic applications.

Summary for a general audience

The discovery of radioactivity in 1896 by Marie Curie and Henri Becquerel paved the way for numerous applications, including the use of radioactive isotopes in nuclear medicine to diagnose and treat cancer. In nuclear medicine, radionuclides are bound to a tracer and are injected into the body. The main function of this tracer is to guide the radionuclide towards the cancer cells. When a radioisotope decays, radiation is emitted, which can be detected to obtain an image of the tumor. Additionally, the energy that is released upon decaying can be used to damage a cancer cell.

For each diagnostic and therapeutic modality, radioisotopes with different decay characteristics are needed. Terbium is a promising element, as there exist 4 terbium isotopes with complementary decay characteristics. Hence, all 4 isotopes have their own applications for diagnostic or therapeutic purposes while their chemistry is the same. Thus, the behavior inside the body will be similar, and tracer research with one isotope can be extended to the other terbium isotopes. Collections of terbium are performed at CERN-MEDICIS (Switzerland) and often start by shooting protons on a target. As a result, terbium is produced amongst other isotopes. Upon heating of this target, terbium and other radionuclides will come out. Subsequently, terbium is accelerated, separated from unwanted particles, collected on a foil, made into a radiopharmaceutical and brought to the hospital for treatment and imaging.

Nevertheless, this production process is not optimized yet. On one hand, we aim to extract the maximum amount of terbium out of the target, while on the other hand, we strive to minimize the loss when shooting them on the collection foil. The first part of the thesis is dedicated to the former. Here, three different collections that were performed in 2022 are analyzed in order to provide insights into the production and separation process. The minimization of the loss when implanting the radionuclides on the foil is addressed in the second part: when terbium particles impinge with a large energy on the foil, their energy is transferred to other foil particles in collisions. These collisions can kick out previously implanted terbium particles, which are thus lost and therefore they cannot be used in the hospital. Due to this process, known as self-sputtering, up to a quarter of the implanted particles were lost in recent collections. Therefore, minimization of self-sputtering is essential.

Finally, an impact of delays is researched: if particles are implanted on a foil and transported towards the hospital, additional losses can occur. More specifically, when an implanted particle undergoes decay after implantation, its daughter nucleus acquires a considerable amount of energy, particularly in certain types of decay. As a result, the daughter nucleus traverses the foil and can kick out previously implanted particles. This effect was investigated in this thesis, but only minor effects are present for the current collections.

In conclusion, using radioactive isotopes in nuclear medicine is crucial for diagnosing and treating cancer. With terbium being a promising radionuclide for nuclear medicine, its production process needs to be optimized. To that end, this thesis is focused on the optimization of producing and implanting terbium.

List of acronyms

BR2	Belgian Reactor 2 (located at SCK-CEN)
CERN	European Organization for Nuclear Research (in French: Conseil Européen pour la Recherche Nucléaire)
CZT	Cadmium Zinc Telluride
FWHM	Full Width at Half Maximum
ARRONAX	Accelerator for Research in Radiochemistry and Oncology in Nantes-Atlantique
HCII	High Current Ion Planter
HIPAC	High Intensity Proton Accelerator (located at PSI)
IBA	International Business Accelerator
ICP-OES	Inductively Coupled Plasma Optical Emission Spectroscopy
IMBL	Ion and Molecular Beam Lab
IRIS	ISOLPHARM Radionuclide Implantation Station
ISOL	Isotope Separation On-Line
ISOLDE	Isotope Separation On-Line DEvice (located at CERN)
ISOLpharm	ISOL technique for radioPHARMaceuticals (located at SPES)
MELISSA	MEDICIS Laser Ion Source Setup At CERN
MEDICIS	Medical Isotopes Collected from ISOLDE (located at CERN)
MYRRHA	Multi-purpose hYbrid Research Reactor for High-tech Applications (located at SCK-CEN)
PSB	Proton Synchrotron Booster (located at CERN)
PET	Positron Emission Tomography
PSI	Paul Scherrer Institute
RBS	Rutherford Backscattering Spectroscopy
RILIS	Resonance Ionization Laser Ion Source
SIMNRA	SIMulation program for the analysis of NRA, RBS and ERDA
SMILES	Mass separation coupled with laser ionization for environmental and healthcare applications (in French : Séparation en Masse couplée à l'Ionisation Laser pour des applications Environnementales et en Santé) (located at ARRONAX)
SPECT	Single Photon Emission Computed Tomography
SPES	Selective Production of Exotic Species
SRIM	Stopping and Range of Ions in Matter
SCK-CEN	Research centre for the applications of nuclear energy (in Dutch: StudieCentrum voor de Toepassingen van de Kernenergie) (in French: Centre d'étude de l'Énergie Nucléaire)
TATTOOS	Targeted Alpha Tumour Therapy and Other Oncological Solutions (located at PSI)
TRIDYN	TRIm simulation code including DYNamic composition changes
TRIM	TRansport of Ions in Matter
VADIS	Versatile Arc Discharge Ion Source

List of most important symbols

SYMBOL	UNIT	MEANING
A	[Bq]	Activity [Becquerel]
c	[A]	Current [ampère]
Δt	[s]	Time difference [seconds]
e	[C]	Electron charge [coulomb]
E	[eV]	Energy [electronvolt]
ε	[%]	Efficiency
f_i	[%]	Fraction compared to the incoming activity
f_r	[%]	Fraction compared to the retained activity after implantation
λ	$[\frac{1}{s}]$	Decay constant [1/seconds]
N		Number of neutrons
N		Number of particles
ψ	$[\text{\AA}^{-2}]$	Fluence [$\text{\AA}\text{ngstr\o{m}}^{-2}$]
ψ_{inc}	$[\text{\AA}^{-2}]$	Incoming fluence [$\text{\AA}\text{ngstr\o{m}}^{-2}$]
ψ_{ret}	$[\text{\AA}^{-2}]$	Retained fluence [$\text{\AA}\text{ngstr\o{m}}^{-2}$]
ψ_{crit}	$[\text{\AA}^{-2}]$	Fluence for which $\psi_{ret} < 0.9\psi_{inc}$ [$\text{\AA}\text{ngstr\o{m}}^{-2}$]
σ_{Δ}	$[\text{\AA}^{-2}]$	Incoming fluence step-size of the TRIDYN simulations [$\text{\AA}\text{ngstr\o{m}}^{-2}$]
σ_{sim}	$[\text{\AA}^{-2}]$	Error on ψ_{crit} [$\text{\AA}\text{ngstr\o{m}}^{-2}$]
σ_{TRIDYN}	$[\text{\AA}^{-2}]$	Error on the retained fluence from the TRIDYN simulation [$\text{\AA}\text{ngstr\o{m}}^{-2}$]
σ_r	[$\text{\AA}$]	Standard deviation on the radius of an impinging beam [$\text{\AA}\text{ngstr\o{m}}$]
t	[s]	Time [seconds]
$t_{1/2}$	[s]	Half-life [seconds]
T	[A, °C]	Temperature [ampère, degrees celcius]
Z		Number of protons

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Introduction

According to the World Health Organization, cancer takes the second place in the death causes all over the world, thus research in both treatment and diagnostics is crucial. One of the emerging fields in cancer research is nuclear medicine. Here, the different decay characteristics are exploited for both treatment and diagnostics. However, the collection process of radionuclides for medical isotopes encounters multiple difficulties. This thesis aims to address some of these challenges to optimize the collection efficiency, with a particular focus on terbium as a case study.

The production process starts with the irradiation of a target in order to produce the nuclide of interest; in this process, other radionuclides are also produced. Therefore, mass separation is used to separate the isotopes of interest, upon which they are implanted in the collection foil. An interesting element is terbium, as there exists a quadruplet of 4 isotopes with complementary decay characteristics. Consequently, various imaging and treatment techniques can be employed while their chemistry, and thus their reaction inside the body, is the same. Nevertheless, the optimization of this process remains a challenge.

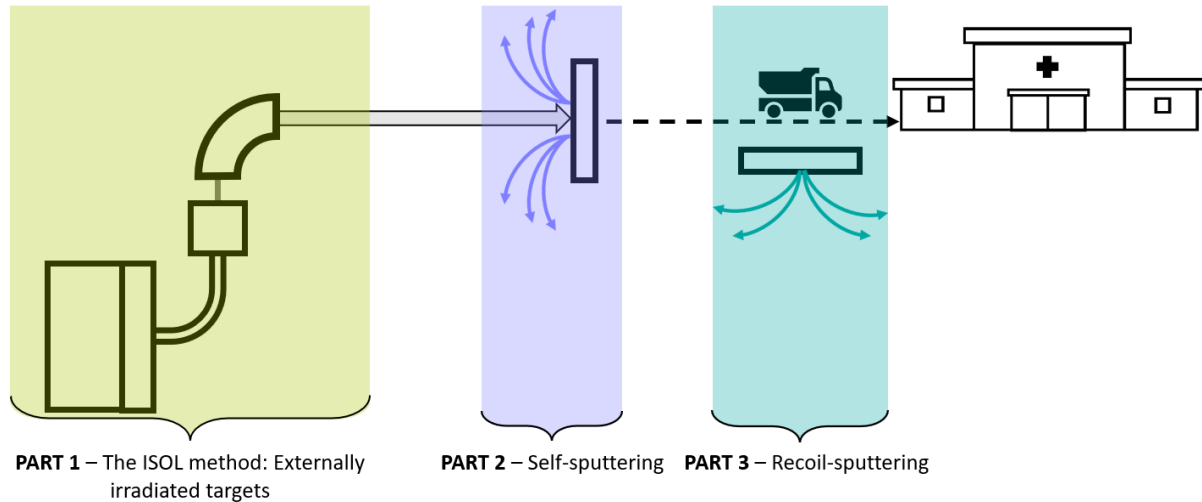


Figure 1: Overview of this thesis with the three main parts.

The main aim of this thesis is to investigate the efficiency of some of the steps of the collection process with terbium as a case-study. To accomplish this goal, this thesis is divided in three main parts (see Figure 1). As a broader introduction, Chapter 1 provides an overview of the state-of-the-art radionuclide production method, highlighting

the importance of terbium in nuclear medicine. In Part I, the three ^{155}Tb collections performed in 2022 with targets irradiated at PSI (Paul Scherrer Institute, Switzerland) are discussed. Moreover, the importance of the target preparation is investigated. Part II explores the problem of self-sputtering, i.e., if the fluence of particles impinging on the foil increases too much, earlier implanted particles can be removed from the foil. The self-sputtering behavior will be analyzed by the use of TRIDYN simulations. Moreover, a systematic study on the dependence on the foil and beam is conducted (with terbium as an example). Additionally, a protocol for experimental verification of the TRIDYN simulations was put forward. Recoil-sputtering in the context of α -decaying radionuclides is shortly introduced in Part III. Here, simulations with TRIDYN and SRIM are combined in order to get a first estimate for the importance of recoil-sputtering in the context of medical isotope collections.

Outline

Chapter 1 serves as a broader introduction by discussing the current state-of-the-art of terbium collections. Chapter 2 focuses on the collections conducted during the summer of 2022, followed by an in-depth discussion on the significance of target preparation in Chapter 3. The key findings from these two chapters are summarized in Chapter 4. Chapter 5 provides a general introduction to self-sputtering, while Chapter 6 presents a systematic study of self-sputtering. In Chapter 7, a protocol for an experimental verification of the used simulations is presented, followed by simulations for a collection of 2021 in Chapter 8. Again, a general summary on self-sputtering follows in Chapter 9. Lastly, recoil-sputtering is discussed in Chapter 10. The code utilized to obtain the results presented in those chapters can be found on GitHub [1]. Moreover, a poster for the LISA conference was presented on the importance of self-sputtering for medical isotope collections and is attached in Appendix G.

1

State-of-the-art of terbium collections

In this chapter, as a broader introduction, the state-of-the art of terbium collections is reviewed. In Section 1.1, an introduction to nuclear medicine is provided. Next, the production of terbium, by irradiating targets with protons (with energies of tens of MeV-GeV) is discussed in Section 1.2. During this production process, other radionuclides are also produced. To isolate the terbium isotope of interest from these other radionuclides, the ISOL (Isotope Separation On-Line) method is often employed, which is discussed in Section 1.3. The main aim of this thesis is to optimize the collection efficiency of medical isotope collections at CERN-MEDICIS, which is introduced in Section 1.4. Lastly, some of the existing and future facilities will be briefly introduced in Section 1.5.

1.1 Introduction to nuclear medicine

In nuclear medicine, radioactive nuclides are brought into the body for diagnostic or treatment purposes, i.e., to detect or destroy cancer tissue. First, a brief introduction to radioactive nuclides is provided, followed by an exploration of their role in diagnostics and therapy. Additionally, the importance of terbium in this context will be addressed.

1.1.1 Radioactivity

A nucleus consists of a number of protons (Z) and a number of neutrons (N). However, not all combinations of N and Z lead to stable nuclei, in fact, the vast majority of nuclei are unstable. This is seen on the nuclear chart (see Figure 1.1), which represents all known nuclei. The stable nuclei are indicated in black and are organized along the line of stability, which deviates from the $N=Z$ line. The main reason for this deviation is that for increasing Z , the repelling Coulomb force increases and thus more neutrons (increase in the strong force) are needed to mitigate for this repulsion in order to form a stable nucleus.

Unstable nuclei decay towards a lower energy configuration: the daughter nucleus. This decay process has a stochastic nature and depends on the half-life of the nuclide ($t_{1/2}$), i.e., the time after which half of the initial nuclei have decayed:

$$\frac{dN}{dt}(t) = -\frac{\ln(2)}{t_{1/2}}N(t) = -\lambda N(t). \quad (1.1)$$

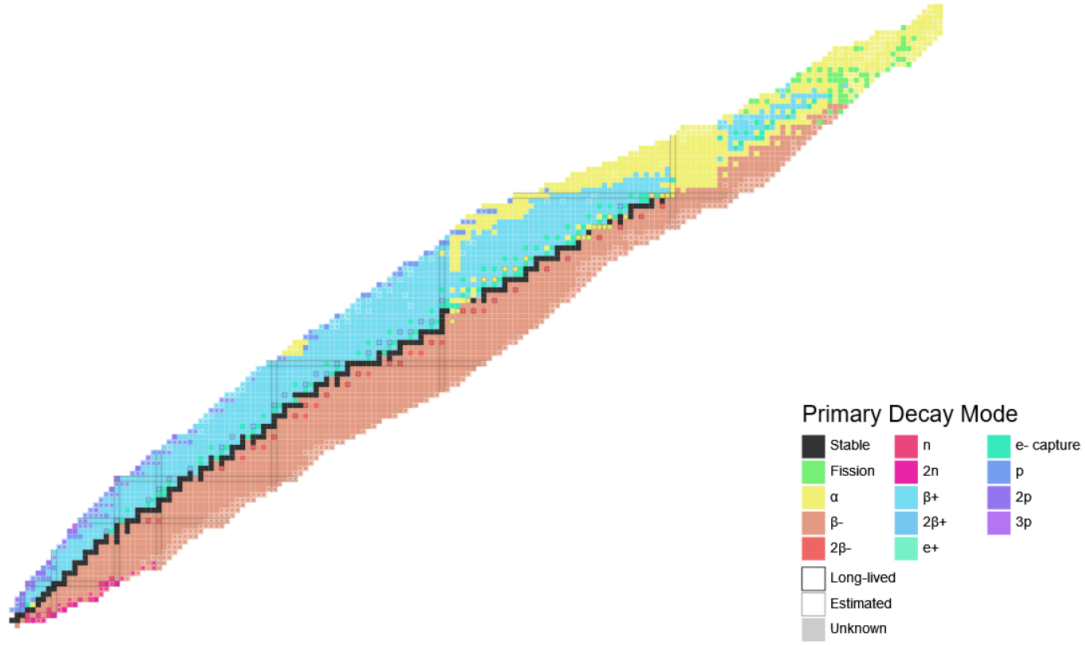


Figure 1.1: Nuclear chart, the number of neutrons is represented on the horizontal axis and the number of protons corresponds to the vertical axis. The black squares indicate the stable nuclei, whereas the colored squares indicate the unstable nuclei (orange = β^- , blue = β^+ , green = electron capture and yellow = α). (figure retrieved from [2])

Here, $N(t)$ is the number of mother nuclei present at time t and $\lambda = \frac{\ln(2)}{t_{1/2}}$ the decay constant. Solving this differential equations yields:

$$N(t) = N_0 e^{-\lambda t}. \quad (1.2)$$

The activity is now defined as the number of decays per second:

$$A = -\frac{dN}{dt}(t) = \lambda N(t), \quad (1.3)$$

which depends on the decay constant λ . This decay constant is important to characterize the decay process and to choose appropriate radionuclides for medical use as it depends on the half-life. In particular, the half-life cannot be too long, since this increases both the dose for the patient after treatment and the amount of long-lived nuclear waste present at the hospital. Therefore, in hospitals, isotopes with a shorter half-life (<1 day) are used [3]. On the other hand, if the half-life is too short, the majority of mother nuclei has decayed before reaching their intended location in the body, leading to an unwanted dose in the rest of the body and a lower dose at their intended location [3].

1.1.2 Types of radioactive decay and their use in nuclear medicine

In nuclear medicine, radioactivity is used for the diagnosis and treatment of various pathologies, in particular cancers. Here, a tracer, for example a sugar-analog, is attached to a radionuclide via a linker (see Figure 1.2). This tracer is then targeted towards processes or organic molecules that are more prominent in cancer cells than in healthy

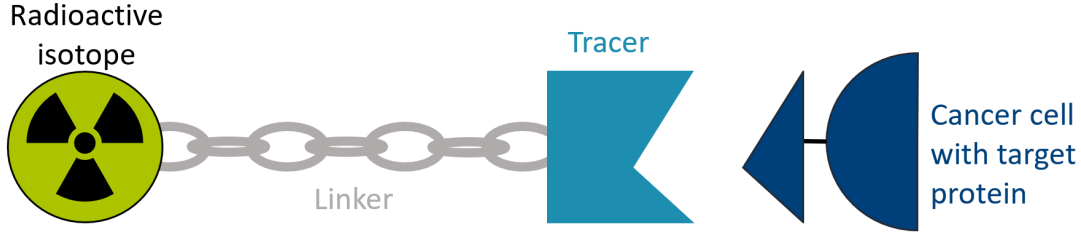


Figure 1.2: Schematic illustration of a radiopharmaceutical: the radioactive nuclide is attached to the tracer using a linker. The tracer ensures that the radiopharmaceutical accumulates at the cancer cells.

cells, e.g., they are targeted towards high metabolic activities. After arrival at the tumor site, the objective is that the radionuclide decays and emits radiation either for imaging or destruction of the cancer cells [4]. By using specific vector molecules, cells in specific parts of the human anatomy can be imaged or killed [4]. Each type of decay (α -, β - and γ -decay) has different properties and can thus be used for different medical modalities.

α -decay During α -decay, a ${}^4_2\text{He}$ nucleus is emitted from the mother nucleus. This process occurs mainly at high masses ($>150\text{u}$), as the Coulomb repulsion increases faster ($\sim Z^2$) than the attractive strong nuclear force ($\sim A$) [5]. Therefore, the nucleus is no longer stable and an α -particle is emitted:

$${}^A_Z\text{X} \rightarrow {}^{A-4}_{Z-2}\text{Y} + {}^4_2\text{He} + Q, \quad (1.4)$$

here Q corresponds to the energy that is released during this process which is equal to the difference in binding energy of the mother and the daughter nucleus. This energy is divided between the daughter nucleus (E_Y) and the α -particle (E_α) according to conservation of energy and momentum [5]:

$$E_\alpha = \frac{Q}{1 + \frac{m_\alpha}{m_Y}} \quad E_Y = \frac{Q}{1 + \frac{m_Y}{m_\alpha}}. \quad (1.5)$$

Here, m_α and m_Y are the masses of the α -particle and the daughter nucleus, respectively. The recoil-energy of the daughter nucleus in α -decay is typically of the order of 0.1MeV . In therapy for cancer, α -decaying particles are employed by using the energy deposited by the α -particle to destroy the cell DNA. The α -particles that originate from α -decays typically have high energies ($4\text{--}9\text{MeV}$) and a short path length ($40\text{--}100\mu\text{m}$). Therefore, a few α -particles are sufficient to cause cell-death in the close vicinity of the decay [6].

β^+ , β^- and electron capture (ϵ) The next type of decay processes is β -decay, which is the main decay path for light, proton-rich (β^+) or neutron-rich (β^-) nuclei [5]. In β^+ -decay, a proton is converted into a neutron, a positron (e^+) and a neutrino (ν_e) ($p \rightarrow n + e^+ + \nu_e$). For β^- -decay, a neutron is converted into a proton, an electron (e^-) and an antineutrino ($\bar{\nu}_e$) ($n \rightarrow p + e^- + \bar{\nu}_e$). Due to the presence of this (anti)neutrino, the β particle has no well-defined energy, but rather a distribution of energies [5].

For proton-rich nuclei, electron capture is also possible (ϵ -decay), in which an atomic electron is captured by the nucleus and forms, together with a proton, a neutron and a neutrino.

Nuclei that decay via β^+ -decay are used for PET imaging (Positron Emission Tomography). In β^+ -decay, a positron is emitted, which quickly annihilates with a nearby electron and consequently two 511keV γ -rays are emitted, with a relative angle of 180° . In PET imaging, these two coincident γ -rays are detected in order to retrieve back the position of annihilation. On the other hand, β^- -decay has therapeutic applications (targeted electron therapy). The emitted β -particles have medium energies (0.5-2.3MeV) and a longer path length than α -particles and are therefore more suited for larger diffuse tumors or for residual diseases [6].

γ -decay Following one of the above-mentioned decays or a nuclear reaction, the nucleus often remains in an excited state. The excess energy associated with this excited state is rapidly released by means of γ -decay. Here, the excited nucleus decays to a lower energy state by emission of one or more γ -rays. These γ -rays are photons with a specific energy, typically between 0.1MeV and 10MeV [5].

γ -decaying isotopes are used in Single-Photon Emission Computerized Tomography (SPECT). Here, the decay of the radionuclides is imaged by measuring the specific γ -rays. In that way, a visualization of the distribution of the isotopes is obtained.

1.1.3 Importance of terbium in the medical context

The terbium-quadruplet consists of ^{149}Tb , ^{152}Tb , ^{155}Tb and ^{161}Tb and has been researched extensively. Each of these isotopes has distinct decay modes (see Figure 1.3), and thus, those different isotopes can be used for different therapy and diagnostics modalities.

Tb 149	Tb 152	Tb 155	Tb 161
4.2 m	4.2 m	5.32 d	6.90 d
ϵ	ϵ	ϵ	β^- 0.5; 0.6...
β^+	β^+ 2.8...	γ 87; 105,	γ 26; 49; 75...
α 3.99	α 3.97...	γ 344;	e^-
γ 796;	γ 344;	586;	
165...	411...	180, 262...	

Figure 1.3: The terbium-quadruplet, together with the different decay modes for each isotope. (figure retrieved from [7])

The fact that different isotopes of terbium decay through different modes, is useful in the context of *theranostics*, in which diagnostics and therapy are combined to optimize the treatment [7]. The main advantage of having four medically interesting isotopes of terbium is that they share identical chemical properties, which ensures similar reactions in the body while their different decay characteristics provide the possibility of different medical modalities. This chemical similarity also enables the use of identical linker molecules, allowing for the extrapolation of research on linker molecules from one isotope to the others. However, to account for differences in half-lives between isotopes, distinct linker molecules must be utilized. For instance, shorter-lived isotopes require transfer molecules that rapidly transfer to the tumor site to reduce the radiation exposure in healthy tissue with respect to the dose given to the target. Conversely, longer-lived isotopes are better combined with transfer molecules with a slower transmission through the body to prevent excretion before a sufficient dose was given to the tumor cells and to minimize radiation exposure to excretion organs such as the kidneys.

Especially ^{149}Tb is a desirable isotope in the context of cancer treatment, in that it is an alpha emitter that decays into a long-lived daughter. Moreover, the long-lived daughter does not decay via α -decay or β -decay and thus no further damage to the tissue is caused. As a result of these properties, ^{149}Tb is highly suitable to use in Targeted Alpha Therapy (TAT) as the additional dose that the patient receives, due to the decay of the daughter nucleus, is limited. Moreover, the half-life of 4.1 hours is long enough to effectively treat the patient, but short enough to limit the additional dose the patient is exposed to. Additionally, ^{149}Tb has a β^+ decay branch, which allows for PET imaging [8]. Hence, the tumor cells can be imaged, while the α -decays destroy the double-stranded DNA of the cancer cells. Also, γ -decay is present and therefore SPECT imaging is also possible.

Another isotope that is suitable for PET imaging is ^{152}Tb , which decays via β^+ . Compared to ^{149}Tb , PET scans are significantly better for ^{152}Tb [6] due to the higher positron emission branching ratio. Nevertheless, when using ^{149}Tb for therapy, no additional dose is needed for the imaging as both PET imaging and alpha therapy are possible for ^{149}Tb via its β^+ and α -decay branch.

^{161}Tb decays β^- , but also electron conversion electrons and Auger electrons are present, which are all used for targeted electron therapy. Moreover, ^{161}Tb is also appropriate for SPECT (imaging). The BR2 nuclear reactor at SCK CEN (Mol, Belgium) is already capable of producing ^{161}Tb [9].

^{155}Tb decays by electron capture, making it suitable for therapeutic applications. Moreover, γ -rays are emitted that can be used for SPECT (imaging) [7]. ^{155}Tb can be paired with ^{161}Tb , forming a theranostic pairing: ^{155}Tb can provide pretherapeutic imaging prior to the therapy with ^{161}Tb [6].

1.2 Proton-induced nuclear reactions

Terbium can be produced by irradiation of a target with protons. The main advantage of irradiation with protons is that protons have a small mass and thus a small energy loss when penetrating through the target, leading to larger ranges of distance travelled through the material compared to heavier particles. Therefore, for a specific energy, using protons often gives the highest production yields per beam power unit compared to heavier projectiles [3].

Two different production modes are distinguished based on the energy of the protons that is used: low to medium proton beam energies (5-70MeV) and high proton beam energies ($> 100\text{sMeV}$). For the low to medium energy proton irradiation, three categories are distinguished based on the proton energy [3, 10]. For low energy protons (5-15MeV), mainly (p,n) and (p, α) reactions dominate, yielding radionuclides that predominantly decay β^+ . These radioisotopes have a short half-life and are thus produced close to or in the hospital. For proton energies in between 20MeV and 35MeV: mainly the (p,2n) and (p,3n) channels dominate. Lastly, for proton energies around 70MeV: (p,4n) and (p,2p) are possible.

For proton beam energies above several hundreds MeV spallation, fission and fragmentation are the dominant processes. The terbium production cross section for these reactions depends strongly on the energy [11]. These are however very dirty reactions in which a lot of impurities are produced.

During any irradiation with protons, contaminants are produced and therefore, separation of the isotope of interest is needed. In the case of offline separation, often, a chemical separation step such as elution (see Section 3.1) is performed after the irradiation. In this way, a large portion of the isobaric contaminants (contaminants with the same mass) can be removed. However, due to the fact that isotopic contaminants (contaminants of the same element) have the same chemistry as the isotope of interest, those cannot be separated using chemical techniques and therefore the ISOL mass separation method is employed. For example, for irradiation of ^{156}Gd with low energy protons, the cross sections for the production of different terbium isotopes are indicated in Figure 1.4. From which it is concluded that, apart from ^{155}Tb , other terbium isotopes also have non-negligible production cross sections [12].

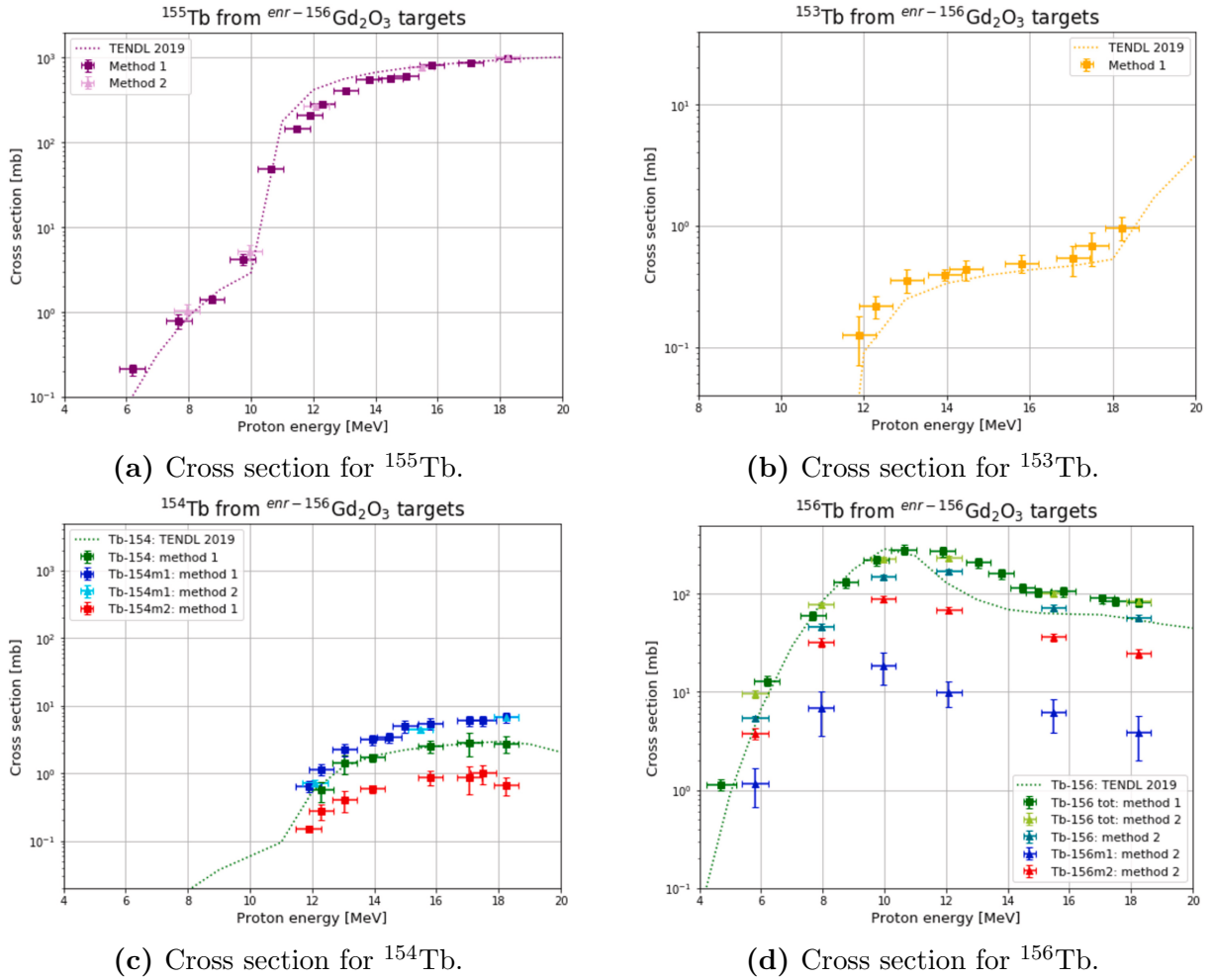


Figure 1.4: Cross section for different terbium isotopes for the irradiation of ^{156}Gd with low energy protons as a function of the energy of the protons. (figure retrieved from [12])

1.3 ISOL method

In this section, an overview of the ISOL method (see Figure 1.5) is provided, including the heating of the target and the subsequent diffusion of the isotopes towards the ion source, mass separator and collection box.

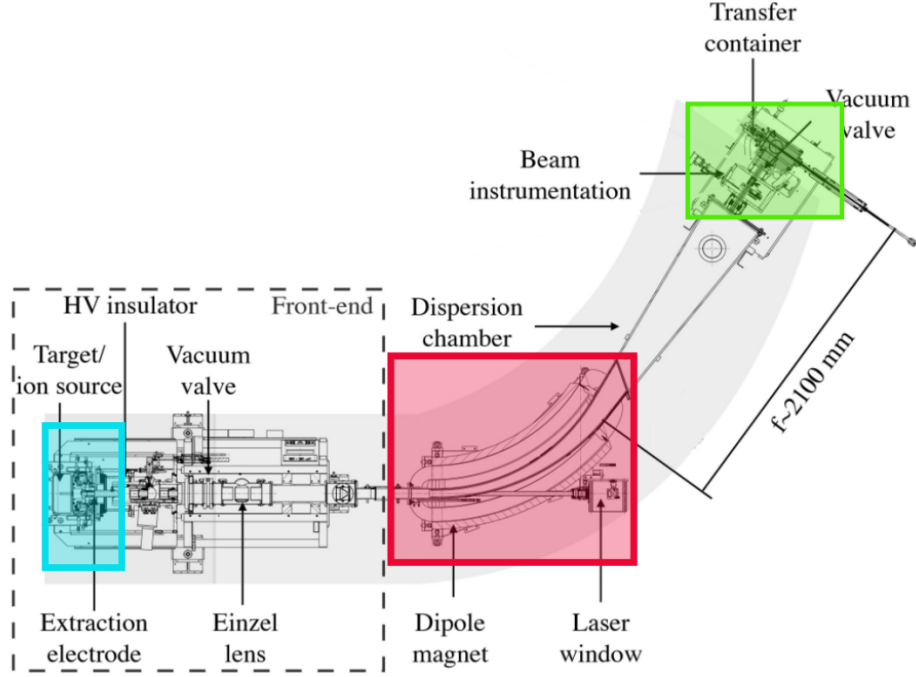


Figure 1.5: Overview of the ISOL method. The extraction and ionization are indicated in blue, the mass separation in red and the collection box in green. (figure retrieved and adapted from [13])

1.3.1 Extraction

The first step in the ISOL method is the extraction. Isotopes are extracted from the target by heating the target (typically $> 2000^\circ\text{C}$ for Tb collections) [14]. First, the isotopes diffuse towards the surface, the concentration of radioisotopes at the surface is determined by Fick's second law of diffusion:

$$\frac{\partial C(\vec{x}, t)}{\partial t} \sim \nabla \cdot \left(\exp\left(-\frac{\Delta H}{RT}\right) \nabla C(\vec{x}, t) \right), \quad (1.6)$$

with C the isotope concentration, R the universal gas constant, T the temperature and ΔH the activation enthalpy of diffusion [15]. Thus, as the temperature increases, the diffusion towards the surface is enhanced.

Next, the radioisotopes are released from the target at a desorption rate: $r_D \sim \exp(-\frac{E_{\text{act}}}{k_B T})$, where k_B is the Boltzmann constant and E_{act} is the activation energy for desorption, which is equal to the negative adsorption enthalpy [15]. Again, increasing the temperature enhances the desorption process [16]. After the diffusion towards the surface and the desorption from the surface, the isotopes effuse out of the target container through a transfer line towards the ion source. In conclusion, the defining factors for extraction are the activation enthalpy of diffusion and the activation energy for desorption. Both depend on the isotope, target material and container material. Hence, different isotopes are extracted at different temperatures and the sequence at which they come out can be approximated via the boiling temperatures, vapor pressures and the adsorption enthalpy when tantalum is used [17, 18].

1.3.2 Ionization

After extraction, the isotopes are ionized; most often surface ionization, resonance laser ionization or electron impact ionization are employed [19]. All three methods are visualized in Figure 1.6.

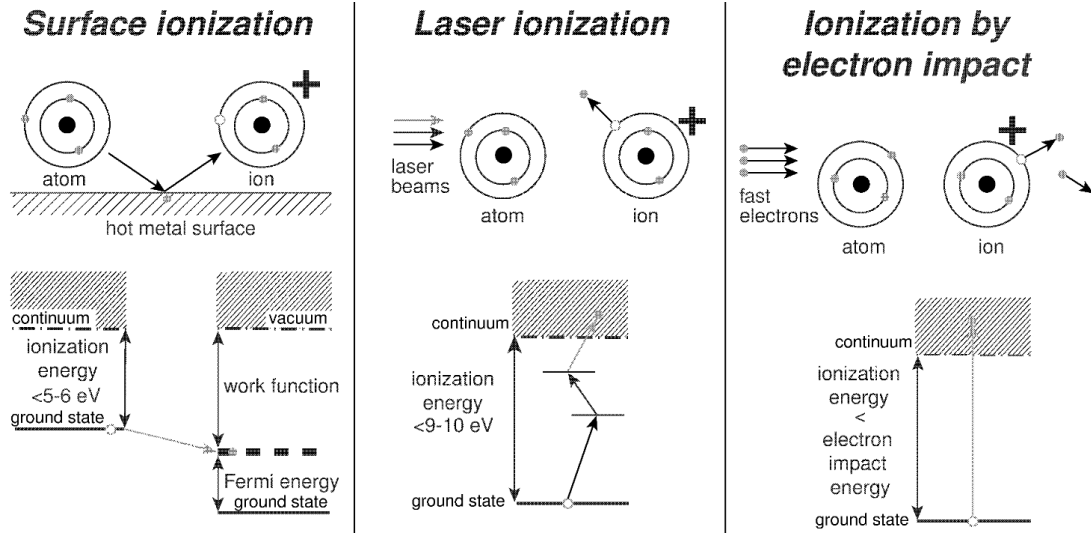


Figure 1.6: Schematic illustrations of surface ionization, laser ionization and electron impact ionization. (figure retrieved from [20])

Surface ionization relies on collisions with the surface, upon which the isotope loses or gains an electron, depending on the work function of the surface (φ) and the ionization potential of the isotope (W):

$$\alpha \sim \exp\left(\frac{\varphi - W}{k_B T}\right). \quad (1.7)$$

Here, α is the ionization degree, T the temperature and k_B the Boltzmann constant [21]. It is concluded that the largest ionization efficiency ($\varepsilon = \frac{\alpha}{1+\alpha}$) is achieved for materials with large work functions like tantalum ($\varphi = 4.19\text{eV}$), tungsten ($\varphi = 4.53\text{eV}$) and rhenium ($\varphi = 5.1\text{eV}$) [21]. Apart from the ionization potential of the isotope, no dependence on the isotope is present for surface ionization, therefore, surface ionization is not very selective, and many elements, other than the one of interest, are surface ionized [21].

The efficiency is usually increased due to the formation of a thermal plasma that creates a potential which confines ions and avoids de-ionization [22]. This effect is taken into account by an amplification factor $\mathcal{N}$: $\varepsilon = \mathcal{N} \frac{\alpha}{1+\alpha}$. If the number of incoming particles is too high (typically, for beam currents above 100nA), the potential is weakened by the many ions and is no longer able to confine new ions. This makes recombination at the surface possible, i.e., the ion source is saturated [23].

A schematic representation of a surface ion source is shown in Figure 1.7. A surface ion source consists of a line, which is a metal tube that has a higher work function than the atom that should be ionized. The temperature of this line can be regulated and is put on similar temperatures as the temperature of the target.

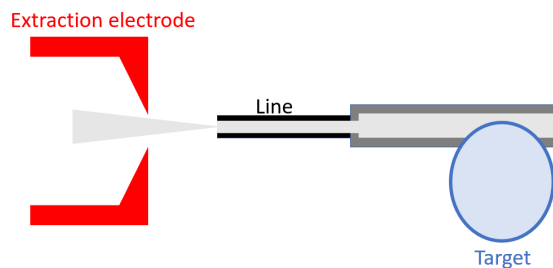


Figure 1.7: Surface ion source together with the extraction electrode and target.

Increased elemental selectivity can be obtained by using a Resonance Ionization Laser Ion Source (RILIS), which is schematically represented in Figure 1.8. Resonance laser ionization employs the electronic structure, which is element specific, to laser-ionize elements in two steps: the first set of lasers (usually 1 or 2 lasers) excites the elements of interest and a second laser is used for ionization of the excited atoms as is shown in Figure 1.9.

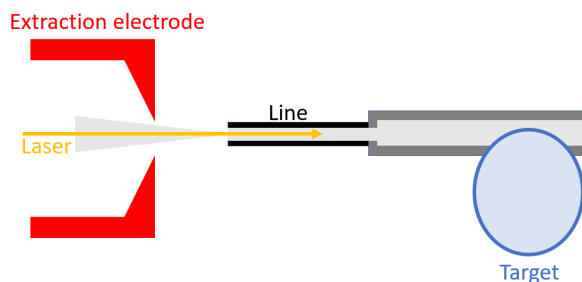


Figure 1.8: RILIS ion source. The laser is shown with respect to a surface ion source.

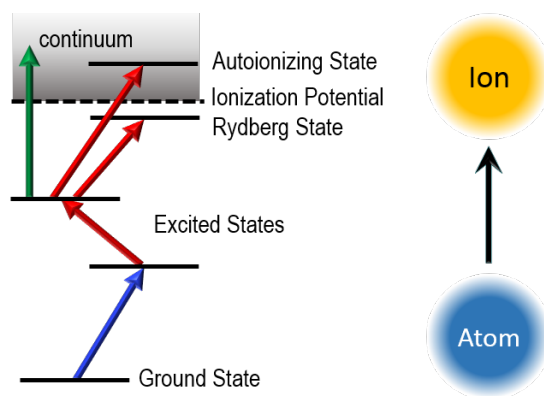


Figure 1.9: Resonance laser ionization principle: the first set of lasers excites the elements of interest and the second laser is used for the ionization of these excited atoms. (figure retrieved from [24])

Next to the increased elemental selectivity, another advantage of using RILIS compared to surface ionization is that the collection time, and thus the decay-induced loss, can be decreased due to the enhanced ionization efficiency.

Electron impact ionization is implemented in a FEBIAD (Forced Electron Beam Induced Arc Discharge) which is visualized in Figure 1.10 [25]. Here, the isotopes effuse out of the target towards the anode volume of the target, where the isotopes are ionized via bombardment with electrons that are extracted from the heated cathode and are accelerated ($E \approx 100 - 200\text{eV}$) towards the anode. The ionization efficiency depends on a number of parameters in the ion source, such as the time spent in the anode volume, the cathode temperature, the anode temperature and the anode voltage. The dependencies are not straightforward and are described in [25].

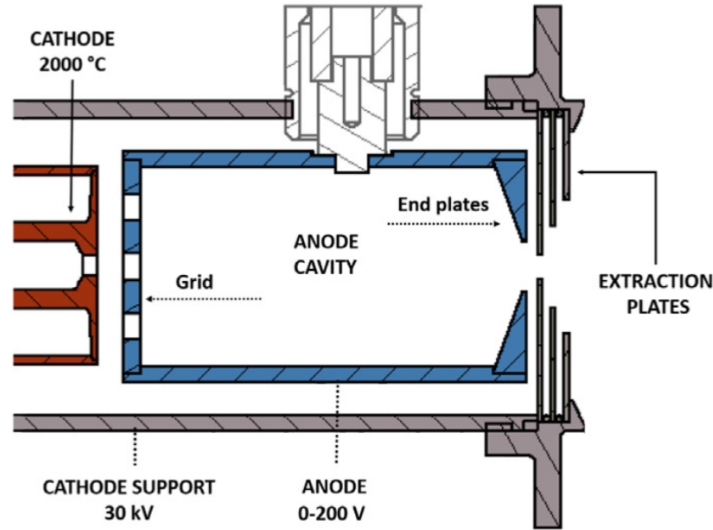


Figure 1.10: Illustration of a FEBIAD ion source. (figure retrieved from [26])

1.3.3 Mass separation

The target and ion source are put on high voltage, while the mass separator is maintained at ground potential. Hence, the ionized isotopes are accelerated towards the mass separator, where isobaric selectivity is obtained by using an analyzing magnet. This analyzing magnet separates the isotopes based on their mass to charge ratio. As the isotopes fly through a homogeneous magnetic field, their trajectory bends with a radius (r) that depends on the mass of the isotope:

$$r = \frac{mv}{qB} = \sqrt{m} \frac{\sqrt{2E}}{qB}, \quad (1.8)$$

where B denotes the applied magnetic field and m , E , v and q correspond to the mass, energy, velocity and charge of the particle [5]. As different masses give rise to different bending radii, the magnetic field can be changed in order to select the desired mass.

Another important factor to take into account is the mass resolving power: $MRP = d \frac{M}{FWHM}$, where M is the mass of the isotope of interest, $FWHM$ is the full width at half maximum of a beam of ions with mass M in the focal plane of the separator and d is the distance between two peaks with mass M and $M + 1$ [25]. Collection of contaminants needs to be avoided, therefore the mass resolving power should be maximal. The mass resolving power is enhanced if the beam is mono-energetic and narrow, which is achieved by using different electrostatic elements such as deflectors and Einzel lenses [25].

1.3.4 Collection box

After the mass separation, the isotopes can be collected on a foil, which is located in the collection box. The collection box is a vacuum chamber in which collection foils can be mounted. The current implanted on these foils can be measured throughout the collection. Moreover, Faraday cups are used, which can be inserted in the beam line to measure the current of the beam [25]. The implantation rate can directly be monitored by measuring the current deposited on the foil, while the collected radioactivity can be monitored with radiation detectors, such as the Kromek GR1 CZT detector [25].

1.3.5 Chemical purification

For the use in medical applications, high specific molar activity is required, i.e., the amount of contaminants needs to be minimized. After the mass separation, isotopes of the same mass are collected, such as, in the case of a ^{155}Tb collection, ^{155}Gd or the isobaric molecule contaminant $^{139}\text{Ce}^{16}\text{O}$ [11]. These contaminants are surface ionized and thus collected on the collection foil. Hence, further chemical separation is still required as mass separation is unable to differentiate between isobaric contaminants [27].

1.4 Optimization of the collections

Each of the steps discussed in the previous sections contributes to the total efficiency of a collection:

$$\varepsilon_{\text{tot}} = \varepsilon_{\text{diff}} \cdot \varepsilon_{\text{eff}} \cdot \varepsilon_{\text{ion}} \cdot \varepsilon_{\text{sep}} \cdot \varepsilon_{\text{transport}} \cdot \varepsilon_{\text{implant}} \cdot c_{\text{decay}} \quad (1.9)$$

Here, $\varepsilon_{\text{diff}}$, ε_{eff} , ε_{ion} , ε_{sep} correspond to the different steps of the ISOL method: diffusion, effusion, ionization and mass separation. $\varepsilon_{\text{transport}}$ is related to the transport of the beam towards the implantation foils. $\varepsilon_{\text{implant}}$ is the implantation efficiency, in other words, the fraction of particles that are retained (= not self-sputtered¹) on the foil compared to the total number of impinging particles. Lastly, c_{decay} is a factor that takes into account the radioactive decay during the collection.

Two main parts ($\varepsilon_{\text{tot}} = \varepsilon_{\text{ISOL}} \cdot \varepsilon_{\text{implant}}$) will be distinguished during this thesis: the efficiency of the ISOL method itself (= target choice, extraction, ionization and mass separation) and the efficiency of the implantation ($\varepsilon_{\text{implant}}$). Both the optimization of the ISOL method and the minimization of self-sputtering are discussed in the next two parts (part I and part II respectively), and are dependent on the operating conditions of each facility, such as the extraction voltage and the material of the foils used for the collections. Since different facilities operate in different conditions, an overview of the currently existing and future facilities is given in the next section.

Additionally, during the collection, activity is lost due to the decay and thus the collection time needs to be minimized. However, also the amount of activity implanted in the foil needs to be maximized. A balance between both needs to be found and, additionally, the transport time needs to be minimized. This time-factor is even more important for the collection of α -decaying isotopes (e.g., ^{149}Tb) as the daughter nuclei

¹If ions are implanted in a foil, earlier implanted isotopes can be lost due to self-sputtering. An elaborate discussion on self-sputtering is provided in Part II.

obtain a significant recoil energy. These recoiling daughter nuclei can induce sputtering of other implanted mother isotopes. Therefore, the mother nuclei are not only lost due to the decay, but also a small part is lost due to the fact that they can be sputtered out due to recoiling daughter nuclei originating from other decaying mother nuclei. A discussion on the importance of this recoil-sputtering for medical isotope collections is provided in Part III of this thesis.

1.5 Different facilities for radionuclide production

In this section, the different medical isotope productions facilities that are currently existing and the future facilities are discussed. There are two main categories of facilities: the facilities with online extraction and the facilities with offline extraction. For online extraction facilities, the irradiation of the target and the extraction of the isotopes are performed simultaneously. For the offline facilities, the irradiation of the target is conducted before the collection of the radioisotopes with the ISOL technique. The latter technique offers two main advantages. Firstly, it allows for an intermediate radiochemistry step to be performed between the irradiation and the collection, in order to have a more efficient extraction of the isotopes of interest. Secondly, short-lived contaminants have decayed already when the extraction starts. However, to collect short-lived isotopes online extraction is favored due to the decay-induced loss of those short-lived isotopes while transporting the target to an offline extraction facility.

Several facilities such as MEDICIS at CERN (Geneva, Switzerland), ISOLpharm at SPES (Legnaro, Italy), ISOL@MYRRHA at SCK-CEN (Mol, Belgium), SMILES at ARRONAX (Nantes, France) and TATTOOS at PSI (Villigen, Switzerland) exist or are planned to be built. The case-study of this thesis is CERN-MEDICIS, therefore, a more elaborate discussion on the MEDICIS facility is provided. For the other facilities, a summarizing table is provided in Table 1.1 and Table 1.2.

The MEDICIS [19] facility is used for the production of different isotopes for research in medical imaging and therapy. MEDICIS is built as an extension of CERN-ISOLDE (Isotope Separation On-Line DEvice), which is a facility where the ISOL method is exploited mainly for fundamental nuclear physics research and for medical nuclear physics. An overview of the MEDICIS facility with respect to ISOLDE is shown in Figure 1.11.

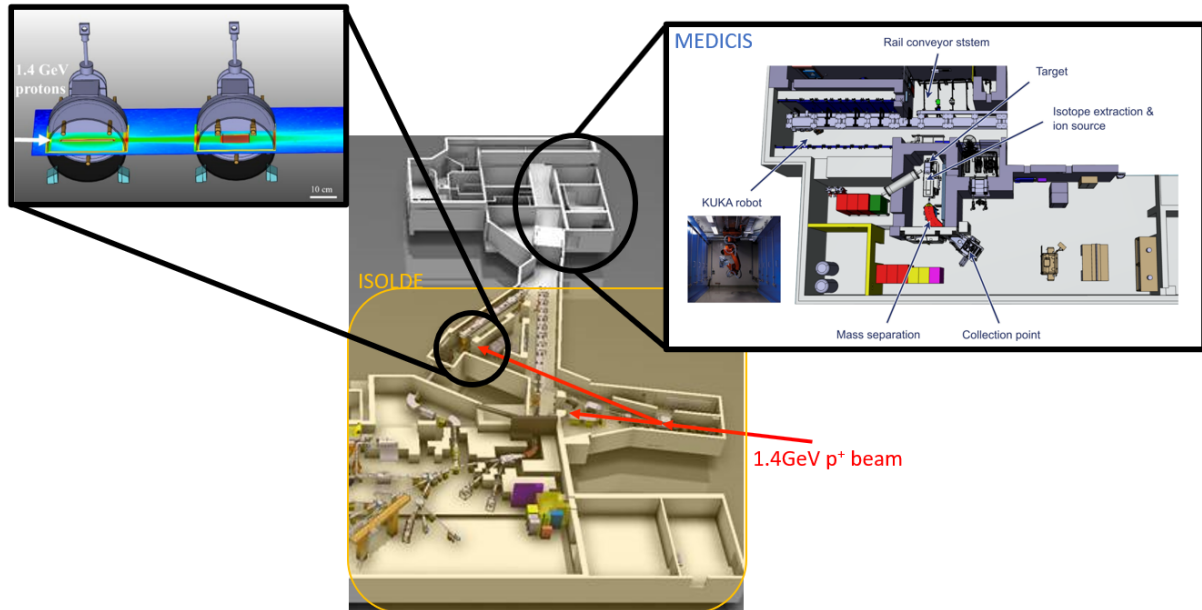


Figure 1.11: Integration of MEDICIS in ISOLDE, together with the top view of the MEDICIS setup (picture on the right). The picture on the left indicates how MEDICIS targets (rightmost target) are irradiated in combination with ISOLDE targets (leftmost target). (figures retrieved from [28, 29, 30])

MEDICIS is an offline collection facility in which two types of targets are used:

- Targets irradiated at ISOLDE: the MEDICIS target is placed in between the ISOLDE target and its beam dump and is irradiated by the proton beam of 1.4 GeV coming from the proton synchrotron booster (PSB) as is shown in Figure 1.11. These high energy protons induce spallation reactions in which many radioisotopes are created, amongst which terbium. Subsequently, the target is transported towards the MEDICIS front-end using a robot [19].
- Targets received from external institutes such as ARRONAX or PSI: these targets allow for collection during periods of long shutdowns at CERN. As a result, MEDICIS stands out as one of the few facilities at CERN able to run during periods in which no protons are available as the full accelerator chain is stopped [19]. The energies of the protons used for the target irradiation at PSI and ARRONAX are of the order of several 10s MeV. After irradiation of the target at the external facility, the target is dissolved and chemical preparation such as elution (see Section 3.1) can be performed. Thereafter, the radioisotopes are deposited on a foil, which is then shipped to MEDICIS and mounted inside a MEDICIS target.

Hereafter, the radionuclide collection can start at MEDICIS, using the ISOL method.

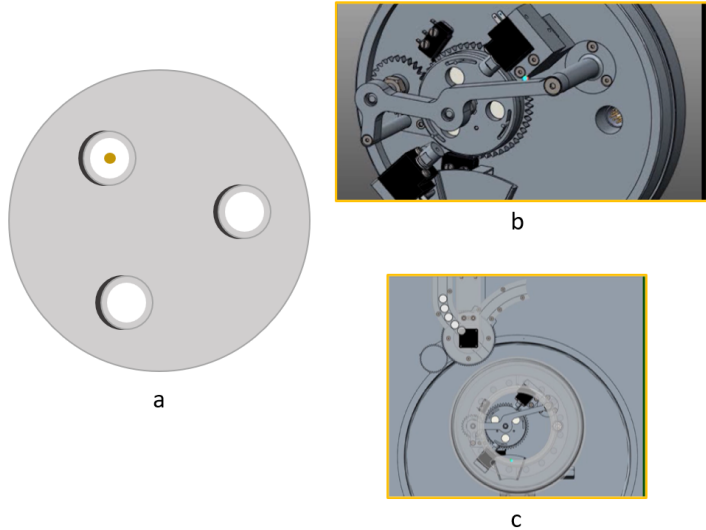
Facility	Type	Production	Extra information
ISOLpharm [31] (In commis- sioning)	Online	A Best Cyclotrons Systems Inc (BSCI) cyclotron is used to accelerate protons up to 70MeV [32] (variable from 35-70MeV [10]) that impinge on a carbide target. Such a carbide target consists for example of UC_2+2C , $TiC+2C$ or ThC_2+2C . The main advantages of these targets are the open porosity and nanostructure to obtain fast release of isotopes and the high thermal properties to efficiently dissipate heat [33].	A potential difference up to 40kV will be used to implant the isotopes in the collection foils [32]. The ISOLpharm radionuclide implantation station is shown in the figure below. This implantation station is equipped with robots that facilitate rotation and refilling of the target foils [33]. The different target foils that are currently researched at ISOLpharm are sodium chloride, sodium nitrate, dextrates and cellulose.  Radiopharmaceutical labelling and chemical separation of the produced nuclei can be performed on-site.
SMILES [34] (Not in com- missioning yet)	Offline	The ISOL-setup is disjunct from the proton accelerator present at AR-RONAX, this IBA accelerator accelerates protons up to 70MeV with a large intensity ($350\mu A$).	Radiochemical separation can be performed on-site.

Table 1.1: The most important characteristics for radionuclide collection at ISOLpharm and SMILES

Facility	Type	Production	Extra information
ISOL@MYRRHA [35] (Construction starting soon)	Online & offline	A 100MeV proton beam that is coming from the MYRRHA accelerator is split by the use of a magnetic kicker, upon which part of the beam is guided towards the ISOL setup. The other part is used for the MYRRHA reactor. Offline extraction will also be possible at ISOL@MYRRHA, with the aim to use external sources with a larger cross section for the production of specific radioisotopes [36].	The extraction will be performed between 30kV and 60kV. Additionally, radiochemistry and radiolabelling will be conducted on-site.
TATTOOS [37] (Not in commissioning yet)	Online	The high-intensity proton accelerator (HIPA) (injector 2) will provide the large intensity proton beam with energies up to 72MeV, also neutron irradiation will be possible at PSI [38].	Everything, starting from the proton irradiation reaction up to the chemical separation after the collection, will be conducted on-site. Moreover, at PSI, it will be possible to use the HIPA to produce a proton beam of 590MeV in order to induce spallation reactions by irradiation of a ^{181}Ta target. The prospects of this method are promising: in theory, 67.3GBq of ^{155}Tb can be produced after irradiation of 12h [38].

Table 1.2: The most important characteristics for radionuclide collection at ISOL@MYRRHA and TATTOOS.

PART I: The ISOL method - Externally irradiated targets

Before anything else,
preparation is the key to success

Alexander Graham Bell

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2

Collections of summer 2022

In this chapter, an analysis on the terbium collections conducted during the summer of 2022 is presented. Three collections were carried out at MEDICIS for ^{155}Tb using targets that were irradiated at PSI. The first section provides an overview of the ^{155}Tb collections performed in 2022. Thereafter, the collections of July 4, July 11 and July 15 are discussed. Lastly, a conclusion based on all three collections is provided. Overall, the analysis in this chapter aims to provide insights into the optimization of the ISOL method for future terbium collections.

2.1 General information about the collections

A flowchart of the collection procedure for externally irradiated targets is shown in Figure 2.1. First, the target is irradiated at PSI, which is described in Section 2.1.1. Thereafter, the target is dissolved and chemical separation is performed at PSI. Subsequently, the isotopes are deposited on a foil and the foil is shipped to MEDICIS, where it is mounted inside a MEDICIS target. Thereafter, the collection is started and performed as was described in Section 1.3. Several measurements are performed during the collections, which are elaborated on in Section 2.1.3.

A summary of all three collections is provided in Table 2.1.

2.1.1 Target production

The targets that were used during the collections in the summer of 2022 were produced at PSI. More specifically, an enriched ^{156}Gd target ($^{156}\text{Gd}_2\text{O}_3$) was irradiated with 22.8 MeV protons ($^{156}\text{Gd}(p,2n)^{155}\text{Tb}$). Following the irradiation, the target is dissolved and undergoes chemical separation (elution, see Section 3.1) in order to remove a portion of the gadolinium present in the target. The basics of elution and its importance will be discussed in Chapter 3. For the collection of July 4 and July 11, HCl was used as an eluent whereas for the collection of July 15, HNO_3 was used as eluent.

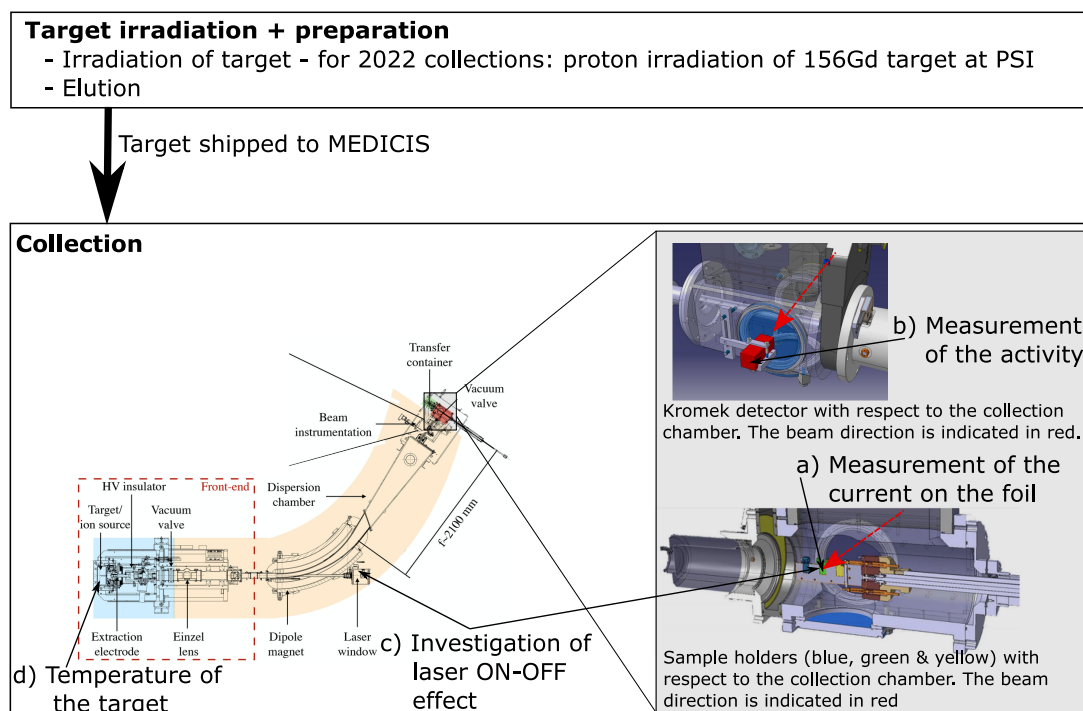


Figure 2.1: General flowchart for the collections that were performed in the summer of 2022. a, b, c and d correspond to the different measurements that were performed throughout the collections. More specifically, a corresponds to the measurement of the current on the foil (both ^{155}Tb and isobaric contaminants), b represents the activity measurement of ^{155}Tb . c corresponds to the laser ON-OFF effect, in other words, the difference in the current on the foil with the lasers on and off (for investigation into laser resonance ionization). Lastly, d corresponds to the temperature of the target container. (figures retrieved and adapted from [13] and [39])

	July 4	July 11	July 15
Target	# 725M	# 764M	# 726M
Facility	PSI		
Target material	$^{156}\text{Gd}_2\text{O}_3$		
Proton energy	22.8MeV		
Beam current	$25\mu\text{A/h}$		
Irradiation time	16h		
Eluent	0.05M HCl	0.05M HCl	0.05M HNO_3
Activity of ^{155}Tb in target at start collection	1018.01MBq	546.62MBq	950.24MBq
Tb to Gd ratio	$> 10^{12}/20$		
Ion source	Ta surface ion source	FEBIAD ion source	Re surface ion source
Efficiency	0.573(32)%	0.01515(22)%	0.3436(136)%

Table 2.1: Summary of the three collections that were performed in 2022 at MEDICIS (see Section 2.2, 2.3 and 2.4).

2.1.2 Laser resonance ionization

For the three collections performed in 2022, the three lasers in MELISSA were tuned for two schemes: 23107.3cm^{-1} & 24338.421cm^{-1} and 24613.87cm^{-1} , as is shown in Figure 2.2. Both schemes do not uniquely laser ionize terbium, but also parasitically ionize gadolinium. However, scheme A is the most efficient scheme for reducing the interference of gadolinium ionization [40]. From 2022 onwards, scheme A and B are subsequently turned off approximately every 5min, for investigation into laser ionization compared to surface ionization.

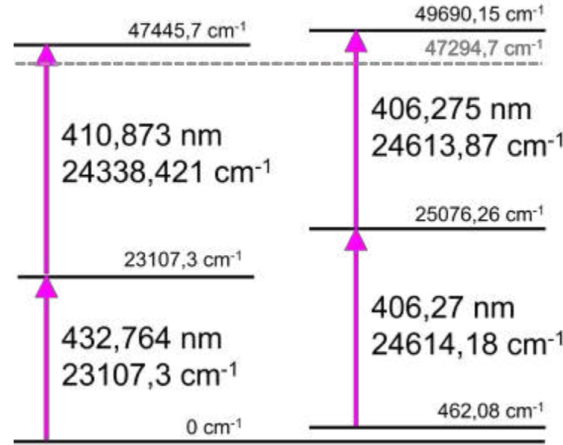


Figure 2.2: Laser scheme A (left) and scheme B (right) used at MEDICIS for the laser resonance ionization of Tb. (figure retrieved from [23])

2.1.3 Measurements performed during the collections

For each collection, several measurements are performed and the most important ones will be discussed in this section. The ion current on the foil is measured continuously throughout the collection (see a in Figure 2.1). This ion current provides a measure for the total number of particles that impinge on the collection foil. The total ion current on the foil includes isobaric contaminants as those are surface ionized, selected by the mass separator and subsequently implanted in the collection foil.

Moreover, an activity spectrum is acquired every 5 minutes by the use of a Kromek CZT detector [41] (see b in Figure 2.1), from which the activity corresponding to ^{155}Tb can be calculated, as is described in Appendix A. Nevertheless, the automation program for the Kromek CZT detector sometimes crashes and at those times, the spectra are not saved. For example, in Figure 2.4a, no activity information is shown between 100 minutes and 950 minutes after the start of the collection due to the fact that the automation program crashed. The activity corresponding to ^{155}Tb in the collection chamber provides a measure for the number of ^{155}Tb particles that are collected on the foil. In order to compare with the total current on the foil, the current that corresponds to the activity of ^{155}Tb is calculated, taking into account the decay process:

$$c_{\text{Tb}}(t + \Delta t) = \frac{e}{\Delta t} \cdot \frac{A_{\text{Kromek}}(t + \Delta t) - A_{\text{Kromek}}(t) \exp(-\lambda_{155\text{Tb}}\Delta t)}{\lambda_{155\text{Tb}}}. \quad (2.1)$$

Here, c_{Tb} represents the part of the current on the foil that corresponds to ^{155}Tb , t is the time, $e = 1.602 \cdot 10^{-19}\text{C}$ the electron charge, $\lambda_{155\text{Tb}}$ the decay constant and A_{Kromek} is the activity of ^{155}Tb measured by the Kromek detector. If the value of Δt is chosen to be too small, random fluctuations of the activity are taken into account and may lead to inconclusive results.

Additionally, the current corresponding to the laser ionization can be calculated (see **c** in Figure 2.1). The lasers are periodically turned off, which results in a decrease in the current on the collection foil as can be seen in Figure 2.3. The minimal value around each periodic position corresponds to the surface ionization as no lasers are present at that point. The total current (surface + laser ionization) is obtained from a polynomial fit (degree=20) around each ON-OFF effect based on the data without the ON-OFF dips. Upon subtraction of the surface ionization current from the total current, a measure for the laser resonance ionization current is obtained. If we assume that there are no other resonances for the used lasers, only terbium will be laser ionized. Under this assumption, ^{155}Tb will be the only contribution to the laser resonance ionization current, as the mass separator is configured on mass 155u. Consequently, the laser resonance ionization current is an estimation for the amount of ^{155}Tb that is laser ionized.

Lastly, the temperature of the target container (see **d** in Figure 2.1) is measured throughout the collections. The temperature on the target container is measured as a current [A]. In order to convert to temperature[°C], $\text{current}[A] = \sqrt{\frac{a \cdot \text{temperature}[^{\circ}\text{C}]}{c + \text{temperature}[^{\circ}\text{C}]} + \frac{b \cdot \text{temperature}[^{\circ}\text{C}]^4}{c + \text{temperature}[^{\circ}\text{C}]}}$ was used to fit the calibration measurements of the target¹.

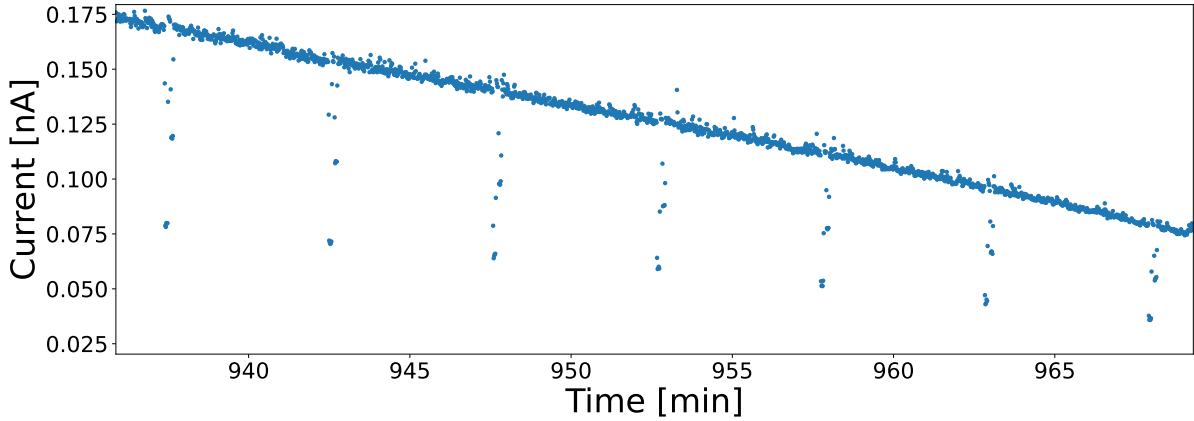


Figure 2.3: Current on the foil during a part where ^{155}Tb is collected as a function of the time after the start of the collection. In intervals of five minutes, the effect of switching the lasers off can be observed as a decrease in the current on the foil.

¹In order to perform this fit, a code was used that is written by Jake Johnson [42].

2.2 Collection of July 4

During the collection that started on July 4, a target from PSI was used. More specifically, a $^{156}\text{Gd}_2\text{O}_3$ (99mg) target was irradiated with 22.8MeV protons for 16h (beam current: $25\mu\text{A/h}$). Subsequently, elution was carried out at PSI using 0.05M HCl and the activity of ^{155}Tb in the target at the start of the collection was 1018.01MBq. In addition, an ICP-OES (Inductively Coupled Plasma Optical Emission Spectroscopy) analysis was conducted after the elution and gadolinium could not be detected, indicating that the terbium to gadolinium ratio is larger than $\frac{10^{12}}{20}$. Hence, the majority of ^{155}Gd present during the collection originates from the decay of ^{155}Tb into ^{155}Gd .

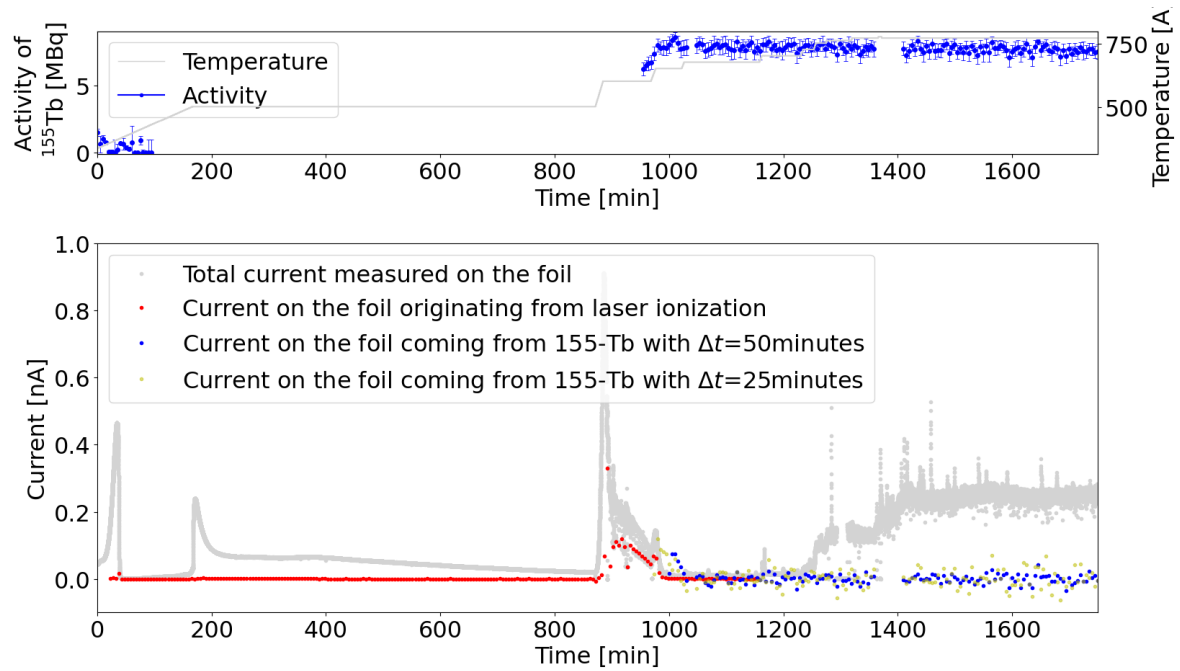
After elution, the isotopes were deposited on a Re foil and subsequently transported to MEDICIS where they were mounted in a MEDICIS target. During the collection, the target was put on high voltage (60kV) and a Ta ion source was used. Moreover, the collected radioisotopes were implanted in an aluminum-coated gold foil (M183), while the mass separator was set to mass 155u. The collection started on July 4 (17h30) and lasted for 41h. Thereafter, another foil was loaded with the objective of collecting more terbium and to understand why lower activities than expected were collected on the first foil. Nevertheless, no significant amount of terbium was collected on the new aluminum-coated gold foil (M184).

The results of the measurements during the collection on the first foil are shown in Figure 2.4a. Upon initiating the heating towards 500A, which corresponds to 1685.4°C, a strong increase in the current on the foil can be seen. However, no laser ON-OFF effect nor increase in the activity of ^{155}Tb is observed, indicating that no ^{155}Tb is collected, but rather a contaminant with mass 155u is collected. After this peak of about 38 minutes, the current on the foil stays rather constant (increase from 0.001nA towards 0.014nA in 100 minutes). Once the temperature reaches 500A (1685.4°C), another peak in the current on the foil is observed, but no laser ON-OFF effect is seen, suggesting that a contaminant at mass 155u is being collected.

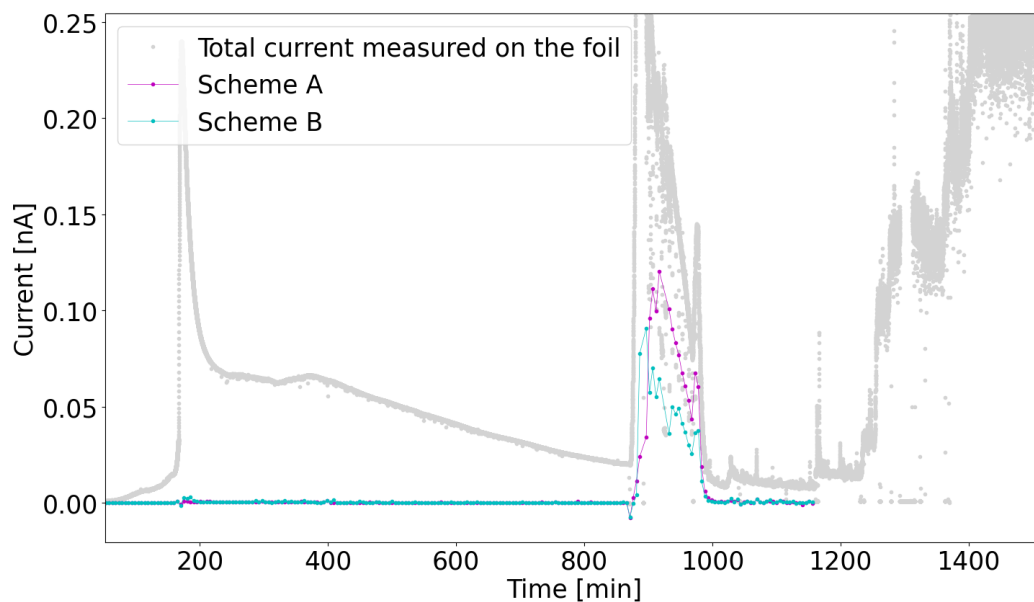
As the temperature rises towards 600A (1988.5°C), a peak in the current on the foil is observed. The first part shows a minor increase in the laser ON-OFF effect ($\frac{\text{ON}}{\text{OFF}} = \frac{0.89\text{nA}}{0.8\text{nA}}$), while the second part shows a strong increase in the laser ON-OFF effect ($\frac{\text{ON}}{\text{OFF}} = \frac{0.23\text{nA}}{0.12\text{nA}}$). The first Kromek measurement (after the crash of the software) yields an activity of ^{155}Tb of 6.19(49)MBq, confirming that ^{155}Tb was collected. From Figure 2.4b, it is observed that during the first part of this peak in the current on the foil, scheme B is predominantly enhanced. The amount of ^{155}Gd that is present in the target at 871 minutes after the start of the collection can be calculated based on the activity corresponding to ^{155}Tb that was present in the target after the chemical separation (1465.92MBq on 01/07/2022 at 14h50):

$$\begin{aligned} N(^{155}\text{Gd}) &= N_0(^{155}\text{Tb}) \left(1 - e^{-\frac{\ln(2)}{t_{1/2}} \cdot (01/07/2022 \ 14\text{h}50 - 05/07/2022 \ 9\text{h}01)} \right) \\ &= \frac{1465.92\text{MBq}}{1.5 \cdot 10^{-6}\text{sec}} \left(1 - e^{-\frac{\ln(2)}{5.32 \cdot 24 \cdot 60\text{min}} \cdot (5411\text{min})} \right) = 1.578 \cdot 10^{13} \text{particles.} \end{aligned}$$

Thus, at least, a current of 0.7nAh of ^{155}Gd is present in the target. Therefore, it is likely that the first part of the aforementioned peak in the current (where mainly scheme B is enhanced) corresponds to ^{155}Gd instead of ^{155}Tb as scheme B has a larger interference for parasitical laser resonance ionization of Gd than scheme A.



(a) Summarizing figure for the ^{155}Tb collection started on July 4. In the upper plot, both the temperature (gray) and the activity of ^{155}Tb measured with the Kromek (blue) are shown as a function of the time after the start of the collection. In the other plot, the total current measured on the foil, the current originating from laser resonance ionization and the current on the foil that is calculated from the activity of ^{155}Tb are shown as a function of the time after the start of the collection.



(b) Laser resonance ionization current corresponding to laser scheme A (magenta) and laser scheme B (cyan) of the laser resonance ionization as a function of the time after the start of the collection for the ^{155}Tb collection started on July 4 (zoomed in Figure of 2.4a with the currents for scheme A and B shown separately).

Figure 2.4: Summary on the collection of July 4

At 980 minutes after the start of the collection, a notable decrease in the current on the foil, the laser ON-OFF current and the current corresponding to the activity of ^{155}Tb is observed. This indicates that ^{155}Tb is no longer being collected. The total current measured at this point is 14nA, which is expected to be below the threshold for ion source saturation (typically, ion source saturation is only expected at currents above 100nA). One possible reason for the fact that Tb is no longer collected may be the formation of a temperature gradient inside the target. Consequently, ^{155}Tb can condense on these colder parts of the target, inhibiting its vaporization from the target. Another possibility is that the target is depleted.

Hereafter, no more ^{155}Tb was collected, the temperature of the target was increased again, but no laser ON-OFF effect nor increase in the activity of ^{155}Tb is observed. Moreover, from the mass scans (see Figure 2.5), it is concluded that no laser enhancement is present. Most likely, lanthanum oxide is collected at this point, which is a contaminant present in the target container.

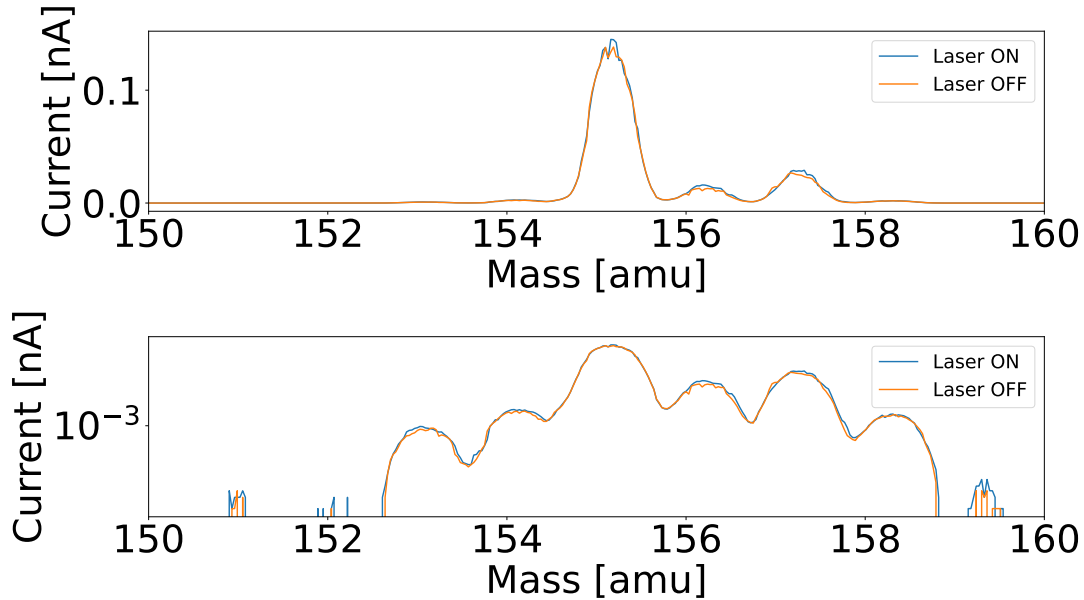


Figure 2.5: Mass scan (current as a function of atomic mass) performed at $\approx 1290\text{min}$ after the beginning of the collection. Two mass scans were performed, one for the lasers on (blue) and one for the lasers off (orange). Both figures show the same mass scans for laser ON and OFF, but the second figure is in logscale.

The activity of ^{155}Tb collected on the foil after removal was measured with a high purity germanium detector and yielded an activity of 5.83(32)MBq at the moment of foil removal. Hence, the efficiency of the collection can now be calculated as:

$$\varepsilon = \frac{\text{Activity of } ^{155}\text{Tb} \text{ measured after the collection corrected for the decay}}{\text{Activity of } ^{155}\text{Tb} \text{ present in the target at the start of the collection}}. \quad (2.2)$$

For this collection, $\varepsilon = \frac{5.83(32)\text{MBq}}{1018.01\text{MBq}} = 0.573(32)\%$, indicating that the collection was not successful.

A possible explanation for this low collection efficiency could be the reaction of terbium with chlorine atoms present in the eluent to form terbium chloride (TbCl_3). This hypothesis suggests that the majority of the ^{155}Tb that was present in the target was converted into TbCl_3 , but not collected, as it was most likely not ionized. More specifically, the ionization potential of TbCl_3 is larger than that of tantalum and the energy levels of TbCl_3 are different from those of Tb and therefore TbCl_3 will most likely not be ionized. However, mass scans were only performed up to the mass of TbCl during the collection and thus, no clear conclusions can be drawn. A more extensive elaboration on this hypothesis is presented in Chapter 3.

2.3 Collection of July 11

The next collection, which started on July 11 and lasted for 42h, aimed to explore the effect of the chlorine used during the elution on the formation of terbium chloride. To achieve this, the mass separator was also configured on the mass of terbium chloride. Similar to the previous collection, a $^{156}\text{Gd}_2\text{O}_3$ (99mg) target was irradiated with 22.8MeV protons for 16h (beam current: $25\mu\text{A/h}$) at PSI. Elution was performed with 0.05M HCl. The activity of ^{155}Tb that was present in the target at the beginning of the collection was 546.62MBq and the terbium to gadolinium ratio is larger than $\frac{10^{12}}{20}$. Instead of a surface ion source, a FEBIAD ion source was used. The main reason for using a FEBIAD ion source is its enhanced suitability for the ionization of molecules [43]. Furthermore, the lasers, for laser resonance ionization, were configured on both terbium schemes. During the collection, the mass separator was first set to the mass of $^{155}\text{TbCl}$ and later adjusted to the mass of ^{155}Tb . However, based on the activity spectra, the mass was suspected to be configured in between 155u and 156u.

The collection was not successful and almost no ^{155}Tb ($< 0.35\text{MBq}$) was collected.

During the collection, activity spectra were acquired every 5 minutes using the Kromek detector. These spectra were analyzed by the use of Interspec [44]. As is indicated in Figure 2.6, Interspec can be used to identify the peaks in the activity spectra. Multiple peaks can be identified, the peak corresponding to ^{139}Ce ($t_{1/2} = 137.6\text{d}$) originates from the ^{139}Ce that was released from the target during the terbium collections in the summer of 2021. ^{139}Ce is present both on the sample holder, due to implantation, and in the collection chamber, due to self-sputtering. For the same reason, ^{153}Sm ($t_{1/2} = 46.27\text{h}$) and ^{131}Ba ($t_{1/2} = 11.5\text{d}$) are present. ^{156}Tb ($t_{1/2} = 5.4\text{d}$) is initially present and corresponds to ^{156}Tb that was collected during the collection of July 4, as indicated by the γ -spectra.

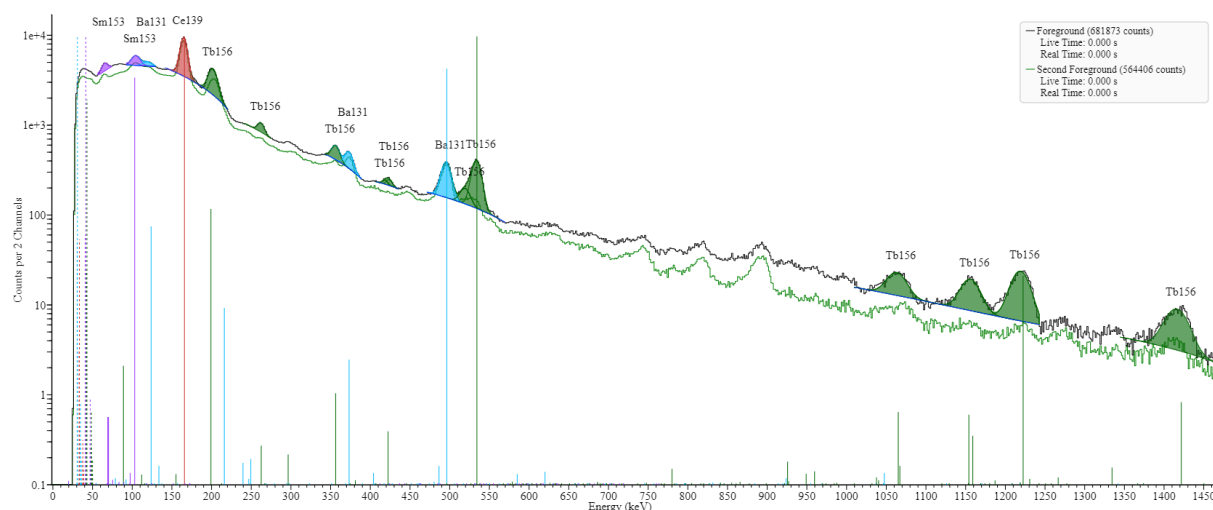


Figure 2.6: Mean spectra (counts as a function of energy) for the collection of July 11 together with the assigned peaks (purple: ^{153}Sm , blue: ^{131}Ba , red: ^{139}Ce and green: ^{156}Tb). The **Foreground** corresponds to the mean value during the period where the mass was configured on $m(\text{Tb})$ and the **Second Foreground** represents the mean value during the first part of the collection where the mass was set on $m(\text{TbCl})$. Note that the decay was not taken into account when calculating the mean spectra.

In Figure 2.6, the mean spectra during the part where the mass was configured on $m(\text{TbCl})$ and on $m(\text{Tb})$ are indicated in green and black, respectively. In the first part, where the mass separator was configured on the mass of TbCl , the temperature of the target container is equal to 150.5A (= 752.3°C). Such a small temperature is chosen because of the low boiling temperature of TbCl_3 (180-200°C [45]). At this point, no radioisotopes are collected or at a lower rate than the decay. Moreover, no peaks in the spectra can be assigned to ^{155}Tb , implying that no ^{155}Tb is collected. However, ^{156}Tb is present, nevertheless the corresponding activity decreases during this part of the collection, indicating that it corresponds to the remainder of the July 4 collection. In conclusion, if terbium chloride was vaporized from the target, it was before the Kromek measurements were started or terbium chloride was not ionized. During the part where the mass separator is configured on $m(^{155}\text{Tb})$, ^{156}Tb is being collected. As can be seen in Figure 2.7 and 2.8, no increase in the activity corresponding to ^{156}Tb is observed when the mass on the mass separator is changed. However, ^{156}Tb is collected when the temperature is increased towards 550A-610A which corresponds to 1803°C-1929°C.

The measurement with the high-purity germanium detector of the collection foil after removal from the collection chamber yielded an activity of 82.8(161)kBq corresponding to ^{155}Tb and 50.2(35)kBq corresponding to ^{156}Tb . Indicating that the efficiency of this collection equals 0.01515(22)% for ^{155}Tb , while the ‘collection efficiency’ for ^{156}Tb would be 0.0826(58)%.

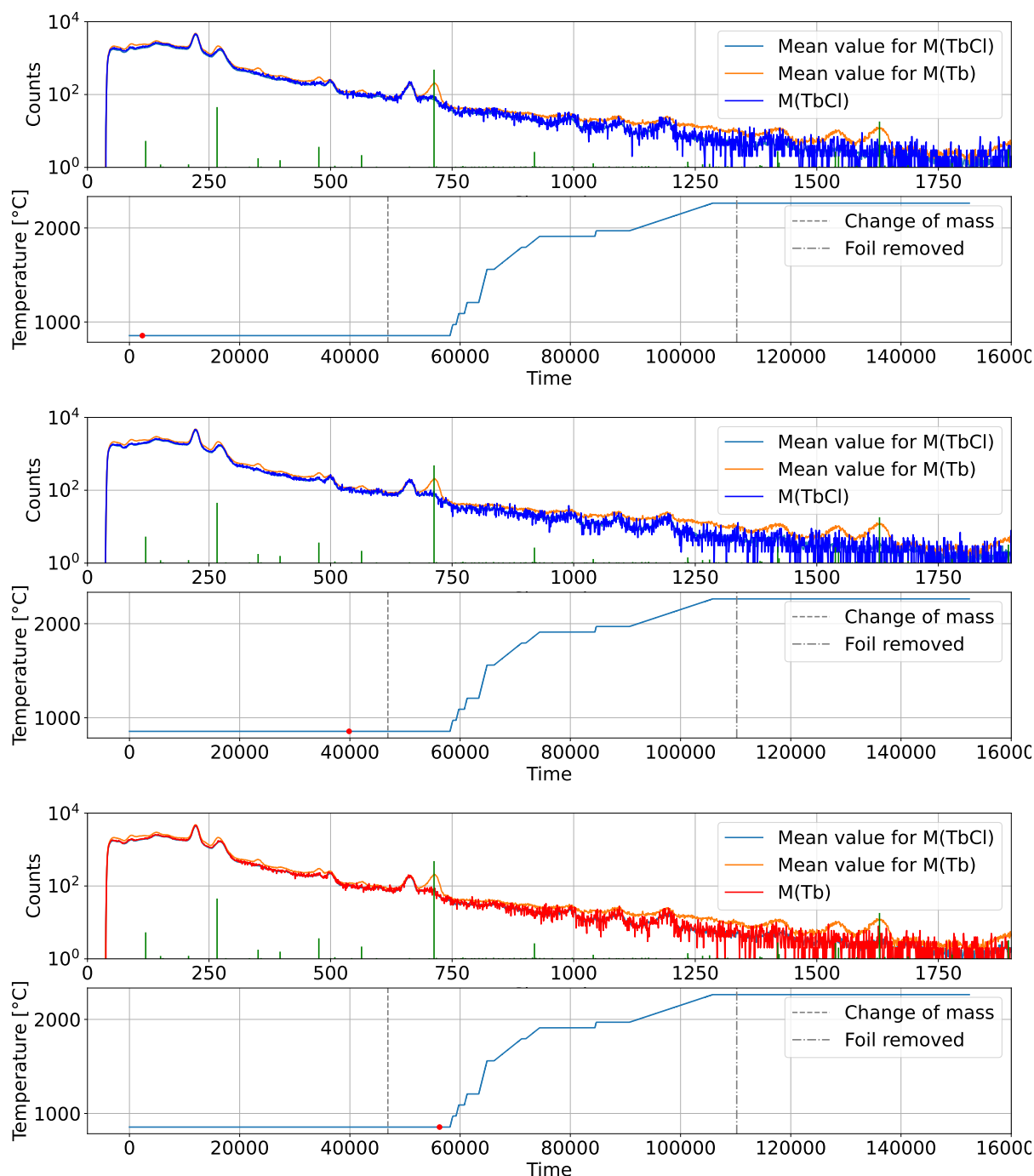


Figure 2.7: Summarizing graphs on the collection of July 11. Both the location in the temperature profile (red dot) and the corresponding spectrum (counts as a function of channel number) is given, together with the mean value when the mass separator was on the mass of Tb or TbCl. The dashed line indicates where the configuration of the mass separator was changed from TbCl to Tb. The green vertical lines indicate the energy lines corresponding to ^{156}Tb , the height of these lines corresponds to their intensity.

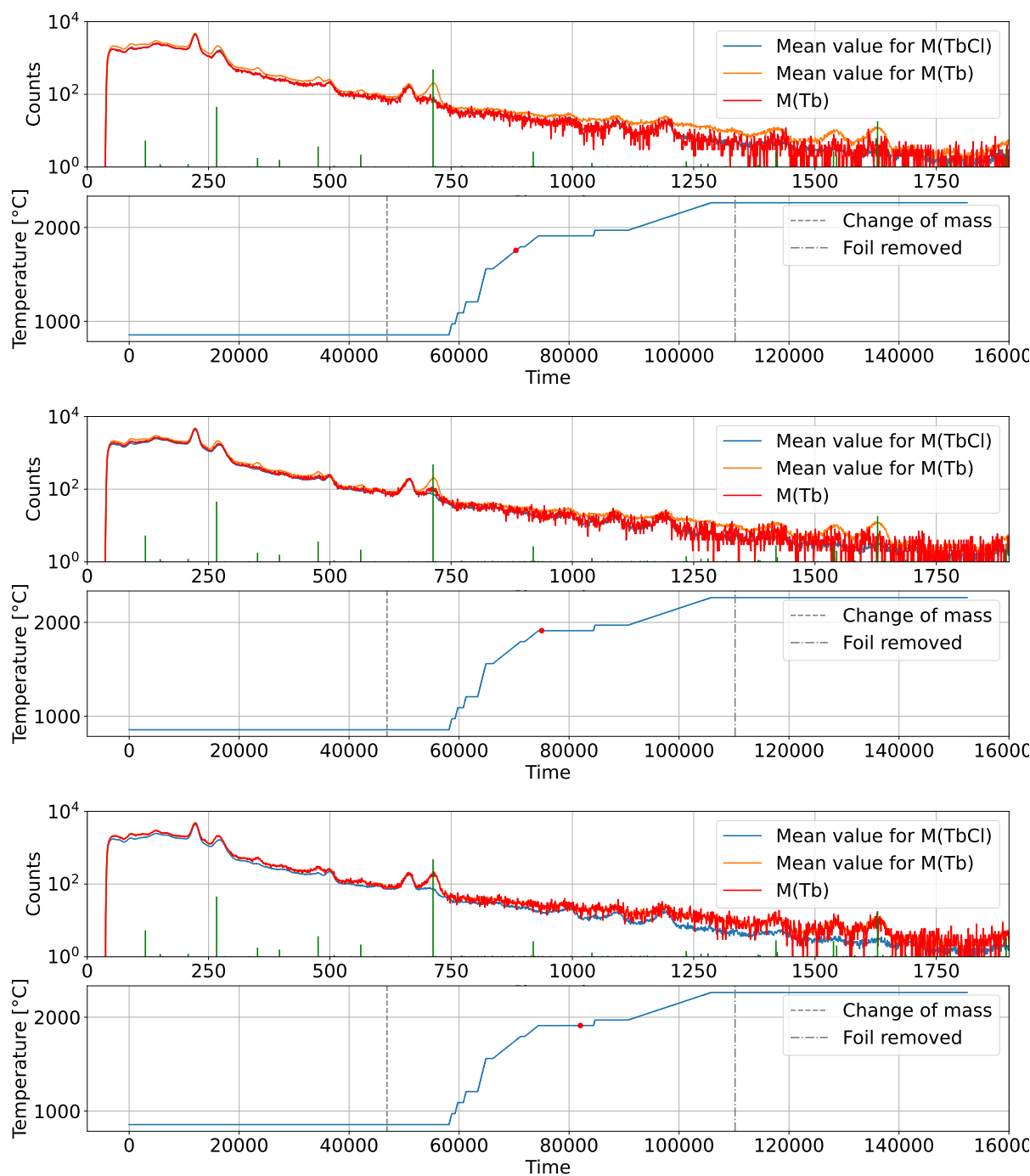


Figure 2.8: Summarizing graphs on the collection of July 11. Both the location in the temperature profile (red dot) and the corresponding spectrum (counts as a function of channel number) is given, together with the mean value when the mass separator was on the mass of Tb or TbCl. The dashed line indicates where the configuration of the mass separator was changed from TbCl to Tb. The green vertical lines indicate the energy lines corresponding to ^{156}Tb , the height of these lines corresponds to their intensity.

2.4 Collection of July 15

In 2020, collections were performed at MEDICIS using targets coming from ARRONAX. The used production reaction was identical to the one used at PSI, but much larger efficiencies were obtained. For these collections, elution was performed with nitric acid (HNO_3) instead of hydrochloric acid (HCl). Therefore, for the collection of July 15, elution at PSI was also performed with nitric acid (HNO_3) in order to obtain a target similar to the ones that were used in 2020 from ARRONAX. Again, the target was a $^{156}\text{Gd}_2\text{O}_3$ (99mg) target that was irradiated for 16h with protons of 22.8MeV (beam current: $25\mu\text{A/h}$). Two aluminum-coated gold foils were loaded in the collection chamber, M191 and M193 and the mass separator was set to mass 155u. The activity of ^{155}Tb at the beginning of the collection present in the target was 950.24MBq and the amount of terbium compared to gadolinium is given by $\frac{Tb}{Gd} > \frac{10^{12}}{20}$. A rhenium ion source and rhenium boat were used in combination with a tantalum container. The collection started on July 15 (12h52), moreover, outgassing was performed during the weekend (16/07/2022-17/07/2022), i.e., the target was heated towards 200A (corresponding to 948.98°C), which is generally too low for terbium to be collected, but some contaminants will be released during this heating. After outgassing (18/07/2022-11h52), the collection spanned 44.8 hours. A summarizing graph is presented in Figure 2.9.

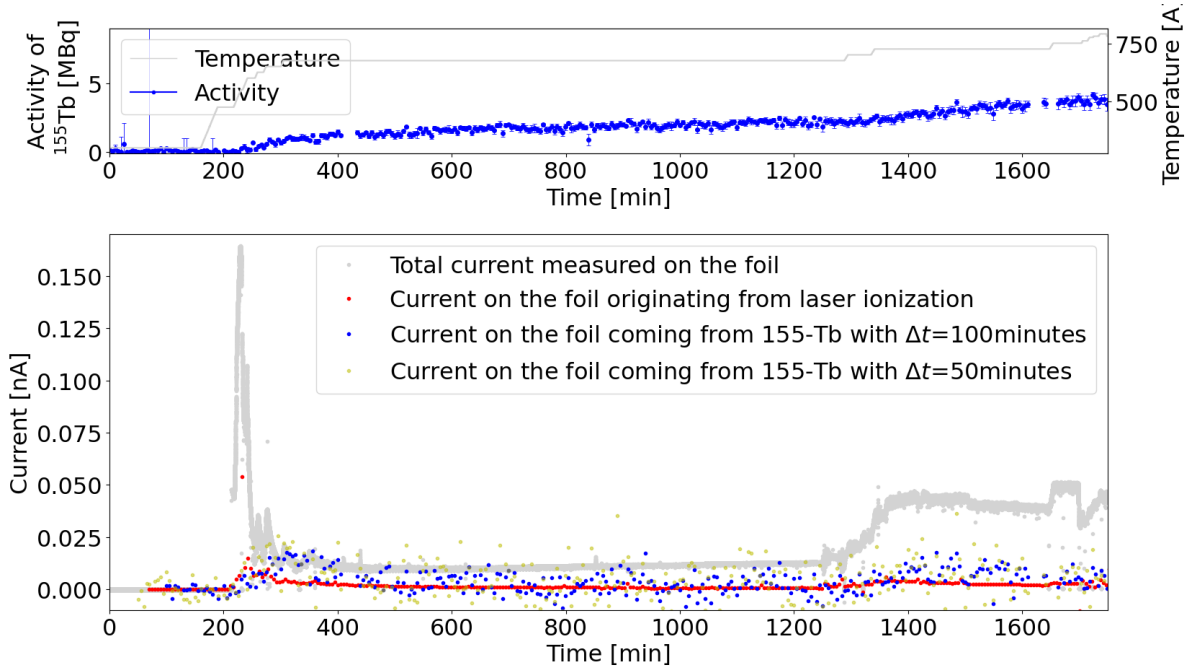


Figure 2.9: Summarizing figure for the ^{155}Tb collection started on July 15. In the upper plot, both the temperature (gray) and the activity of ^{155}Tb measured with the Kromek (blue) are shown as a function of the time after the start of the collection. In the other plot, the total current measured on the foil, the current originating from laser resonance ionization and the current on the foil that is calculated from the activity of ^{155}Tb are shown as a function of the time after the start of the collection.

For this collection, a laser ON-OFF effect is already present ($\frac{ON}{OFF} = \frac{0.045\text{nA}}{0.0438\text{nA}}$) at the moment of the Faraday cup removal. Moreover, the activity corresponding to ^{155}Tb increases immediately after removal of the Faraday cup, indicating that ^{155}Tb is already collected at 300A which corresponds to 1316.2°C. Nevertheless, at 250 minutes after the start of the collection, the current on the foil, the laser ON-OFF current and the current corresponding to the activity of ^{155}Tb start to decrease. The total beam current is approximately 20nA, showing that this decrease is not caused by saturation of the ion source. At about 30 minutes before the heating of the target towards 700A, the current on the foil and the laser ON-OFF effect start to increase again. Moreover, the activity of ^{155}Tb also increases during this heating. This indicates that ^{155}Tb is being collected. However, from 1500 minutes onwards, the laser ON-OFF current and the current corresponding to the activity of ^{155}Tb start to decrease again while the current on the foil stays constant. Probably the target is depleted and mainly LaO is being collected. At around 1750 minutes after the start of the collection, the foil is replaced by foil M193.

After the collection, a measurement with a high purity germanium detector was performed on both foils to determine the activity of ^{155}Tb collected on the foil. It was found that 2.919(133)MBq was present on the first foil at the moment of foil removal. For the second foil, an activity of ^{155}Tb of 0.579(37)MBq was found (corrected for the decay). Thus, the efficiency of this collection is 0.3436(136)%, which is comparable to the efficiency obtained in the collection that started on July 4.

Conclusion

The analysis of the three collections conducted at MEDICIS during the summer of 2022 has provided valuable insights into the production of ^{155}Tb . Several conclusions can be drawn based on the findings. To begin with, it is noticed that although chemical separation of ^{155}Tb and ^{155}Gd is performed, contaminants, such as ^{155}Gd originating from the decay of ^{155}Tb , are still collected. Therefore, the Kromek measurements and the laser ON-OFF current are essential to distinguish whether ^{155}Tb or a contaminant is mainly collected. Another crucial aspect during the collection process is to investigate the isotope of terbium being collected. Lastly, the analyzed collections exhibited lower efficiencies (0.3%-0.6%) compared to previous collections (1.6%-6.1% for targets irradiated at ARRONAX where the same production reaction was employed). This indicates that although the production mechanism of terbium is understood, there are still unresolved issues affecting the efficiency. It is postulated that the underperformance of these collections may be attributed to the different chemical preparations of the irradiated targets prior to the collection. More specifically, in the first two collections of 2020, elution was performed with HCl while at ARRONAX, HNO_3 is used. Consequently, for the third collection, nitric acid was used as an eluent at PSI. Nevertheless, elution with HNO_3 at PSI did not yield a significant improvement in the collection efficiency, probably due to the difference in concentration of HNO_3 . Therefore, in the next chapter, a discussion on the target preparation is presented and subsequently a global conclusion on the ISOL method for externally irradiated targets is provided.

3

Importance of the target preparation

In this chapter, to obtain a better understanding of the low efficiency of the collections performed in 2022, the different target preparation methods for externally irradiated targets are investigated. Both the chemical preparation (elution, see Section 3.1) and the irradiation conditions (energy and current of the protons and the target material) are considered.

The comparison based on the chemical preparation is discussed in Section 3.2. In particular, the collection of July 4 (2022) and July 15 (2022) are considered, as two different eluents were used, HCl and HNO₃ respectively.

The comparison based on the irradiation conditions is discussed in Section 3.3. Here, the collection of August 3, 2020 and July 15, 2022 is utilized. The collection of August 3 was performed with targets received from ARRONAX [46]. For both collections, HNO₃ was utilized as an eluent, but the irradiation conditions differ.

3.1 Elution

Elution is a chemical analysis technique that is used for separation of two elements: X and Y, for example terbium and gadolinium. A column, consisting of narrow-bore tubes, is packed with a finely divided inert solid (= adsorbent) [47]. First, a sample containing both X and Y is introduced to the column, both X and Y can bind to the adsorbent (see Figure 3.1). These components X and Y are then forced through the column by continuously adding eluent (mobile phase in Figure 3.1). As the eluent travels down the column, it can bind to an adsorbent, which was initially bound to a component (X or Y), and thus it can cause descent of the component. This process depends on how strongly the component is retained by the adsorbent, and thus if component Y is more strongly retained, component Y will move down slower. Consequently, component X will reach the end of the column earlier than component Y as is shown in Figure 3.1. Therefore, separation of component X and Y or thus terbium and gadolinium is possible by changing the reservoir at the end of the column at specific times [47].

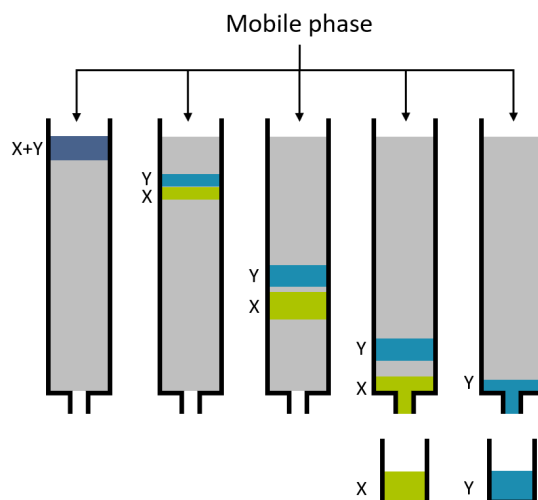


Figure 3.1: Visualization of column chromatography separation.

3.2 Importance of the eluent

The main difference between the collection of July 4, 2022 and July 15, 2022 is the eluent used during the chemical separation. As was discussed previously, it is proposed that for the collection of July 4, the chlorine of the eluent reacted with the terbium inside the target and therefore terbium chloride (boiling point = 180-200°C) was formed. This hypothesis is supported by the dose rate measurement that is performed inside the MEDICIS bunker. The dose rate probe is shown in Figure 3.2 with respect to the target and is mainly used for safety issues. In general, a change in the dose that is measured indicates that activity is being transported in the MEDICIS bunker.



Figure 3.2: Dose rate probe (red) with respect to the MEDICIS target (blue).

The dose rate measurement together with the temperature is shown in Figure 3.3 for the collection of July 4¹. It is observed that the dose increases when the temperature of the target is around 450°C. This temperature is too low for terbium to be collected in

¹For this graph, and all the others in this chapter, a linear fit was made based on the target calibration to obtain the temperature [°C] from the current on the target. This is justified as we focus on temperatures below 1000°C.

elemental form (boiling point terbium = 3230°C [48]). Therefore, a proposed hypothesis is that terbium chloride (boiling point = $180\text{-}200^{\circ}\text{C}$) is vaporized from the target and transported towards the ion source, causing an increase in the dose that is measured with the dose rate probe. Nevertheless, the released terbium chloride is not collected on the foil as it is most likely not ionized [43].

Moreover, the pressure on the front-end (see Figure 3.4) shows an increase just before the increase in the dose, indicating that something is being vaporized from the target.

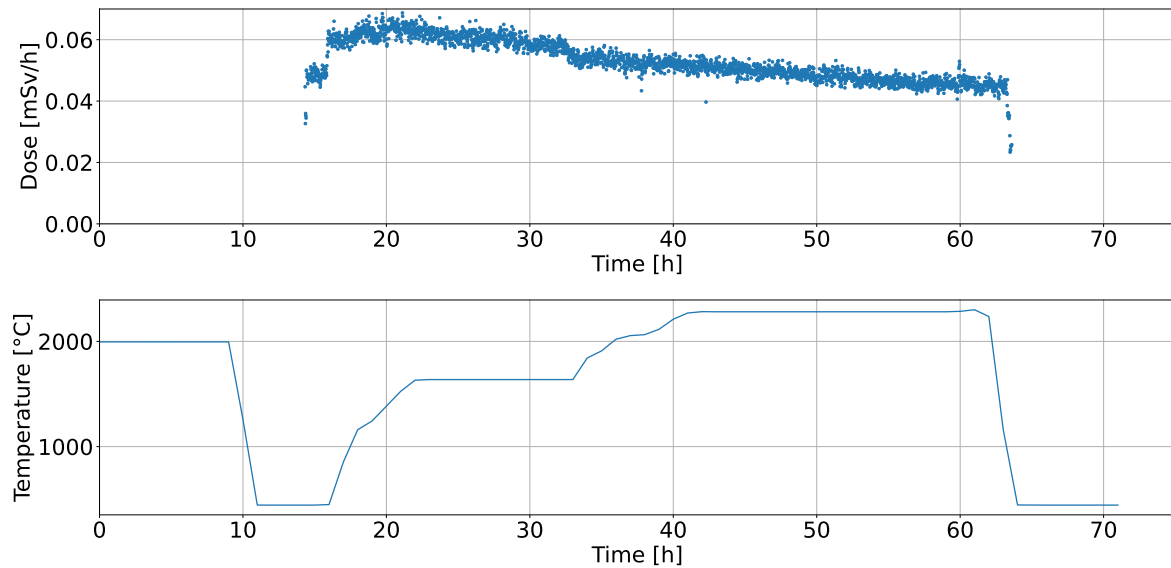


Figure 3.3: Dose rate measurement together with the temperature of the target as a function of the time for the collection that started on July 4.

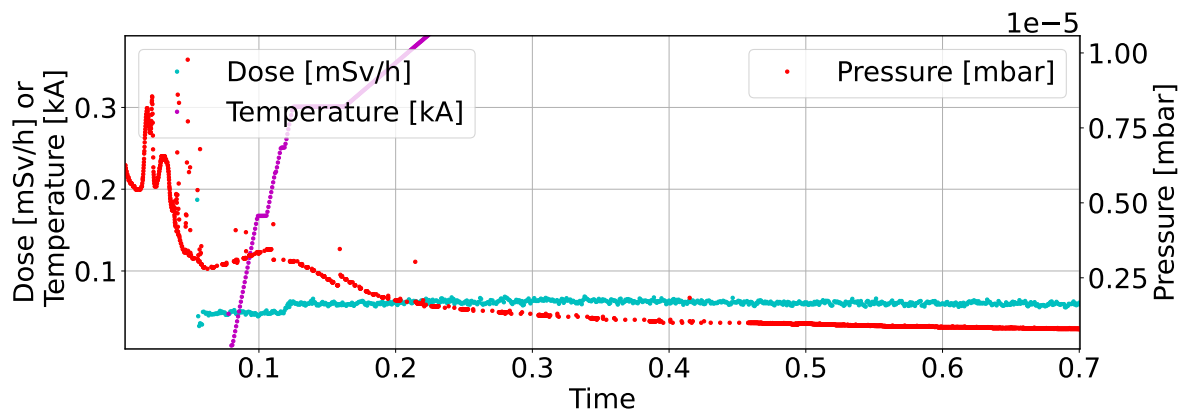


Figure 3.4: Overview of the dose, the pressure on the front end and the temperature during the collection that started on July 4 as a function of the time after the start of the collection. The dose and temperature are read on the left axis, whereas the pressure is indicated on the right axis.

For the collection that started on July 11, similarly as for July 4, HCl was used as an eluent. Therefore, a similar behavior is expected for the July 11 collection. When considering the dose together with the temperature (Figure 3.5) a similar behavior is indeed seen with an increase of the dose at around 634°C. However, when considering the pressure (Figure 3.6), no increase in the pressure at the moment of the dose increase can be observed. Thus, no clear conclusion for the formation of terbium chloride can be drawn based on these collections and further research on the formation of terbium chloride is planned.

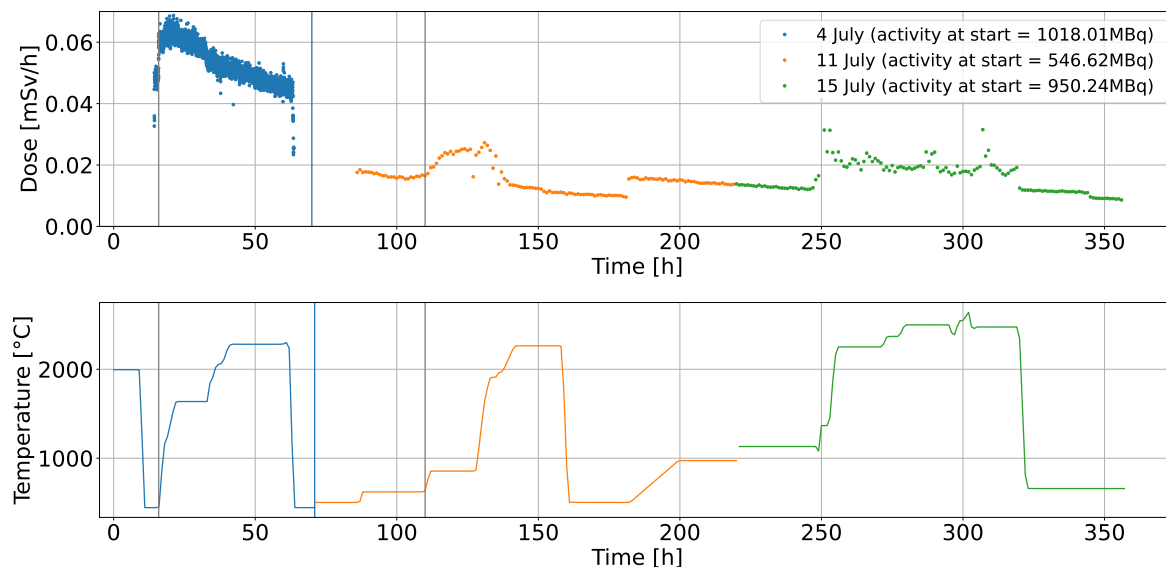


Figure 3.5: Overview of the dose and the temperature during all three collections of 2022 as a function of the time after the start of the collection. The vertical blue line indicates a jump in time. The two gray lines indicate the hypothetical time at which terbium chloride was released.

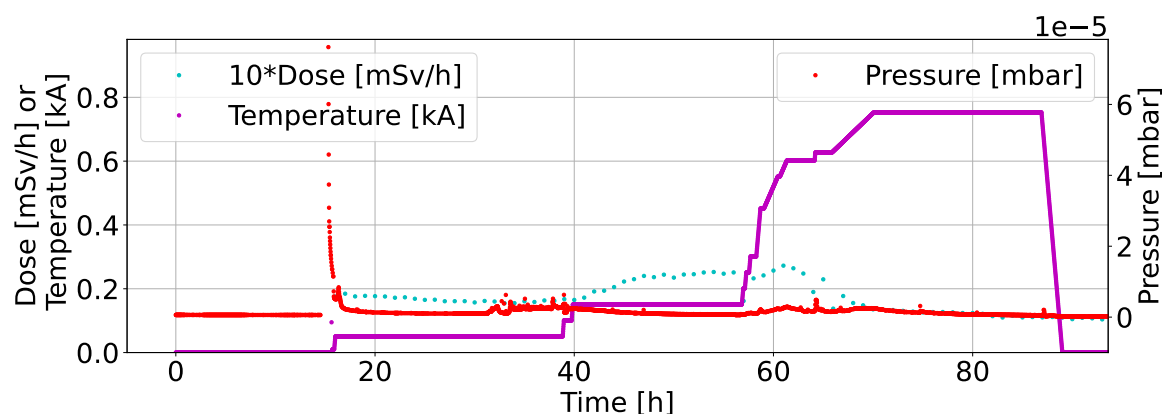


Figure 3.6: Overview of the dose, the pressure and the temperature during the collection that started on July 11 as a function of the time after the start of the collection.

3.3 Importance of the irradiation conditions

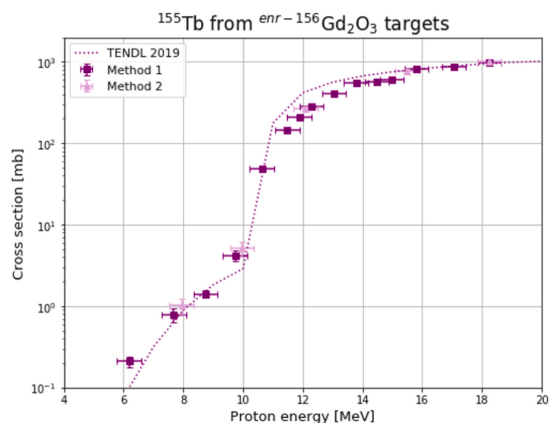
	15/07/2022	3/08/2020
Facility	PSI	ARRONAX
Target material	$^{156}\text{Gd}_2\text{O}_3$ (99mg)	$^{nat.}\text{Gd}_2\text{O}_3$ (0.3114g)
Proton energy	22.8MeV	34MeV
Current of the beam	25 $\mu\text{A}/\text{h}$	30 $\mu\text{A}/\text{h}$
Time of irradiation	16h	12h08min
Eluent	HNO_3	HNO_3
Activity of ^{155}Tb in target at start collection	950.24MBq	299MBq
Tb to Gd ratio	$> 10^{12}/20$	1/5.2
Ion source	Re ion source (726M)	Re ion source (671M)
Laser scheme	both A and B	only A
High voltage	60keV	60keV
Efficiency	0.003436(14)	0.0126(26)

Table 3.1: Full comparison for the collections of July 15, 2022 and August 3, 2020.

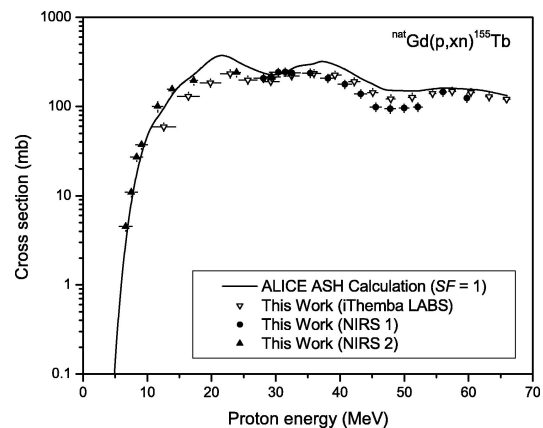
In order to investigate the effect of the proton energy, the used target material and the proton current on the collections, a summary of the collection of July 15, 2022 and August 3, 2020 is given in Table 3.1. For the collection of 2022, the activity of ^{155}Tb after irradiation is equal to 2192.4MBq and for the collection of 2020, the activity of ^{155}Tb equals $\approx 928.77\text{MBq}$. Contrastingly, the efficiency for the collection of 2020 is almost 10 times better than for the collection of 2022: for the collection of 2022, $\varepsilon = 0.3436(136)\%$ is found and for the collection of 2020, $\varepsilon = \frac{2.89(59)\text{MBq}}{229\text{MBq}} = 1.26(26)\%$ is found.

Several explanations are now provided for the difference in efficiency and the difference in activity of ^{155}Tb present in the target. The activity of ^{155}Tb present in the target is larger for the 2022 collection than for the collection of 2020, the reason for this is two-fold. First, by slight extrapolation of Figure 3.7a and by using Figure 3.7b, it is concluded that the cross section for irradiation of enriched $^{156}\text{Gd}_2\text{O}_3$ with 22.8MeV protons is larger than irradiation of natural Gd_2O_3 with 30MeV protons. Note that the proton-beam energy decreases when traversing the sample. Secondly, the irradiation corresponding to the collection of 2022 was performed with a larger integrated current, 400 μA for 2022 and 218.33 μA for 2020. Therefore, the number of incoming particles, and thus the yield, increases for the collection of 2022 compared to 2020.

From Section 3.2, it was concluded that the chemical preparation is also important. The chemical preparation of both collections is similar as the same eluent is used, but the eluent that is used in the last step of the preparation has a different concentration: 0.05M and 0.01M for the collection of 2022 and 2020 respectively. In [49], for ^{161}Tb which has the same chemical properties as ^{155}Tb , it was found that the purity of ^{161}Tb after elution is strongly dependent on the concentration of eluent. This could explain why the fraction of ^{155}Tb to ^{155}Gd was much larger during the collection of July 15, 2022 ($> 0.5 \cdot 10^{11}$) than for the collection of August 3, 2020 (1/5.2).



(a) Cross section for irradiation with protons of different energies of enriched $^{156}\text{Gd}_2\text{O}_3$. (figure retrieved from [12])



(b) Cross section for irradiation with protons of different energies of natural Gd_2O_3 . (figure retrieved from [50])

Figure 3.7: Cross section for irradiation of a gadolinium target as a function of the proton energy.

Although the amount of Tb compared to Gd is larger for 2022, the efficiency is smaller. Different reasons are proposed based on the concentration of eluent. The higher concentration of HNO_3 in the collection of 2022 could facilitate the formation of more Tb_2O_3 atoms than for the collection of 2020. Consequently, these terbium oxide atoms are not released from the target due to their high boiling temperature (3123°C [51]). Another possible explanation is that the higher concentration of HNO_3 may lead to the formation of a precipitate together with the terbium. As a result, the physical contact with the rhenium will differ, leading to a change in thermal contact. This change in thermal contact could affect the efficiency of the collection process.

4

The ISOL method - Conclusion and outlook towards future collections and facilities

As a closing remark on this part, Equation 1.9 will be revisited:

$$\varepsilon_{\text{tot}} = \varepsilon_{\text{diff}} \cdot \varepsilon_{\text{eff}} \cdot \varepsilon_{\text{ion}} \cdot \varepsilon_{\text{sep}} \cdot \varepsilon_{\text{transport}} \cdot \varepsilon_{\text{implant}} \cdot c_{\text{decay}} \quad (4.1)$$

Here, $\varepsilon_{\text{diff}}$, ε_{eff} and ε_{ion} correspond to different steps of the ISOL method: diffusion, effusion and ionization. ε_{sep} and $\varepsilon_{\text{transport}}$ are related to the mass separation and the transportation of the beam towards the implantation foils. Those efficiencies are very high and can be approximated by 100% [46]. $\varepsilon_{\text{implant}}$ is related to the self-sputtering, which will be addressed in Part II. Lastly, c_{decay} is a factor that takes into account the radioactive decay during the collection.

$\varepsilon_{\text{diff}}$, ε_{eff} are strongly dependent on the target temperature. The collection of terbium occurs at nominal ISOL operating temperatures, ranging from 1316.2°C to 1929°C. Moreover, temperature gradients within the target container could hinder the release of terbium. Additionally, the choice of eluent plays a role in the release behavior. In particular, for the collection where HNO_3 was used as eluent, terbium is collected at lower temperatures than for the collections where HCl was used as eluent. This observation may be attributed to many effects, ranging from the vaporization of the different molecular compounds to the formation of a precipitate of terbium and HNO_3 , which alters the thermal contact with the rhenium. Furthermore, the results from the July 4 collection provide evidence for gadolinium being extracted earlier compared to terbium as a function of the target temperature. More specifically, the laser ionization current corresponding to scheme A was observed to increase with a delay corresponding to scheme B, suggesting that, most likely, gadolinium is released prior to terbium, but their release temperatures are very similar.

For ε_{ion} , it has been observed that for too high total beam currents (typically $>100\text{nA}$), the ion source can be saturated [23]. However, for the discussed collections, the total beam current is of the order of tens of nA, which is below the saturation threshold. Therefore, ion source saturation should not be a limiting factor in these collections. This relatively low beam current is attributed to the chemical preparation. Hence, for offline collections with an intermediate chemical separation step, ion source saturation effects are less probable compared to collections without chemical preparation.

Lastly, the optimization of c_{decay} can be achieved through the monitoring of the laser resonance ionization current and the activity. These measurements enable the calculation of the amount of ^{155}Tb that is collected in the collection foil. Moreover, they provide an

excellent benchmark for when to stop the collection. A decrease in the Kromek signal or laser-ionization current indicates a lower collection of ^{155}Tb , prompting the removal and shipping of the collection foil from the MEDICIS setup towards a radiochemical facility in order to minimize the loss due to the decay.

Based on this analysis, different improvements that can be made to increase the efficiency of the ^{155}Tb collections are proposed.

Implementing the laser ON-OFF automatically with a period of 5 minutes was a beneficial choice. Together with the current corresponding to the activity, it should be used as a benchmark for when to stop the collection. Additionally, conducting Kromek measurements of the background before and after the collection, both with and without the sample holder present in the chamber, could provide valuable insights of what has been collected and what is a remnant left in the chamber from a previous collection. Moreover, for the activity calculation, it is important that the efficiency calibration of the Kromek detector is reliable. However, it was noted that different efficiency curves as a function of the energy can be fitted to the available datapoints from [39] (see Appendix A). Especially at energies between 100keV and 300keV, the fit is unreliable. Therefore, sources such as ^{133}Ba , ^{152}Eu , ^{241}Am , ^{57}Co , ^{226}Ra , ^{137}Cs , ^{158}Tb and ^{229}Th can be used for new efficiency measurements to improve the reliability of the fit. Nevertheless, due to the large dead time, the accuracy of the detector is limited [39].

Plans are made to collect terbium via its constituent molecules, such as terbium chloride. However, further research is required to gain a more comprehensive understanding of these molecules, as many aspects remain unknown. Additionally, when the aim is to collect elemental terbium, it is imperative that the chemistry of the targets is investigated. Considering the target preparation, irradiation of natural gadolinium with protons is less favorable than irradiating enriched ^{156}Gd . Moreover, consistent investigation into the used eluent is needed in order to optimize the different parameters. Therefore, more facilities evolve towards offline collections in order to have the chemical separation in the same facility. This allows to make the performed collections reproducible by doing systematic research on the effect of the used eluent.

PART II: Self-sputtering

Whatever you do, don't lose
yourself in the process

Tiffany Moule

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5

Self-sputtering

During several previous MEDICIS collections, indications that not all implanted material is retained on the foil were found (i.e., $\varepsilon_{\text{implant}} < 1$) [52]. For example, the ^{155}Tb collections performed during the summer of 2021 showed a large inconsistency ($\approx 26.5\%$) between the activity measured in the collection chamber and the activity measured on the foil once the foil was removed from the collection chamber. This indicates that ^{155}Tb was left in the collection chamber [23]. The reason proposed for this discrepancy is that self-sputtering occurred during the implantation. Consequently, only a fraction of the terbium ions would be retained in the foil, while the rest would be self-sputtered inside the collection chamber. These self-sputtered particles contribute to the activity measured inside the chamber but remain in the collection chamber once the foil is removed. Therefore, a smaller activity is measured on the foil after removal. Another indication for self-sputtering was found by measuring the repeller plate, which is a plate that is installed in between the collimator and the collection foil in the backward direction of the beam. When measuring this plate after the winter shut-down of MEDICIS, ^{139}Ce was detected, indicating that something had moved antiparallel with respect to the beam, suggesting that those ^{139}Ce particles were (self-)sputtered from the collection foil.

The purpose of this second part of the thesis is to characterize the systematic effects of self-sputtering for different radionuclide collections. Hence, simulations using TRIDYN (TRIm simulation code including DYNamic composition changes, [53, 54, 55]) will be performed in order to simulate the self-sputtering behavior. This chapter serves as a stepping stone towards an understanding of self-sputtering by introducing the physical processes regulating sputtering and the TRIDYN simulations.

5.1 Physical principle

When an ion is implanted in a foil, the ion interacts both with the electrons and the nuclei of the foil material, leading to electronic and nuclear stopping effects, respectively. At large energies (typically $> 1\text{MeV}$), the electron-ion interaction cross section is larger than the nucleus-nucleus interaction cross section. Thus, for larger energies, the ion dominantly loses its energy by interactions with the electrons. At medical collection facilities, the voltage difference between the target and the extraction electrode is typically 30-60kV, leading to an ion energy of 30-60keV. Therefore, the nuclear stopping effects dominate

rather than electronic stopping effects (energy lost per distance travelled: $\frac{dE}{dx}|_{\text{nuclear}} \approx -300 \frac{\text{eV}}{\text{\AA}}$ and $\frac{dE}{dx}|_{\text{electronic}} \approx -2 \cdot 10^{-10} \frac{\text{eV}}{\text{\AA}}$ for terbium implanted in aluminum).

If an ion impinges on the foil, it collides with the nuclei in the foil. In these collisions, some momentum is transferred to the foil nuclei, which in turn can transfer their energy to other foil nuclei. In that way, a nuclear collision cascade emerges and eventually a certain fraction of the foil nuclei obtain a momentum in the outward direction. If such a nucleus is located near the surface of the foil, it can be removed from the foil; in other words, the nucleus has *sputtered* away from the foil. If the sputtered element is of the same ion species as the ones impinging, the sputtering process is called *self*-sputtering. Self-sputtering can occur during isotope collections. More specifically, as the number of incoming ions on the implantation foil increases, more of the foil material is sputtered away, removing a thin layer from the sample. Therefore, the surface of the foil moves closer to the layer of previously implanted ions (and thus ions will be implanted deeper) and eventually the previously implanted ions will be sputtered away, as is visualized in Figure 5.1. This process is enhanced if the atomic number of the ion is larger than the atomic number of the foil, as more implanted ions will, in this case, increase the stopping power of the foil. The latter increases the amount of self-sputtering as new ions are implanted closer to the surface.

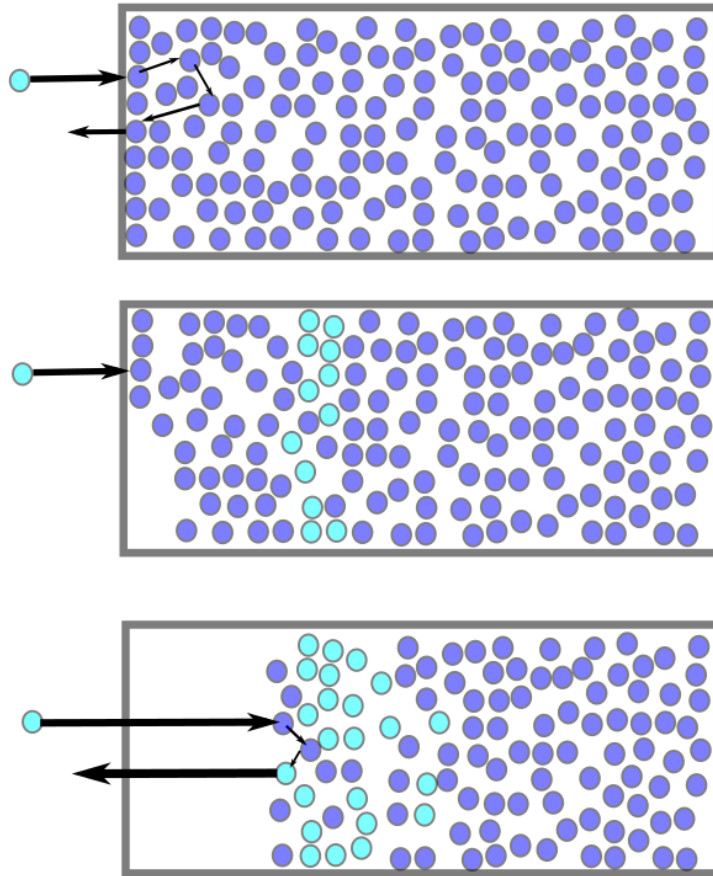


Figure 5.1: A simplified visualization of the self-sputtering process: as more ions are implanted, the foil is slowly sputtered away. Thus, the previously implanted ions move closer towards the surface until they are eventually sputtered out.

A self-sputtering curve shows the retained fluence as a function of the implanted fluence. The latter equals the fluence (number of particles per unit area) of particles that is incoming, whereas the former corresponds to the amount of these incoming particles that is retained on the foil per unit area. A typical self-sputtering curve is given in Figure 5.2. Indeed, in the beginning, where no self-sputtering has occurred yet, the retained fluence is equal to the incoming fluence. However, as the incoming fluence increases, self-sputtering starts to occur: the foil material is sputtered away and eventually the initially implanted ions reach the surface and are self-sputtered away, leading to the strong decrease of the retained fluence compared to the incoming fluence (in this case around $10\text{\AA}$).

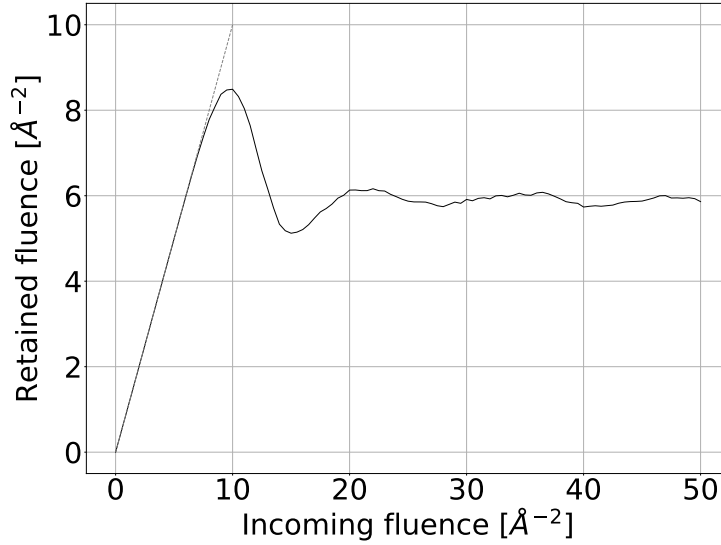


Figure 5.2: A typical self-sputtering curve, the retained fluence is plotted as a function of the incoming fluence, the $y = x$ line corresponds to the gray dashed line.

This behavior has very strong implications, e.g., if more particles are impinging on the foil, a smaller retained fluence can be observed. Additionally, a slight oscillation can be resolved, which can be explained as follows in the case of terbium implanted in aluminum: as the fluence of terbium increases, more aluminum is sputtered away, and thus the implanted terbium moves closer towards the surface. Consequently, the average Z of the foil increases and thus the implantation depth decreases (as the stopping power increases). As the terbium moves towards the surface, more terbium is sputtered away, yielding a smaller retained fluence. This decreases the amount of terbium present in the foil (mainly near the surface) and thus both the sputtering yield for terbium and the average Z decrease, implying that more terbium is retained deeper in the foil. In other words, the foil has undergone a ‘soft reset’, i.e., the terbium at the surface is mostly removed and terbium is again implanted at a larger depth. Several of these iterations are needed to reach the equilibrium state.

Different physical models have been proposed to describe the sputtering mechanism, such as Sigmund’s theory, which is based on Boltzmann transport theory [56]. However, the primary objective of this thesis is to describe the systematic effects of self-sputtering rather than providing a theoretical description of self-sputtering. Therefore, the phenomenology of self-sputtering is discussed by using simulations performed in TRIDYN.

5.2 TRIDYN

The simulations for self-sputtering were, throughout this whole thesis, performed in TRIDYN. TRIDYN is a Monte Carlo based simulation package that allows to keep track of dynamic changes to the target. In the Monte Carlo simulations, the binary collision approximation is used to model the collisional processes. Contrastingly to SRIM (Stopping and Range of Ions in Matter, [57]), dynamical simulations can be performed, where changes to the target due to implantation and sputtering are taken into account. Moreover, different radiation conditions can be applied simultaneously or subsequently via layered simulations, in which the depth profile at the end of the previous step is taken as a start profile for the target in the subsequent simulation. Further investigation into these layered simulations can be found in Section 8.2.1.

The parameters that are given as inputs in the simulations are depicted in Table 5.1.

	Code	Explanation
First line	filename+description	The first 10 characters are used as the name for the output files, thereafter, a description of the simulation can be given
cdata	x y z	x: Initial composition 0 for constant fractions over the whole depth 1 for fractions that vary with the depth (determined by a depth profile file) y: Total fluence [$\frac{\# \text{Particles}}{\text{\AA}^2}$] z: Mode 0 for static (no update of the target during simulation) 1 for dynamical (update of the target during the simulation)
geom	k l m	k: Thickness of the target [$\text{\AA}$] l: Number of simulation layers in which the target is divided m: Thickness of thin film on top of the target [$\text{\AA}$]
atda	A B C	Symbols of the elements used in the simulation (both target and beams)
comp	a b c	Initial composition of the target (a = fraction of element A, ...) If x = 1, this line should not be provided
irra	o p q r	Irradiation condition, multiple irradiation conditions can be provided in multiple lines: o: Number (corresponding to the sequence of atda) of the element that is implanted p: Energy of the beam [keV] q: Angle of incidence r: Fraction of the total irradiation fluence (sum over r-values of all irradiation conditions should give 1)
rand	XXXXX	5-digit number used as a random seed
outp		Activation of the depth profile output
outi	ardn	ardn gives the areal densities for all fluences
fout	d e f	d: Frequency of the outputs given in the command prompt e: Frequency of the saved ardn outputs f: Frequency of the saved depth profiles

Table 5.1: Parameters used as an input in the TRIDYN simulations.

In order to compare the self-sputtering behavior for different conditions quantitatively, a measure for when self-sputtering becomes significant is needed. Throughout this part, the start of self-sputtering is defined to be the incoming fluence for which the retained fluence equals 90% of the incoming fluence. This fluence is defined as ψ_{crit} and is visualized in Figure 5.3. Moreover, the error on this value is defined both by the systematic error

due to the discreteness of the simulation (σ_Δ) and the error due to the uncertainty on the simulation (σ_{sim}). The former is defined to be the stepsize of the incoming fluences taken in the simulation, whereas the latter is defined from the statistical uncertainty on the simulation, i.e., the incoming fluence for which $\psi_{\text{crit}} \pm \sigma_{\text{TRIDYN}}$ is equal to 90% of the incoming fluence, yielding $\sigma_{\text{sim},1}$ and $\sigma_{\text{sim},2}$ as is shown in Figure 5.3. σ_{TRIDYN} is determined by performing 10 simulations with a different random number seed [58] and is indicated in orange in Figure 5.3. Ultimately, the total error on ψ_{crit} is given by:

$$\left[-\sqrt{\left(\frac{\sigma_\Delta}{2}\right)^2 + \sigma_{\text{sim},1}^2}, +\sqrt{\left(\frac{\sigma_\Delta}{2}\right)^2 + \sigma_{\text{sim},2}^2} \right]. \quad (5.1)$$

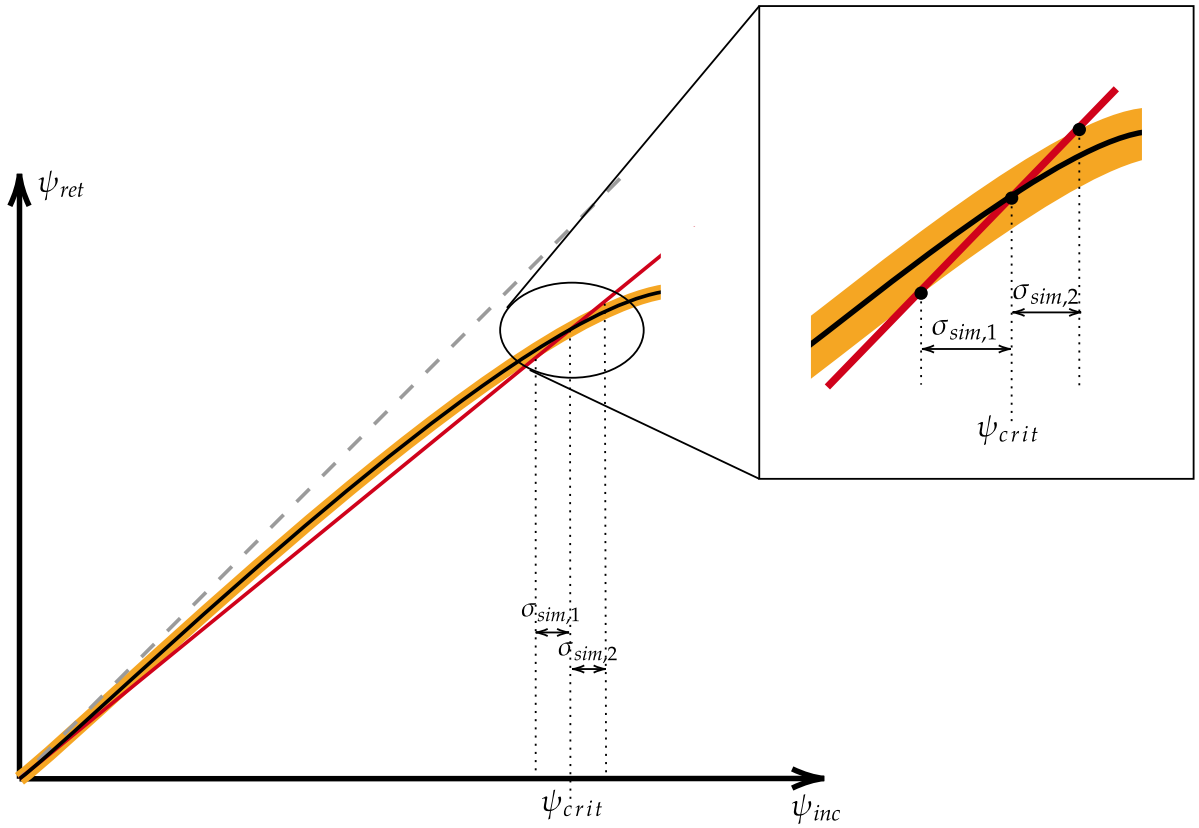


Figure 5.3: Retained fluence (ψ_{ret}) as a function of the incoming fluence (ψ_{inc}), where the dashed gray line indicates the $y = x$ line, the red line corresponds to the $y = 0.9x$ line. The black curve indicates the self-sputtering curve with its statistical uncertainty indicated in orange.

6

Systematic study on the dependence of self-sputtering on the foil and beam

In the collections performed at MEDICIS before 2022, most implantations were performed in zinc-coated gold foils. However, in the more recent collections, aluminum-coated gold foils have been used in order to minimize the self-sputtering, under the assumption that it would reduce this effect. Therefore, investigation into the dependence of self-sputtering on the foil-type will be performed to validate this assumption and to create a framework that could guide further improvements in the future. Lighter elements are not used for implantation foils at MEDICIS due to complications: hydrogen, helium, oxygen and neon are gases, lithium is flammable and quite active in the body, beryllium is very toxic, graphite needs to be burned for extraction of the radionuclide, fluorine and boron are too reactive and separation of nitrogen and the radionuclide through chemical purification is difficult. However, at ISOLpharm at SPES, investigation into implantation in sugars and salts is performed and, therefore, also self-sputtering for these implantation materials is discussed.

At the different production facilities, different potential differences can be used to extract the radioisotopes. Therefore, in Section 6.2.1, the impact of the resulting beam energy on the self-sputtering behavior is explored.

From Chapter 2, it was noted that gadolinium comes out of the target before or simultaneously with terbium, therefore its effect on the self-sputtering will be investigated. Lastly, as was discussed in Part I, there are plans to collect molecular beams of terbium instead of elemental terbium ions. Therefore, a consideration into the implantation of different forms of Tb is made, considering an elemental Tb^+ ion beam in comparison with molecular beams of terbium chloride and terbium fluoride.

6.1 Dependence on the foil material

In this section, investigation into the use of different foil materials is performed; implantations in zinc, aluminum, sugars and salts are considered.

6.1.1 Implantation in zinc

A simulation has been performed for 60keV terbium ions implanted in zinc¹. The result is shown in Figure 6.1, from which it is noted that significant self-sputtering starts around $0.48^{+(14)}_{-(6)} \text{ \AA}^{-2}$ (see Chapter 5 for the formalism to determine this value and the corresponding error). Ten simulations were performed with different random number seeds in order to get an estimate for the statistical errors; both the error on a single simulation and the error on the mean value for ten simulations are provided.

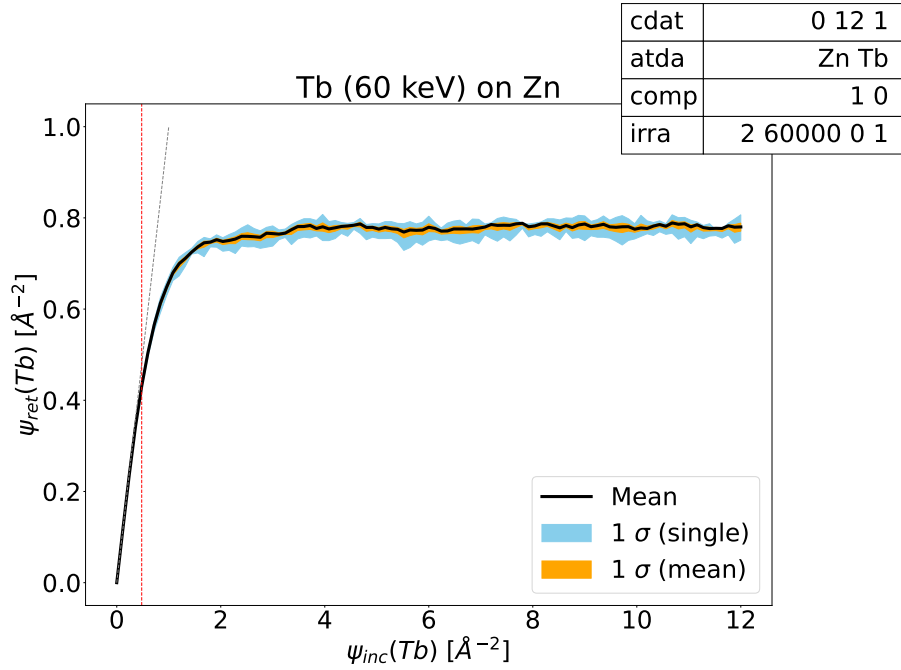


Figure 6.1: Summarizing graph for the implantation of terbium in zinc. The retained fluence (ψ_{ret}) is shown as a function of the incoming fluence (ψ_{inc}) and the red (dashed) line indicates when self-sputtering starts ($\psi_{\text{retained}} < 0.9\psi_{\text{incoming}}$). The gray line indicates the $y = x$ line.

6.1.2 Implantation in aluminum

The results of the simulation for implantation of terbium in aluminum are given in Figure 6.2. Self-sputtering starts to be important around $3.72^{+(6)}_{-(14)} \text{ \AA}^{-2}$. As expected, self-sputtering is less significant for implantation in aluminum than in zinc due to the lower Z-number.

¹For the analysis of the TRIDYN simulations, a code written by Michael Heines was used. However, this code was extended towards including the systematic error, multiple irradiation conditions and layered simulations.

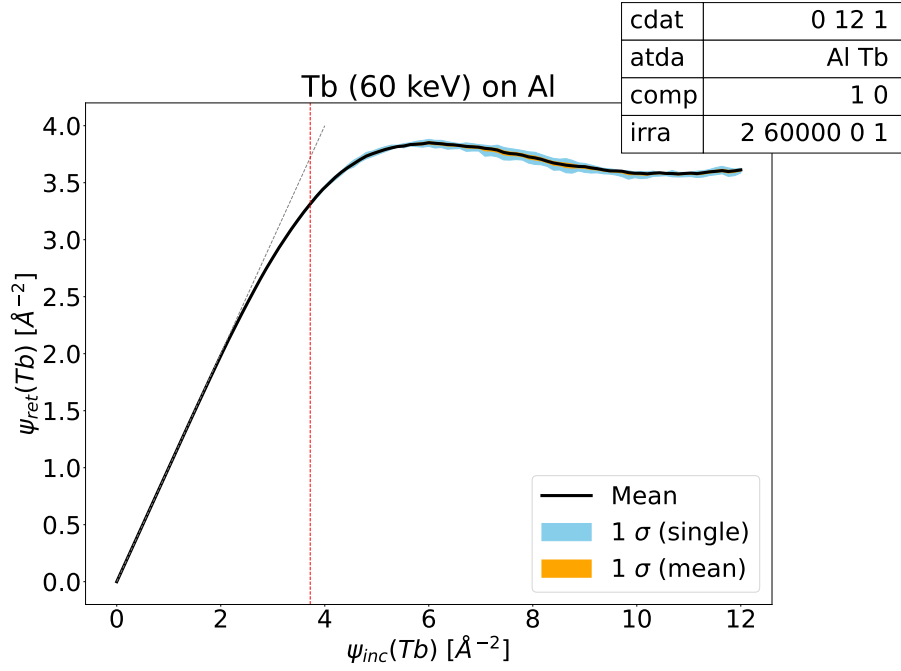


Figure 6.2: Summarizing graph for the implantation of terbium in aluminum. The retained fluence (ψ_{ret}) is shown as a function of the incoming fluence (ψ_{inc}) and the red (dashed) line indicates when self-sputtering starts ($\psi_{\text{retained}} < 0.9\psi_{\text{incoming}}$). The gray line indicates the $y = x$ line.

6.1.3 Implantation in sugar

At ISOLpharm, investigation into the collection in sugar tablets is performed. Two types of tablets are currently researched: cellulose ($\text{C}_6\text{H}_{10}\text{O}_5$) and dextrans ($\text{C}_5\text{H}_5\text{O}_6$). Simulations have been performed for both cellulose and dextrans and the results are shown in Figure 6.3 and Figure 6.4. It is noted that self-sputtering for cellulose and dextrans starts around $11.64^{+(6)}_{-(6)} \text{Å}^{-2}$ and $9.48^{+(14)}_{-(6)} \text{Å}^{-2}$, respectively. This is a substantial improvement compared to implantation in metals, which is mainly due to the fact that the average Z-value of sugar is much smaller than for aluminum or zinc. Despite this improvement, it should be noted that the microstructure (density, porosity, ...) can affect the self-sputtering behavior and that this microstructure is not taken into account in the TRIDYN simulations. This dependence on the microstructure was observed for example in [59], where a different behavior for the self-sputtering in glassy carbon compared to pyrolytic graphite was found.

6.1.4 Implantation in salt

At ISOLpharm and MEDICIS, implantation in salts is researched, where both sodium chloride and sodium nitrate are used. Simulations in TRIDYN have been performed and yielded that self-sputtering starts around $1.68^{+(6)}_{-(6)} \text{Å}^{-2}$ and $3.12^{+(6)}_{-(6)} \text{Å}^{-2}$ for NaCl and NaNO_3 respectively (see Figure 6.5 and 6.6). Surprisingly, the value for NaNO_3 indicates earlier self-sputtering compared to implantation in Al, despite NaNO_3 having a lower Z-number.

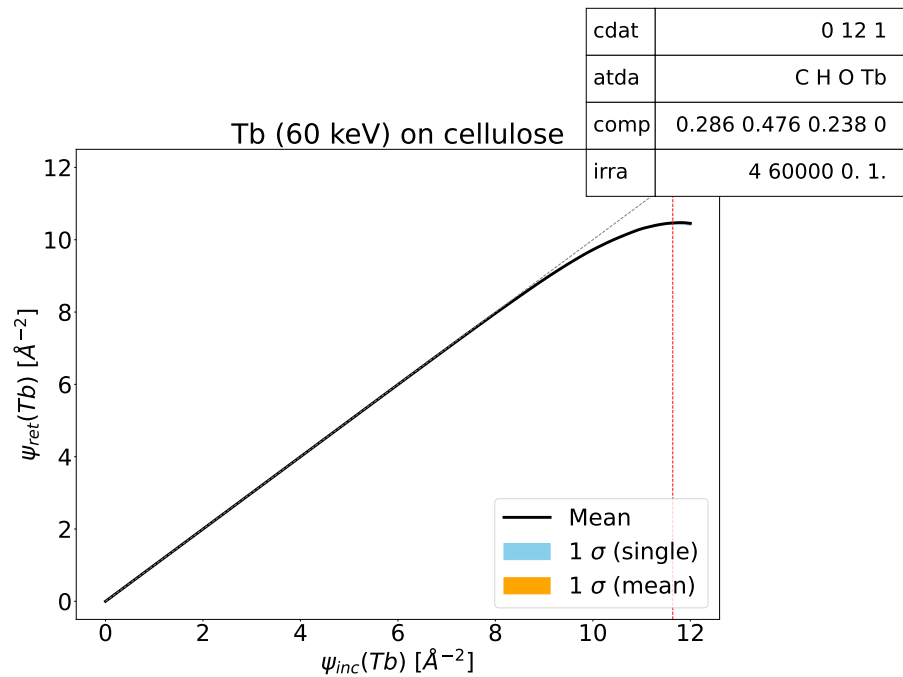


Figure 6.3: Summarizing graph for the implantation of terbium in cellulose. The retained fluence (ψ_{ret}) is shown as a function of the incoming fluence (ψ_{inc}) and the red (dashed) line indicates when self-sputtering starts ($\psi_{\text{retained}} < 0.9\psi_{\text{incoming}}$). The gray line indicates the $y = x$ line.

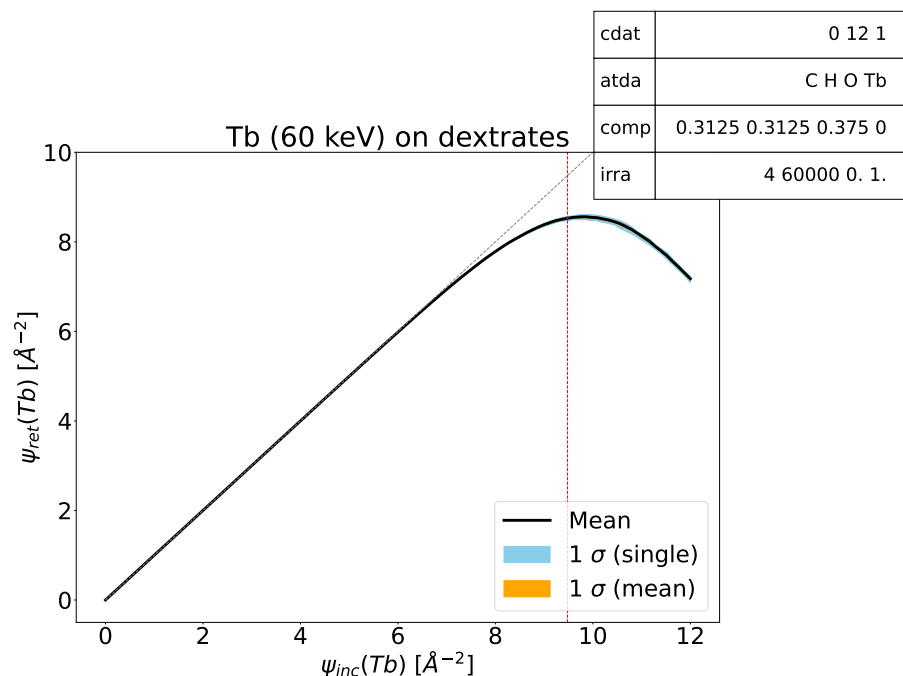


Figure 6.4: Summarizing graph for the implantation of terbium in dextrans. The retained fluence (ψ_{ret}) is shown as a function of the incoming fluence (ψ_{inc}) and the red (dashed) line indicates when self-sputtering starts ($\psi_{\text{retained}} < 0.9\psi_{\text{incoming}}$). The gray line indicates the $y = x$ line.

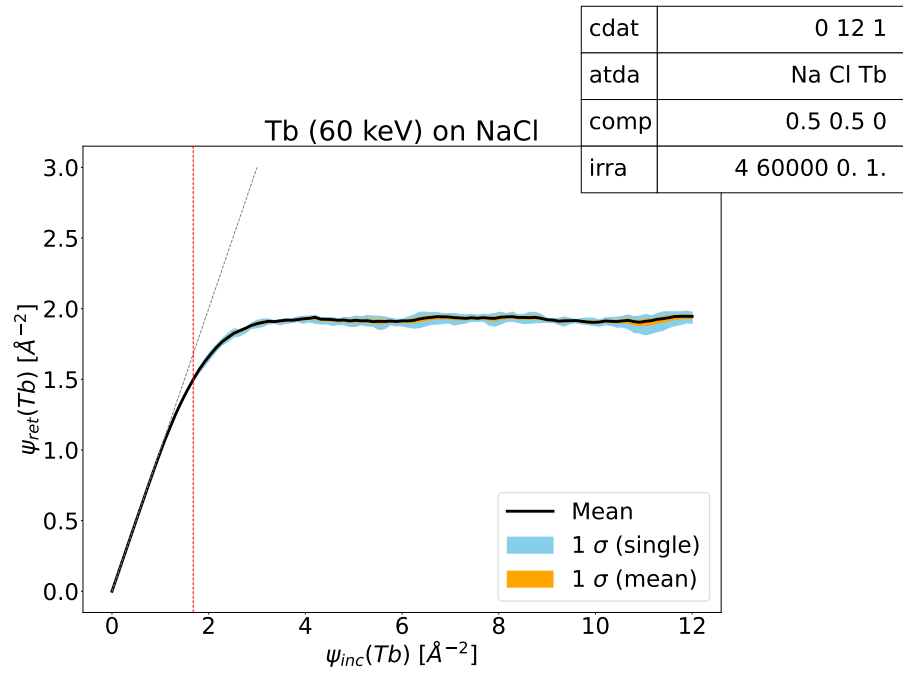


Figure 6.5: Summarizing graph for the implantation of terbium in NaCl. The retained fluence (ψ_{ret}) is shown as a function of the incoming fluence (ψ_{inc}) and the red (dashed) line indicates when self-sputtering starts ($\psi_{\text{retained}} < 0.9\psi_{\text{incoming}}$). The gray line indicates the $y = x$ line.

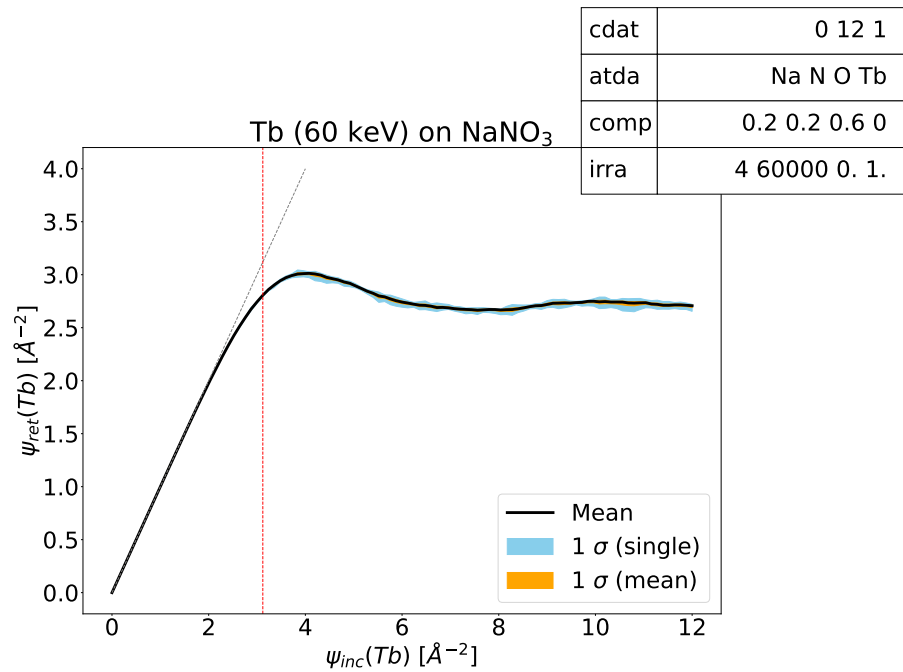


Figure 6.6: Summarizing graph for the implantation of terbium in NaNO_3 . The retained fluence (ψ_{ret}) is shown as a function of the incoming fluence (ψ_{inc}) and the red (dashed) line indicates when self-sputtering starts to be important ($\psi_{\text{retained}} < 0.9\psi_{\text{incoming}}$). The gray line indicates the $y = x$ line.

6.2 Effect of the beam characteristics

In this section, investigation into the effect of the beam on self-sputtering is performed. First, the impact of the energy of the beam on self-sputtering is considered. Thereafter, the effect of a non-pure Tb beam is investigated. Lastly, the implantation of molecular beams of terbium are compared.

6.2.1 Effect of the energy of the beam

The high voltage applied on the target compared to the rest of the beamline, and thus the energy of the impinging ions, can be varied at the different collection facilities. Therefore, in order to choose an appropriate energy, investigation into self-sputtering as a function of the energy of the impinging beam is imperative. Simulations for energies ranging from 30keV to 200keV were performed and the results are provided in Figure 6.7 and Table 6.1.

A strong increase in the incoming fluence at which self-sputtering becomes important is observed for increasing beam energies, mainly due to the deeper implantation. Thus, it is recommended to keep the target at the highest high voltage possible. Nevertheless, the voltages applied on the target are limited due to stability-issues² and the availability of transformers. For example, the maximum high voltage at MEDICIS is 60kV.

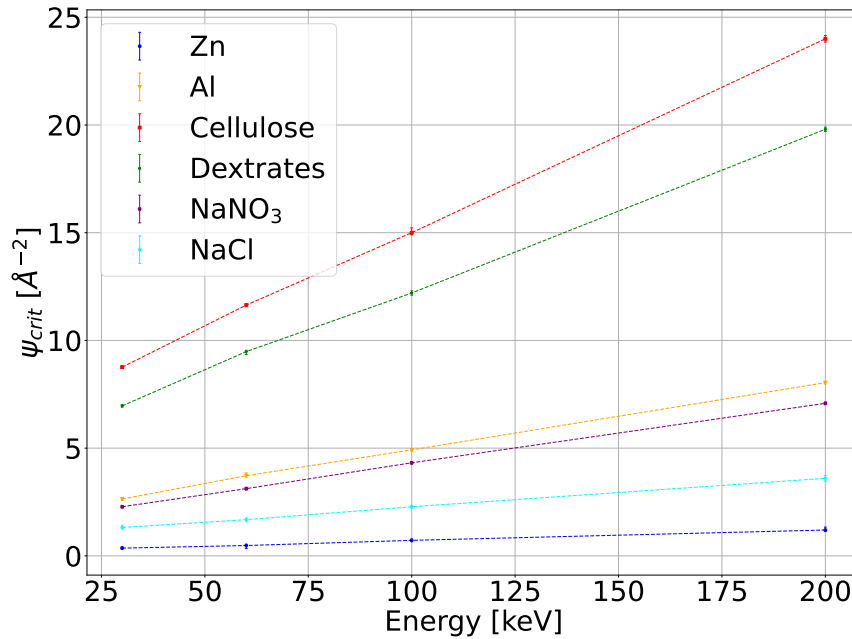


Figure 6.7: Fluence at which self-sputtering starts (ψ_{crit}) as a function of the energy for implantation in different foils for implantation of terbium.

²If the voltage on the target increases, the electrical field gradient becomes too large, resulting in ionization and subsequent discharge of the electric field due to the finite vacuum.

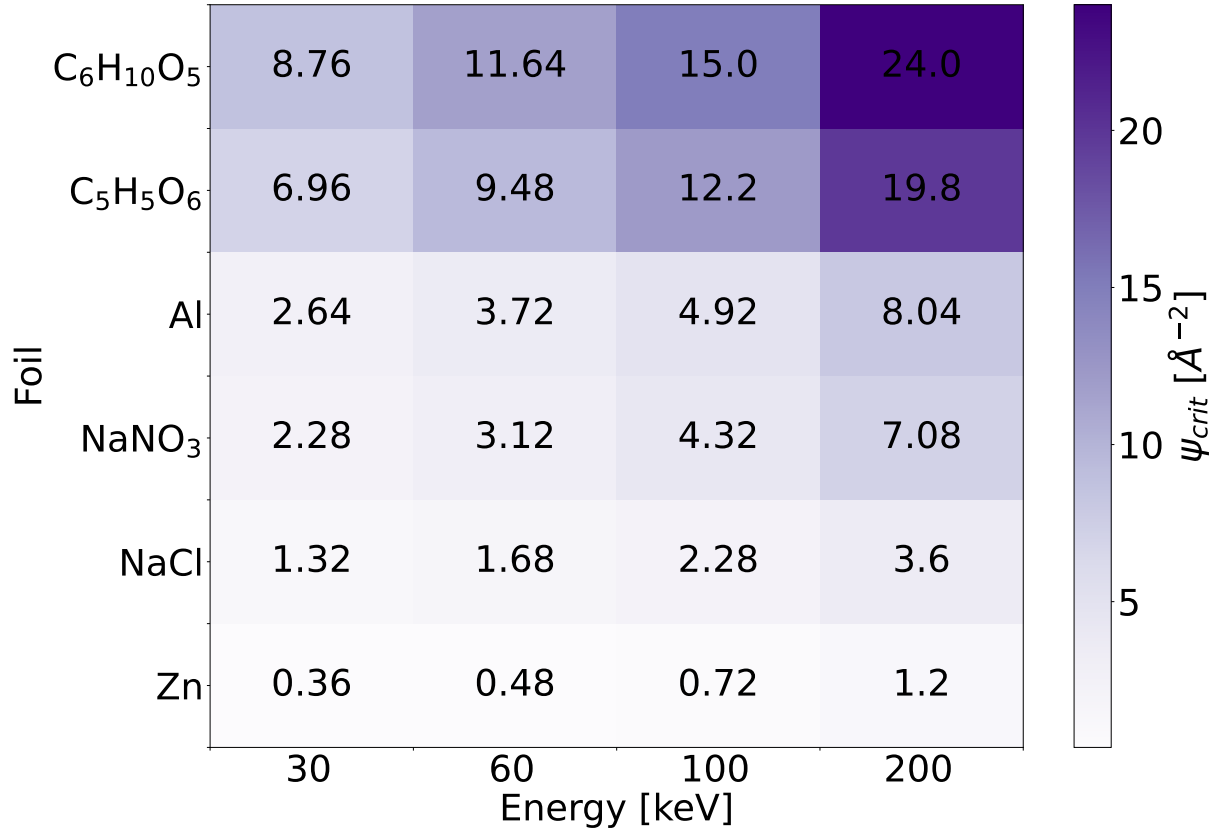


Table 6.1: Incoming fluence of terbium at which the self-sputtering starts (ψ_{crit}).

6.2.2 Effect of a non-pure beam

In the analysis of the different collections, it was observed that gadolinium is also extracted from the target. Currently, everything is implanted in the same foil, however upon implantation of gadolinium, more high-Z elements are present in the target. As a consequence, the implantation depth decreases while the sputter yield increases, both leading to more self-sputtering of Tb. Therefore, in this section, the effect of the amount of gadolinium that is implanted is explored.

Gadolinium implanted prior to terbium implantation

To investigate the effect of gadolinium implanted before terbium implantation, different simulations were performed. The aluminum foil is first irradiated with a pure gadolinium beam, followed by a simulation of irradiation with a pure Tb beam on the gadolinium-irradiated foil. The gadolinium fluence that is implanted is varied between $0\text{\AA}^{-2}$ and $4\text{\AA}^{-2}$ (reasonable values considering the collection of July 2021, see Figure 6.8) and the fluence of terbium is kept constant at $10\text{\AA}^{-2}$.

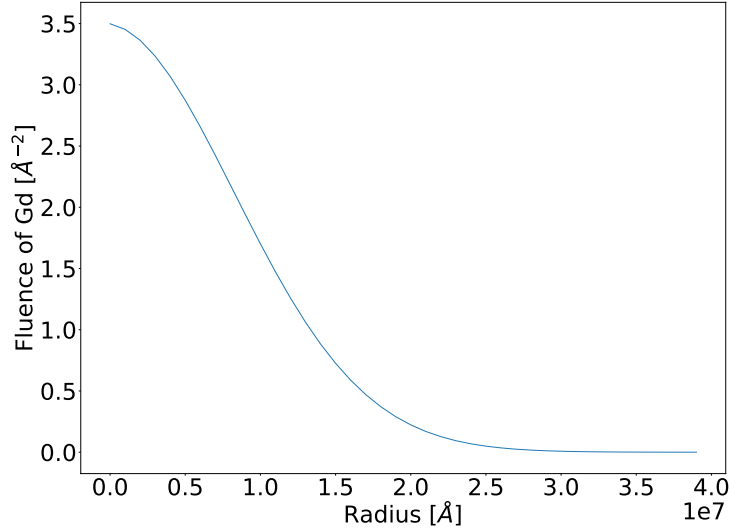
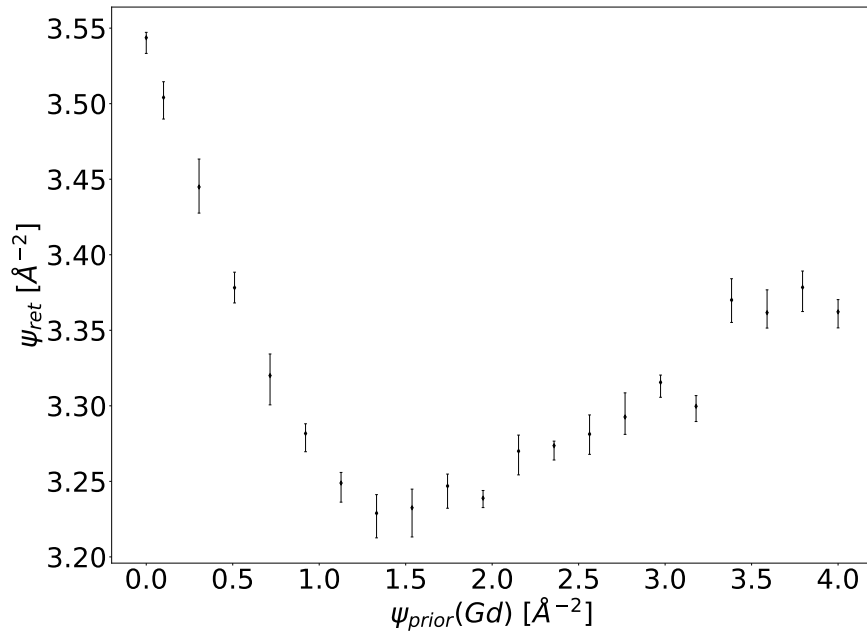


Figure 6.8: Total fluence of implanted gadolinium during the 2021 July collection as a function of the radius of the beam [23].

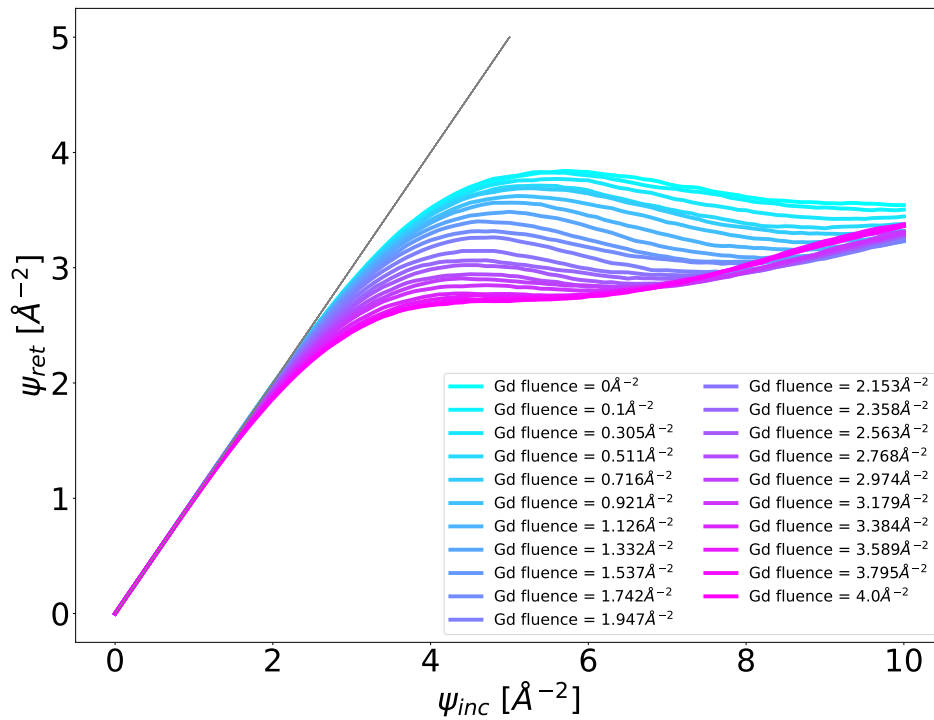
In Figure 6.9a, the retained fluence of terbium is plotted as a function of the fluence of gadolinium that was implanted prior to the terbium implantation. The amount of gadolinium implanted prior to terbium implantation has a significant effect on the retained fluence of terbium. As the amount of gadolinium present in the foil increases, initially, the amount of terbium that is retained decreases, due to the increase of the average Z-value of the foil. However, as the fluence of gadolinium increases more, the sputtering yield of gadolinium for the implantation of terbium increases too, yielding an increase in the rate³ at which gadolinium is removed from the foil. This is also enhanced by the fact that a larger fluence of gadolinium leads to more self-sputtering of gadolinium during the gadolinium implantation. Thus, for larger fluences, the gadolinium is closer to the surface and is thus removed faster once terbium is implanted. Figure 6.9b shows that all curves tend to converge for large incoming fluences: the terbium fluence becomes so large that all the gadolinium is sputtered away and the *history* of the foil is lost.

For curves with large Gd fluences, the retained fluence increases for large incoming Tb fluences. This is due to the fact that for more gadolinium, the gadolinium is removed faster from the foil as a function of the incoming Tb fluences. Therefore, the average Z-value of the foil decreases for increasing Tb fluences and thus the self-sputtering yield of terbium decreases, yielding an increase in the retained fluence. Nevertheless, this increase will never exceed the retained fluence if no gadolinium is present in the foil.

³The rate here is defined as a function of implantation fluence of terbium.



(a) The retained fluence (ψ_{ret}) of terbium for an incoming fluence of $10\text{\AA}^{-2}$ as a function of the gadolinium fluence incoming prior to the terbium implantation ($\psi_{\text{prior}}(\text{Gd})$).



(b) The retained fluence (ψ_{ret}) as a function of the incoming terbium fluence (ψ_{inc}) for different values of the gadolinium fluence implanted prior to the terbium implantation.

Figure 6.9: Retained fluence of terbium as a function of the incoming terbium fluence for different implantation fluences of gadolinium prior to the terbium collection.

Behavior as a function of the total fluence implanted

During the collections, the total current on the implantation foil is continuously measured, therefore the total number of particles impinging on the foil is available at each time. Hence, we want to elaborate on the *total* fluence at which self-sputtering of terbium becomes important, so a more general behavior as a function of the total fluence (the sum of the terbium and gadolinium fluence) will be described. The aim is to obtain an estimate for the total incoming fluence at which self-sputtering of Tb becomes significant, that is independent of the composition of the beam.

Firstly, from the results found above, the fluence at which terbium self-sputtering becomes prominent can be plotted as a function of the fluence of previously implanted gadolinium. If more gadolinium is implanted prior to terbium implantation, the total fluence (Tb+Gd) at which self-sputtering of Tb starts, increases (see Figure 6.10). Thus, as a function of the total current, self-sputtering of Tb for a pure Tb beam occurs earlier than if Gd was implanted prior to the Tb implantation. However, for the latter, less terbium will be retained, as was seen in Figure 6.10. This is mainly an effect of the fact that for larger Gd fluences implanted prior to Tb implantation, Tb is being implanted at a larger *total* incoming fluence and thus the whole self-sputtering process of Tb starts later as a function of the total incoming fluence.

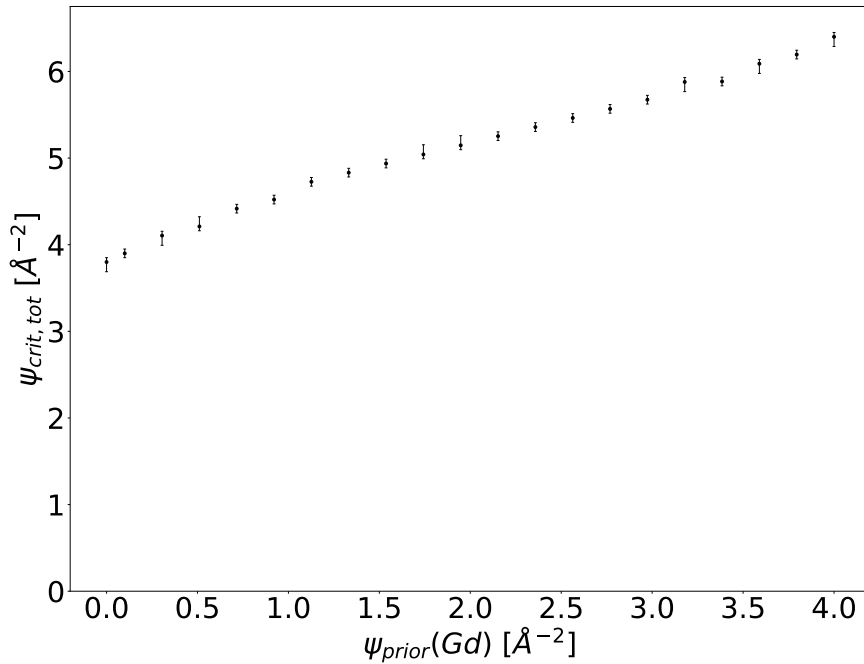


Figure 6.10: The total (Gd + Tb) fluence at which sputtering starts ($\psi_{crit,tot}$) as a function of the fluence of Gd that is implanted prior to the Tb implantation ($\psi_{prior}Gd$).

Secondly, a simulation was performed for terbium and gadolinium implanted simultaneously with different fractions of terbium compared to gadolinium. In Figure 6.11, the total (Tb+Gd) critical fluence is plotted as a function of the fraction of Tb present in the beam. It follows that the total (Tb+Gd) fluence at which self-sputtering of terbium becomes important is rather constant throughout the different fractions of terbium compared to gadolinium. This is mainly due to the similar Z-number of terbium and gadolinium and therefore an impinging Gd ion will have a similar effect as for a Tb ion.

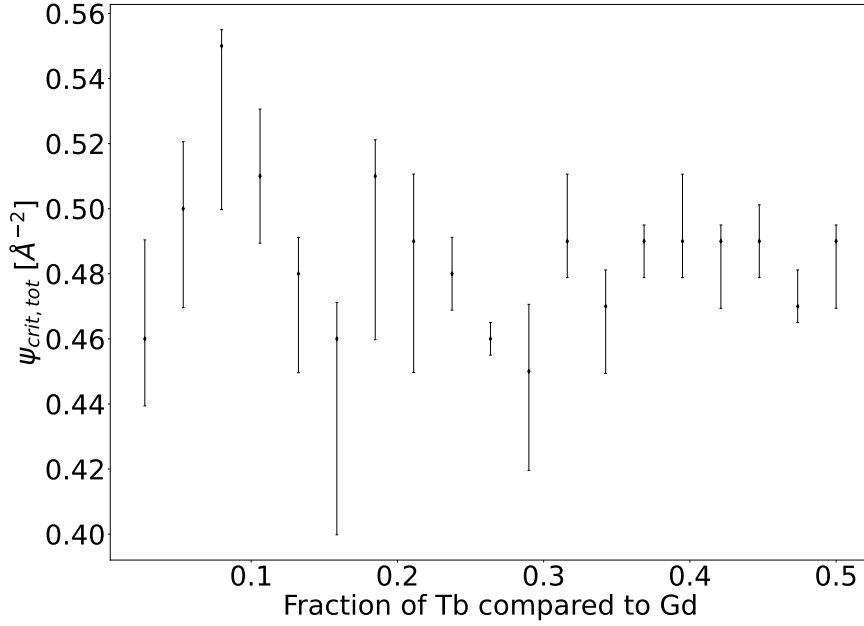


Figure 6.11: The total fluence (Gd fluence + Tb fluence) at which sputtering starts ($\psi_{crit, tot}$) for simultaneous implantation of Tb and Gd as a function of the fraction of Tb compared to Gd.

It is concluded that using the fluence at which self-sputtering of terbium starts for a pure Tb beam as an estimate for the total beam fluence when a mixed beam is impinging on the target is a reasonable approximation. If gadolinium was implanted prior to the terbium beam, self-sputtering will not have significantly started yet, but the pure Tb value can be used as a lower limit for the start of self-sputtering. If gadolinium is implanted simultaneously with terbium, the sputtering of terbium due to incoming terbium or incoming gadolinium is similar and thus self-sputtering of Tb can be described in first approximation as a function of the total fluence.

More specifically, by calculating the critical fluence for a pure terbium beam (ψ_{pure}^{crit}), an estimate for the total fluence (ψ_{tot}^{crit}) at which self-sputtering starts to be important can be found: $\psi_{pure}^{crit} \lesssim \psi_{tot}^{crit}$. The fluence implanted at the center of the beam equals $\frac{N}{2\pi\sigma_r^2}$ (see Appendix B). Here, N is equal to the number of particles implanted on the foil and σ_r is equal to the standard deviation of the beam. Hence, an estimate for the integrated current on the foil at which self-sputtering becomes important at the center of the beam ($c_{int.}^{crit}$) can be calculated from the critical fluence for pure Tb implantation (ψ_{pure}^{crit}):

$$c_{int.}^{crit} = 2\pi e \sigma_r^2 \psi_{pure}^{crit}, \quad (6.1)$$

where e is the electron charge. This provides, given the standard deviation of the beam, an approximate lower limit for the current on the foil at which self-sputtering of terbium will become important given the critical fluence for a pure terbium beam.

This calculation is of great use to know when to replace the foils during a medical isotope collection. In particular, the fractions of Tb compared to its contaminants change continuously throughout a collection, making precise TRIDYN simulations too time-consuming. Moreover, if multiple stable contaminants are present, no distinction between them is possible. Hence, this approximation where only the standard deviation of the beam is needed provides a good measure for when the foils should be replaced.

6.2.3 TbCl and TbF

Recently, investigation is underway at MEDICIS into the use of terbium chloride and terbium fluoride to volatilize Tb in the target due to the reduction in boiling temperature. Fluorine has the advantage of having only one stable mass ($m(\text{F}) = 19$), whereas chlorine has two stable masses (35 and 37). More specifically, for the collection of $^{155}\text{TbCl}$ both mass 190 and 192 can be used. However, for selection on mass 190, $^{153}\text{Tb}^{37}\text{Cl}$ is present as a contaminant whereas for configuration on mass 192, $^{157}\text{Tb}^{35}\text{Cl}$ is present as a contaminant. Hence, enriched ^{155}Gd is needed in order to collect $^{155}\text{TbCl}$, as ^{157}Tb cannot be produced from a ^{155}Gd target irradiated with protons or enriched chlorine is needed.

In this section, self-sputtering for implantation of TbCl, TbCl₂, TbF and TbF₂ in comparison with implantation of elemental Tb ions is explored. The results for implantation in Al are shown in Figure 6.12. Self-sputtering for implantation of TbCl, TbCl₂, TbF and TbF₂ starts respectively at $2.200^{+(25)}_{-(104)} \text{Å}^{-2}$, $1.650^{+(37)}_{-(69)} \text{Å}^{-2}$, $2.750^{+(56)}_{-(56)} \text{Å}^{-2}$ and $2.310^{+(37)}_{-(37)} \text{Å}^{-2}$. It is concluded that, according to the self-sputtering, implantation of terbium fluoride is favored over implantation of terbium chloride and implanting TbX₁ is better than implantation of TbX₂. This is mainly due to the fact that the mass of fluorine is lower than the mass of chlorine, indicating that the incoming chlorine sputters away more of the already implanted Tb than the incoming fluorine. Moreover, terbium obtains less energy upon break-up of terbium chloride than for terbium fluoride, leading to smaller implantation depths. In the same way, terbium coming from the break-up of TbX₁ has a larger implantation depth than for TbX₂. Self-sputtering for implantation of Tb⁺ ions in Al starts to be significant only at $3.72^{+(6)}_{-(14)} \text{Å}^{-2}$ indicating that implantation of a molecular beam is less favorable considering self-sputtering. In conclusion, while collecting molecules could be favorable for extraction of Tb, molecular beam collection causes more self-sputtering compared to elemental Tb collections.

	$\psi_{\text{crit}}(\text{Al})[\text{Å}^{-2}]$
Tb ⁺	$3.72^{+(6)}_{-(14)}$
TbCl	$2.200^{+(25)}_{-(104)}$
TbCl ₂	$1.650^{+(37)}_{-(69)}$
TbF	$2.750^{+(56)}_{-(56)}$
TbF ₂	$2.310^{+(37)}_{-(37)}$

Table 6.2: Summarizing table for the implantation of TbCl, TbCl₂, TbF, TbF₂. The critical fluences for the implantation in aluminum ($\psi_{\text{crit}}(\text{Al})$) of Tb⁺, TbCl, TbCl₂, TbF and TbF₂ are indicated.

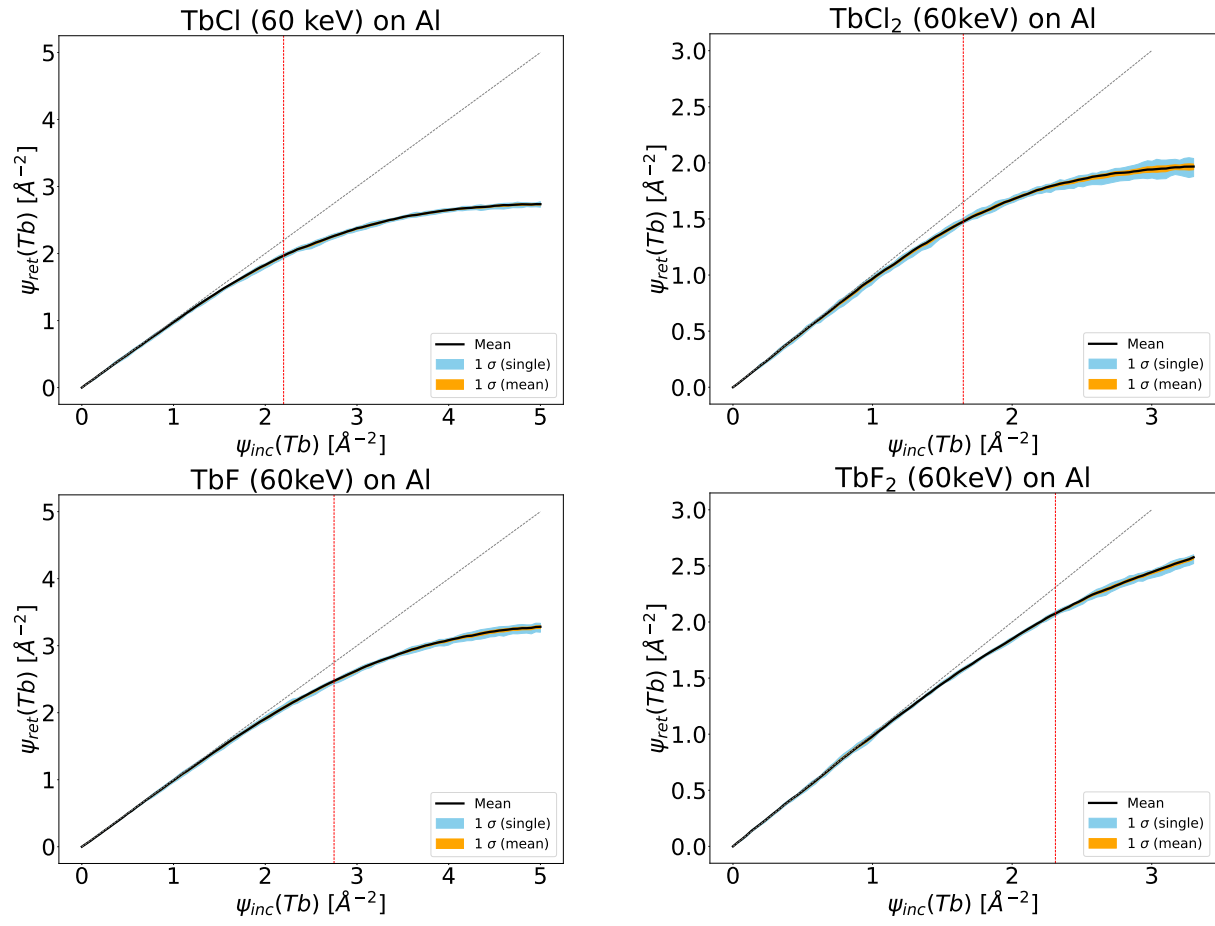


Figure 6.12: Summarizing graphs for the implantation of TbCl, TbCl₂, TbF, TbF₂. The retained fluences (ψ_{ret}) are shown as a function of the incoming fluences (ψ_{inc}) and the red (dashed) lines indicate when self-sputtering starts ($\psi_{retained} < 0.9\psi_{Incoming}$).

7

Experimental verification of the TRIDYN simulations

In this chapter, a protocol is put forward to evaluate the predictive power of the TRIDYN simulations in the lanthanide region. The validity of those simulations in the region of interest for radiopharmaceuticals is planned to be tested by experiments. Yb-implantation will be performed with the High-Current Ion Implanter (HCII) at KU Leuven for similar conditions as those reachable at the different facilities discussed in Section 1.5. After implantation, the foils will be examined using Rutherford Backscattering Spectroscopy (RBS) in order to determine the retained fluence in the foil. A RADIATE [60] proposal for these experiments was submitted in January 2023 (see Appendix C).

First, a short introduction to experimental setup is given and thereafter the protocol is discussed.

7.1 Experimental setup

The first step in the experimental confirmation is the implantation of Yb in zinc and aluminum at 60keV using the high-current ion implanter at KU Leuven. Thereafter, RBS analysis will be performed on the implanted samples in order to determine the Yb-fluence retained in the foil.

7.1.1 High-current ion implanter

For the different implantations, the HCII at KU Leuven will be used [61], which accelerates ions through a potential difference. Subsequently, these ions are mass separated and implanted in a foil. The ions are created from a plasma in the ion source, after which they are extracted with an extraction voltage of up to 40kV. Thereafter, the ions pass through a mass separator. The mass-selected ions are now post-accelerated with a voltage up to 160kV. This results in a maximal acceleration voltage of 200kV, which corresponds, for singly ionized ions, to an energy equal to 200keV. Thereafter, the beam is implanted in the foils, multiple (generally about 20 samples) can be mounted at once.

7.1.2 Rutherford backscattering spectrometry

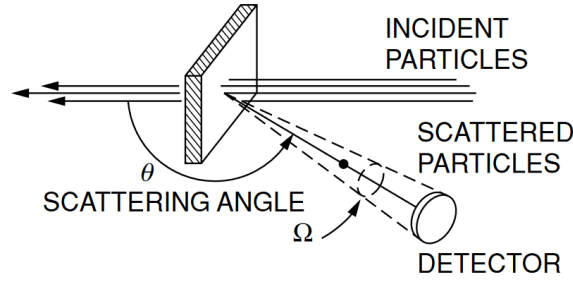


Figure 7.1: Simplified RBS setup. (figure retrieved from [62])

The Rutherford Backscattering Spectrometry (RBS) setup is indicated in Figure 7.1. An α -particle beam is incident on a foil and the backscattered particles are detected at a scattering angle θ . The incident beam of α -particles has an energy of 1 to 2 MeV, indicating that when the α -particles traverse the foil, they lose their energy mainly due to electronic stopping described by the Bethe-Bloch equation [62]:

$$\frac{dE}{dx} = - (e^4) 8\pi \frac{N Z_2}{E} \frac{M_1}{m_e} \ln \left[4 \frac{m_e}{M_1} \frac{E}{I} \right]. \quad (7.1)$$

Here, e is the electron charge, N and Z_2 , respectively, the atomic density and the Z -value of the foil material, E the energy of the α -particles at the interaction point, M_1 the mass of the α -particles, m_e the mass of an electron and $I = 10Z_2$. While traveling through the foil, the α -particles lose energy until they are backscattered. The energy lost by the α -particle due to the backscattering is determined from the kinematics:

$$E_{\text{After backscattering}} = K E_{\text{Incoming}} \quad K = \left[\frac{(M_2^2 - M_1^2 \sin^2 \theta)^{1/2} + M_1 \cos \theta}{M_2 + M_1} \right]^2, \quad (7.2)$$

where K is the kinematic factor, M_1 and M_2 correspond to the mass of the incident particle and foil particle, respectively. Thus, by measuring the number of backscattered particles at each energy, a spectrum is obtained from which the depth and composition of each layer in the target can be determined. The number of backscattered particles is determined by the Rutherford cross section:

$$\sigma(\theta) = \left(\frac{Z_1 Z_2 e^2}{4E} \right)^2 \frac{1}{\sin^4(\theta/2)}. \quad (7.3)$$

Moreover, it can be shown that the number of particles of a certain element is proportional (in first order) to the integral of its corresponding peak in the spectrum [1, 63]. The latter will be used in the next section to determine the fluence that is retained in the foils.

A typical RBS spectrum is given in Figure 7.2, for terbium implanted in aluminum. The spectrum is generated from the output of a TRIDYN simulation, using SIMNRA [64], a simulation package for RBS experiments. This package can be used to simulate RBS spectra, but also to fit experimental spectra in order to obtain the number of particles that are retained in the foil.

Some characteristics of this spectrum will be discussed now. The edge at $\approx 900\text{keV}$ corresponds to α -particles scattered from the surface with an energy $K_{\text{Al}}E_0$, the kinematic factor of aluminum multiplied by the incident energy of the α -particle. For lower energies, the α -particle has penetrated deeper into the foil and has lost some energy due to the electronic stopping prior to and after the backscattering. Moreover, the number of counts increases as the cross section increases for lower energy α -particles (see Equation 7.3). The dip observed in the proximity of the edge is attributed to the reduction in aluminum concentration at the terbium implantation depth. The α -particles scattered from terbium correspond to the peak at $\approx 1400\text{keV}$, which corresponds to the energy of an α -particle that travelled through a layer of aluminum, that was backscattered on a terbium particle and that travelled back through the layer of aluminum.

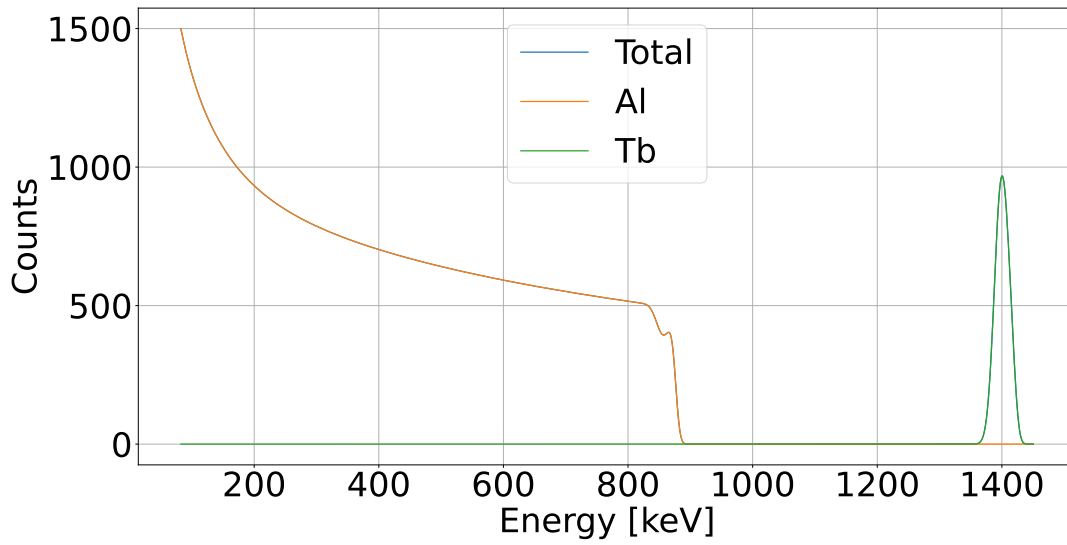


Figure 7.2: Simulated RBS spectrum (counts as a function of the energy of the backscattered α -particles and the corresponding channel number) for terbium implanted in aluminum. The code to generate the RBS spectra was provided by Goele Magchiels.

7.2 Experimental protocol

Once RBS is performed, the fluence of Yb that is retained in the foil can be determined by integrating the spectrum over the energy region of helium ions that are backscattered from Yb (indicated in orange in Figure 7.3). More specifically, for a spectrum where the integral over the peak in the spectrum corresponding to Yb equals I_{RBS} , $\frac{\psi_{\text{cal}}}{I_{\text{cal}}} I_{\text{RBS}}$ equals the fluence of Yb. The values of I_{cal} and ψ_{cal} are obtained from a calibration measurement, where I_{cal} is obtained by integrating a simulated RBS peak for Yb with a fluence of ψ_{cal} , here ψ_{cal} is chosen based on estimates.

Due to the varying number of He ions used in different measurements, renormalization with respect to the current of He ions is performed:

$$\psi_{\text{RBS}} = I_{\text{RBS}} \frac{\psi_{\text{cal}}}{I_{\text{cal}}} \frac{c_T(\text{cal})}{c_T(\text{exp})}, \quad (7.4)$$

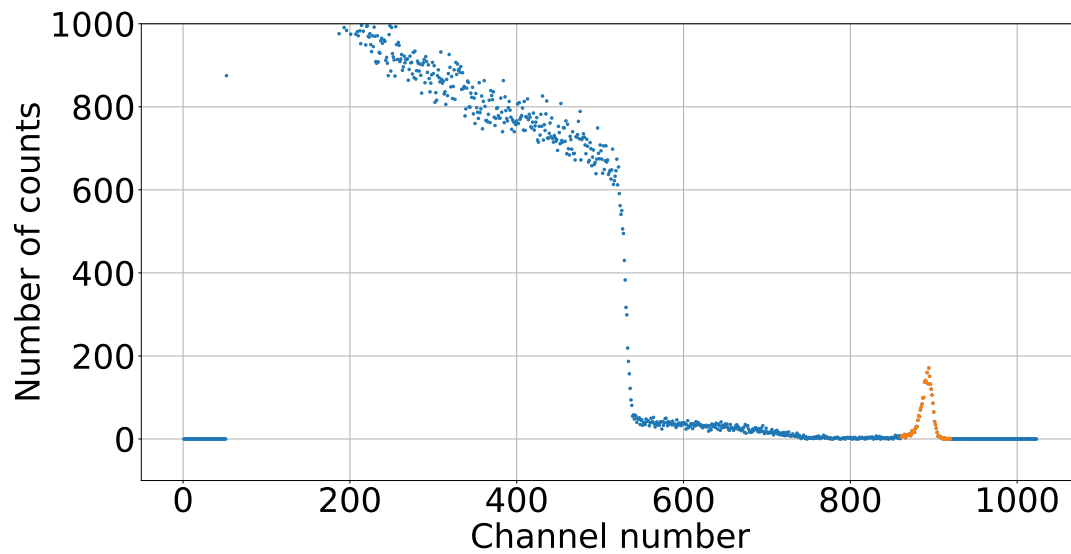
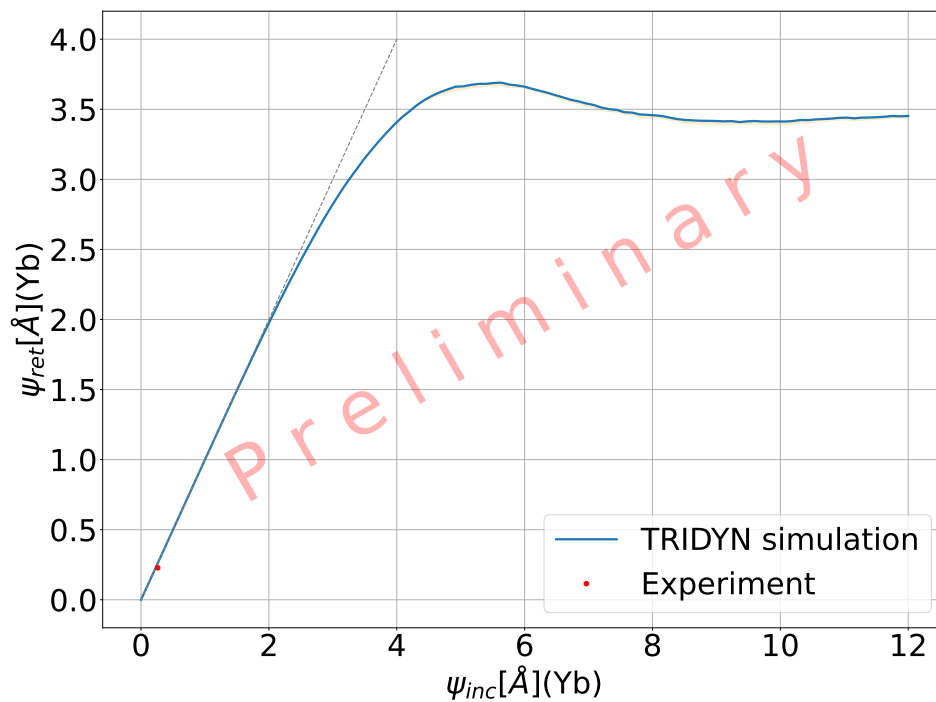


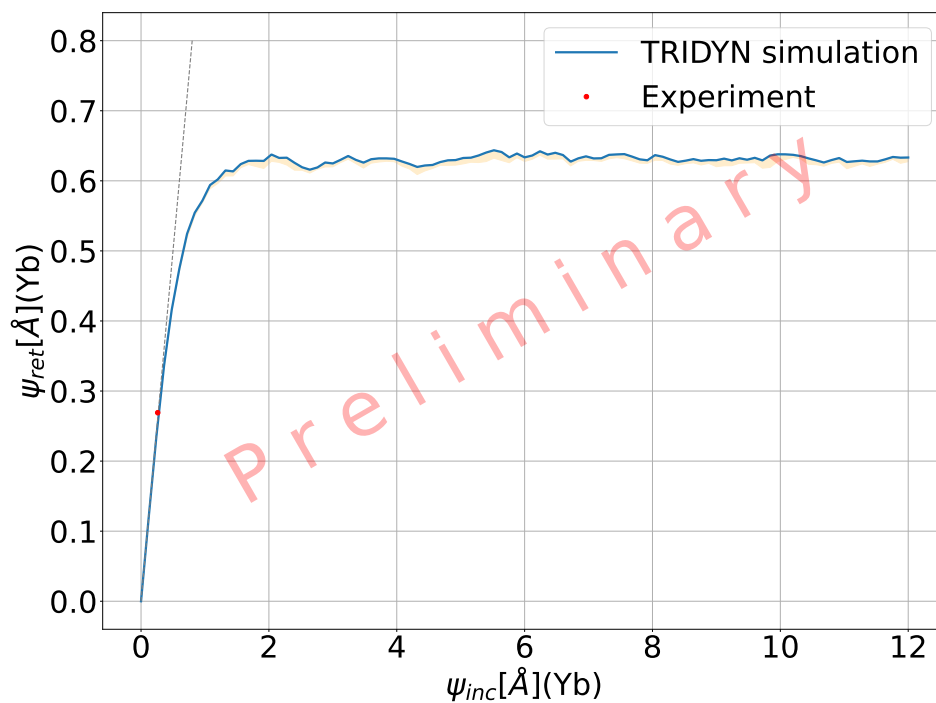
Figure 7.3: RBS spectrum (counts as a function of the channel numbers corresponding to the energy of the backscattered α -particles) for aluminum implanted with ytterbium. The peak corresponding to Yb is indicated in orange.

Here, $c_T(\text{cal})$ and $c_T(\text{exp})$ correspond to the currents of He ions incoming for the calibration measurement and the considered measurement, respectively.

Due to time-restrictions of the high-current ion implanter at KU Leuven, only a limited number of implantations could be performed within the timeline of this thesis. The preliminary results of these experiments are shown in Figure 7.4. Further analysis and implantation at higher fluences are required to provide an evaluation of the TRIDYN simulations in the ytterbium region. Nevertheless, evidence for the feasibility of those experiments to be conducted in the future is now obtained.



(a) Implantation in Al.



(b) Implantation in Zn.

Figure 7.4: Experimental results (red points) of the Yb implantations compared to the TRIDYN simulations (blue curve). The retained fluence is shown as a function of the incoming fluence.

8

TRIDYN simulations for the collection of July 2021

In this chapter, the aim is to verify the statement that was made in [23] that self-sputtering occurred during the collection of ^{155}Tb in July 2021. First, the collection is summarized. Thereafter, a simulation is performed in which the retained activity is calculated. Also, a layered simulation is performed in order to take into account the changing ratios of particles in the beam.

8.1 Collection information

For the collection of July 2021, a tantalum target was irradiated at ISOLDE using 1.4 GeV protons to induce spallation. For the implantation in the *second* foil during the July collection, which was the most successful collection of 2021, the activity spectra were acquired 4 times during the collection and the activities were calculated from the Kromek spectra as is described in Appendix A. The activities are recalculated (taking into account the systematic error) compared to [23] (see Table 8.1) and the values mostly agreed within 1σ . The total collected activity at each point can be calculated as:

$$A_{\text{collected}}(t + \Delta t) = A_{\text{Kromek}}(t + \Delta t) - A_{\text{Kromek}}(t) \exp(-\lambda \Delta t) \quad (8.1)$$

$$A_{\text{collected}}^{\text{total}} = \sum_{i=1}^4 A_{\text{collected}}(i), \quad (8.2)$$

where $A_{\text{Kromek}}(i)$ is the activity measured with the Kromek detector for the i th measurement, λ the decay constant and Δt the time between two measurements (see Appendix A for the uncertainty on this activity and the extension towards isotopes with multiple peaks in the γ -spectrum). $A_{\text{collected}}^{\text{total}}$ is the activity that impinged on the foil during the collection, and can thus be used to calculate the number of particles that are incoming. Note that this activity will differ from the final Kromek measurement. The results are summarized in Table 8.1, from which it can be concluded that the total activity of terbium collected on the *second* foil is equal to 5.41(25) MBq. Contrastingly, the activity on the foil after extraction from the detection chamber is 2.686(87) MBq (value partially corrected for the decay over time).

Timestamp	¹⁵⁵ Tb activity [MBq]		¹³⁹ Ce activity [MBq]	
	Using A	From [23]	Using A	From [23]
-1	1.59(27)	1.56(31)	0.1611(89)	0.1542(81)
0	5.61(45)	5.93(66)	0.501(21)	0.552(30)
1	8.57(69)	8.54(114)	0.842(33)	0.834(43)
2	9.12(63)	9.10(124)	0.961(23)	0.894(45)
3	8.60(59)	8.53(118)	1.054(25)	1.017(52)
4	7.94(58)	8.28(114)	1.002(27)	0.953(48)

Table 8.1: Summary of the activity during the July 2021 collection corresponding to ¹⁵⁵Tb and ¹³⁹Ce. The zeroth time step indicates the change of the foil.

On the other hand, for the *first* foil that was used in the collection of July 2021, an activity of 3.745(125)MBq was impinging and the activity measured on the foil after extraction is 3.439(121)MBq (corrected for the decay). Thus, compared to the second foil, less activity was impinging, but more was retained. This is mostly an effect of the self-sputtering: for this smaller incoming fluence, self-sputtering limits are not reached yet and thus more terbium is retained.

From the γ -spectra acquired with the Kromek CZT detector it was observed that ¹³⁹CeO is present as an isobaric contaminant. The activity of ¹³⁹Ce measured in the collection chamber during implantation in the second foil is 0.5539(176)MBq. Moreover, stable gadolinium is present as a contaminant.

The number of particles corresponding to terbium and cerium is now calculated:

$$N = \frac{A}{\lambda}, \quad (8.3)$$

where N is the number of particles, A is the activity and λ the decay constant. By doing so, $3.606 \cdot 10^{12}$ particles for ¹⁵⁵Tb and $9.5 \cdot 10^{12}$ particles corresponding to ¹³⁹Ce are found. As the stoichiometry of cerium and oxygen is one to one, the number of oxygen particles is equal to that of cerium. The total number of particles impinging on the foil, calculated from the total ion current, is larger than the number of particles corresponding to terbium, cerium and oxygen (calculated from the activities). This excess of incoming particles is most likely caused by stable gadolinium (¹⁵⁵Gd). Other stable contaminants such as LaO will be present but those are not considered in this simulation.

The fluence is defined as the number of particles per surface area. Thus, by assuming that the beam has a Gaussian distribution, the fluence implanted in an infinitesimal ring defined by the radii r and $r + \Delta r$ ($R(r, r + \Delta r)$) is given by (see [Appendix B](#)):

$$\psi_{R((r,r+\Delta r))} = \frac{N}{\pi((r + \Delta r)^2 - r^2)} \left(\exp\left(-\frac{r^2}{2\sigma_r^2}\right) - \exp\left(-\frac{(r + \Delta r)^2}{2\sigma_r^2}\right) \right), \quad (8.4)$$

with N the total number of particles and $\sigma_r = \sqrt{\sigma_x \sigma_y}$, the standard deviation of the Gaussian beam profile. The fluence for each of the incoming particles can now be calculated from the total number of particles of that element.

At MEDICIS, the target is placed on 60kV, thus the singly charged ions have an energy equal to 60keV. For ^{139}CeO , this energy is distributed according to conservation of momentum and energy, yielding $E_{\text{Ce}} = 53.8\text{keV}$ and $E_{\text{O}} = 6.2\text{keV}$.

8.2 Simulation

The incoming fluence of each element and the corresponding energies are known and thus the simulation for the collection of 2021 can be performed. The results are shown in Figure 8.1. Ten different random number seeds were used to obtain a measure for the statistical error on the simulation, both the error on one simulation and the error on the mean value for 10 simulations are provided. From this curve, the retained activity for this collection, taking into account the implantation profile, can be calculated according to the following procedure:

1. Make a polynomial fit of fourth degree for the retained fluence graph (as is shown in Figure 8.1). No constant term is used in the polynomial in order to impose conservation of particles, i.e., zero retained fluence if no fluence is incoming.
2. Calculate the incoming fluence (ψ_{in}) in $R(r, r + \Delta r)$ by using Equation 8.4.
3. By using the polynomial fit (determined in step 1), the retained fluence (ψ_{ret}) for an incoming fluence equal to ψ_{in} can be calculated (see e.g., Figure 8.2).
4. From the retained fluence (ψ_{ret}), the activity corresponding to an incoming beam between radius r and $r + \Delta r$ can be calculated:

$$A = \psi_{\text{ret}} \pi ((r + \Delta r)^2 - r^2) \lambda_{^{155}\text{Tb}}$$
5. Start this scheme for $r = 0$ and repeat for $r = r_{\text{previous}} + \Delta r$ until convergence is obtained.
6. The sum of all the partial activities is equal to the total activity.

By using this scheme, an activity of 2.5956(147)MBq is found. In [23], a measurement of the activity after the collection was performed and yielded 2.686(89)MBq. Thus, very good agreement is found, indicating that the simulation software is suited for the self-sputtering problem.

Moreover, in Figure 8.2, where the retained fluence is shown as a function of the incoming fluence, a Gaussian curve is expected if no self-sputtering had occurred. For large radii, indeed, the Gaussian behavior is observed, up to the point where self-sputtering becomes important. At this point (around 1.2mm), for a slightly smaller radius, more particles are self-sputtered and thus less is retained in the foil than for slightly higher radii. Upon decreasing the radius further an oscillation is observed due to the fact that a slight oscillation is present in the self-sputtering graph (see Figure 8.1).

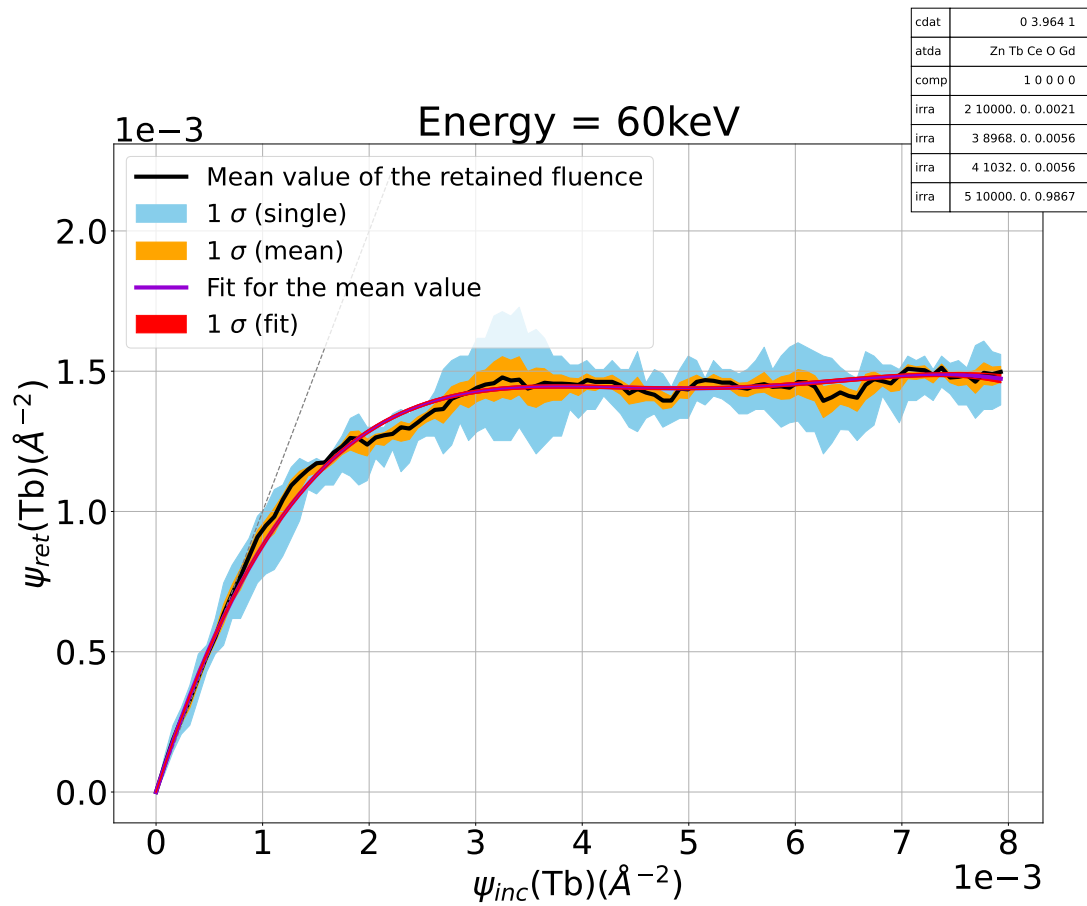


Figure 8.1: Summarizing graph on the retained fluence of Tb for the collection of 2021 where the retained fluence is shown as a function of the incoming fluence of terbium. Together with the fits that were used for the activity calculation.

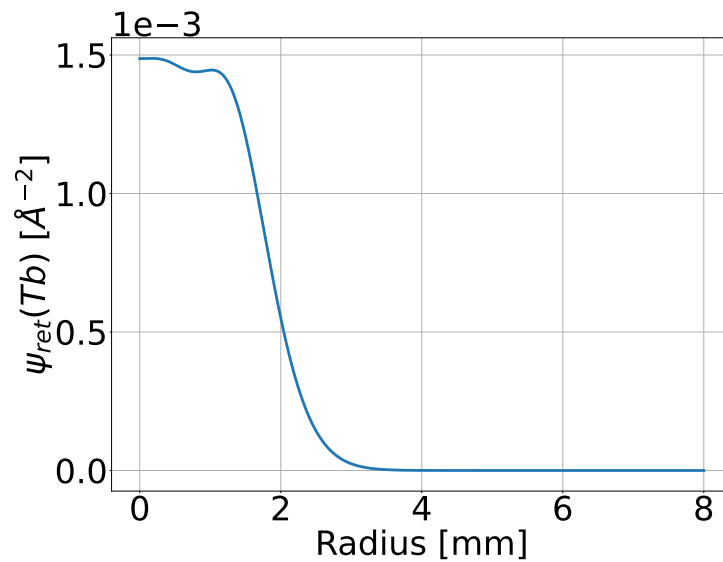


Figure 8.2: The retained fluence as a function of the position in the beam (0 mm corresponds to the center of the beam).

Lastly, a summarizing figure is presented, which will be useful when layered simulations are considered. In Figure 8.3, the x-axis represents the radius (center of the beam has radius 0), on the y-axis the incoming fluence corresponding to this radius is indicated, the retained fluence is represented by the colorbar. Note that the incoming fluence is Gaussian distributed, thus equal timelines follow a Gaussian relation between the incoming fluence and the radius. For the same timeline, the fluence impinging on the spot corresponding to the center of the beam is larger than for a non-zero radius.

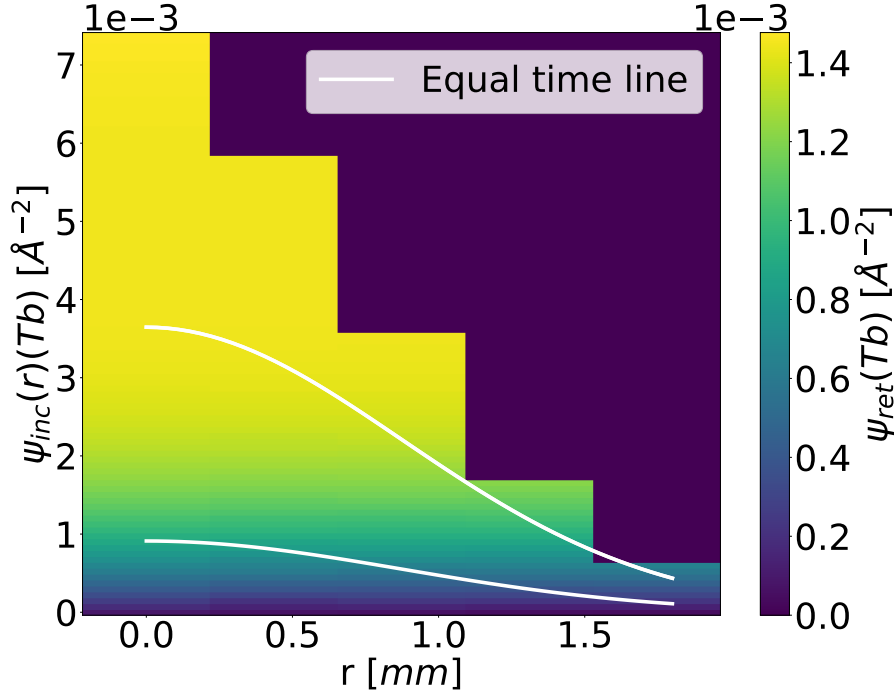


Figure 8.3: Heatmap for the 2021 July collection for an energy of 60keV. The x-axis represents the radius (r), where 0 corresponds to the center of the beam, on the y-axis the corresponding incoming fluence (ψ_{inc}) is given, note that for larger radii the incoming fluence at the same time (same time line indicated in white) is smaller as the beam profile is Gaussian. The colorbar indicates the retained fluence (ψ_{ret}) for a specific radius and incoming fluence.

As the incoming fluence increases, the retained fluence increases up to a point where self-sputtering becomes significant. At this point, terbium atoms are removed from the foil, leading to a flattening behavior in the amount of retained terbium. At slightly larger radii, the incoming fluence is smaller. Due to this lower incoming fluence, no self-sputtering has occurred yet and more terbium is retained.

In this case, symmetry for the radius is present, i.e., symmetry of the retained fluence in the horizontal direction of the graph. However, throughout the collection, the beam composition changes continuously, leading to changing fractions of terbium compared to gadolinium and cerium. Consequently, the self-sputtering behavior will depend on the radius: initially, a beam with a specific fraction of terbium is implanted. During this implantation, larger radii are exposed to lower fluences, which results in a smaller amount of terbium being present when switching to another fraction of terbium. Hence, different results for different radii are expected. Therefore, further investigation into this time-dependent behavior is performed.

8.2.1 Layered simulation

Up to this point, all particles were implanted with relative fractions that remain constant throughout the simulation. However, these fractions are time-dependent for actual collections. Therefore, a layered simulation is performed, in which different simulations are performed in subsequent steps. In the first step, Tb, Ce, O and Gd are implanted with specific fractions (calculated from the first activity measurement and the total current on the foil). The total incoming fluence that is implanted in this first step is calculated from the integrated total current on the foil. After this first simulation, a subsequent simulation is performed. Again the fractions of Tb, Ce, O and Gd and the total incoming fluence are calculated from the (subsequent) activity measurement and total current on the foil. The novelty of TRIDYN is that for this second simulation, the depth profile of the previous simulation can be used as a starting foil composition. During the collection of July 2021 (with the second foil), four γ -spectra were acquired with the Kromek detector and thus four subsequent simulations will be performed. Using these activity spectra, the activity for terbium and cerium that was collected during a given time interval Δt ($A_{\text{collected}}$) can be calculated as is described in Appendix A. Hence, the total number of particles of terbium, cerium and oxygen that are incoming on the foil can be calculated by the use of Equation 8.3. The results are summarized in Table 8.2.

		^{155}Tb	^{139}Ce	^{16}O	^{155}Gd
Activity collected (MBq)	1	3.045(182)	0.3416(143)		
	2	1.114(146)	0.12019(179)		
	3	1.249(73)	0.09216(145)		
	4	0	0		
Number of particles collected	1	$2.03 \cdot 10^{12}$	$5.86 \cdot 10^{12}$	$5.86 \cdot 10^{12}$	$2.53 \cdot 10^{14}$
	2	$7.43 \cdot 10^{11}$	$2.06 \cdot 10^{12}$	$2.06 \cdot 10^{12}$	$3.17 \cdot 10^{14}$
	3	$8.33 \cdot 10^{11}$	$1.58 \cdot 10^{12}$	$1.58 \cdot 10^{12}$	$9.59 \cdot 10^{14}$
	4	0	0	0	$1.50 \cdot 10^{14}$

Table 8.2: Summary of the collected number of particles during the collection of July 2021 for the different time intervals (1-4). Since oxygen and gadolinium are stable, no activities are given. The uncertainties that are indicated take into account the systematic error due to the uncertainty on the efficiency as well as the statistical uncertainty as is described in Appendix A.

The retained activity can now be calculated similar to the scheme given in Section 8.2. Multiple layered simulations were performed for different radii to take into account that upon going to the next step in the simulation (changing the beam composition), different fluences were implanted at different radii. More specifically, the retained fluence can be calculated using the following procedure:

1. Calculate the incoming fluence in $R(r, r + \Delta r)$ by using Equation 8.4 for each of the four layers in Table 8.2.
2. Perform the layered simulation with the incoming fluences calculated in the previous step, using TRIDYN. Here, the first simulation starts from a pure Zn foil and the subsequent simulations use the output depth profile of the previous layer as an input

depth profile (see Table 8.3 for the basic simulation input values). In doing so, the retained fluence (and thus the retained activity) in $R(r, r + \Delta r)$ is obtained.

3. Start this scheme for $r = 0$ and repeat for $r = r_{\text{previous}} + \Delta r$ until convergence is obtained.
4. The sum of all the partial activities is equal to the total activity.

This scheme is implemented by combining Python with a bash script to automatize the process. In this way, for every radius, the retained fluence for a given incoming fluence (Gaussian distributed) can be obtained as is visualized in Figure 8.4. From this figure, it is deduced that the incoming fluence for which the maximal retained fluence is obtained decreases with increasing radius. Also, for the last equal time line, the retained fluence at a radius of about 1.5Å is larger than for radius 0. From these simulations, a total retained activity equal to 1.7919MBq is found, which is significantly lower than what was found experimentally and for the non-layered simulation. This discrepancy could be an effect of the fact that only five measurements with the Kromek detector are performed, while there is a lot of fluctuation in the activity that is collected in the foil. Additionally, in the simulation, it was assumed that all excess in the total current on the foil compared to the activity spectra was originating from stable ^{155}Gd . However, other stable contaminants such as LaO can also contribute to this excess. Due to the lower mass and Z-value of LaO, less self-sputtering will occur, which could possibly explain why self-sputtering is overestimated in the simulation.

	Layer 0	Layer 1	Layer 2	Layer 3
atda	Zn Tb Ce O Gd	Zn Tb Ce O Gd	Zn Tb Ce O Gd	Zn Tb Ce O Gd
comp	1 0 0 0 0			
irra	2 60000. 0. 0.00761	2 60000. 0. 0.00231	2 60000. 0. 0.00087	2 60000. 0. 0
irra	3 53806. 0. 0.02195	3 53806. 0. 0.00642	3 53806. 0. 0.00164	3 53806. 0. 0
irra	4 6194. 0. 0.02195	4 6194. 0. 0.00642	4 6194. 0. 0.00164	4 6194. 0. 0
irra	5 60000. 0. 0.94849	5 60000. 0. 0.98486	5 60000. 0. 0.99585	5 60000. 0. 1

Table 8.3: Table with the most important input values for the layered simulation for the 2021 July collection.

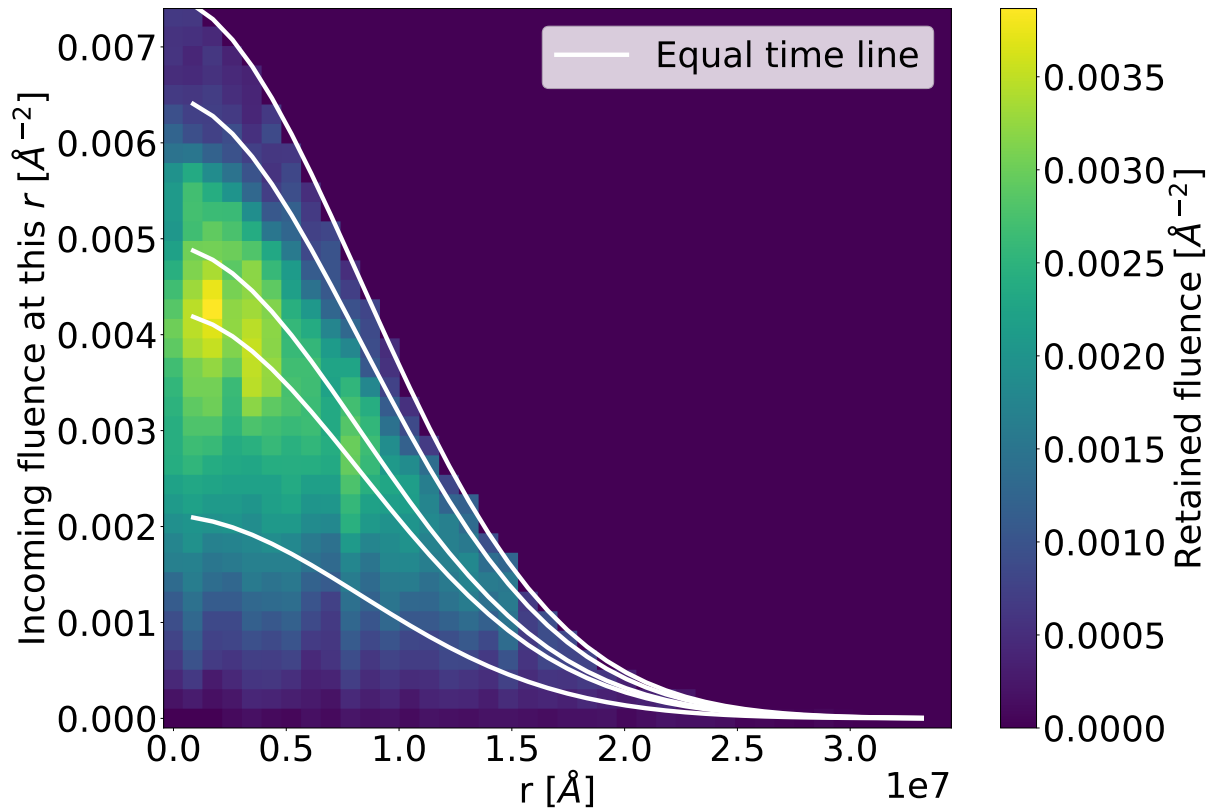


Figure 8.4: Heatmap for the 2021 July collection for the layered simulation. The x-axis represents the radius (r), where 0 corresponds to the center of the beam, on the y-axis the corresponding incoming fluence (ψ_{inc}) is given, note that for larger radii the incoming fluence at the same time (same time line indicated in white) is smaller as the beam profile is Gaussian. The colorbar indicates the retained fluence (ψ_{ret}) for this radius and this incoming fluence.

9

Self-sputtering - Conclusion and outlook towards future collections and facilities

For increasing collection fluences, self-sputtering becomes more important. In this chapter, different methods are proposed that could help to limit the self-sputtering as much as possible.

First of all, implanting mainly in Al rather than in Zn during MEDICIS collections was a good decision, as self-sputtering in aluminum only starts at 7 times larger incoming fluences compared to zinc. As implantation in Al and NaNO_3 are comparable, the choice between both depends on the desired chemical post-processing (aluminum is dissolved in acid whereas NaNO_3 is dissolved in water). Nevertheless, implantation in sugar is even more promising, with a 3-fold increase in the fluence at which self-sputtering starts for cellulose compared to aluminum. However, it has to be noted that the simulations for self-sputtering in sugar do not take into account the microstructure of sugar, which could deteriorate the self-sputtering behavior or its retention capabilities. The latter should, therefore, be experimentally verified.

The main conclusion for the energy of the radionuclide beam is to take an energy that is as high as reasonably achievable. To date, the extraction voltage at the different facilities is limited by $\approx 60\text{kV}$, mainly due to the reduced stability and the available transformers. The behavior as a function of the energy of the beam needs to be taken into account when building new facilities, as the incoming fluence at which self-sputtering starts for cellulose is approximately trice as much for implantation at 200keV compared to 30keV . It could, for example, be promising to consider raising the collection chamber to a negative voltage. By doing so, the energy of the impinging particles increases without the instability and transformer availability issues.

It has to be noted that for collections where the target was irradiated with high-energy protons in order to induce spallation reactions, more contaminants are present, such as ^{139}CeO [23]. Those isotopes have a larger mass than the aluminum foil, leading to similar sputtering effects as for gadolinium. Furthermore, minimizing the time interval between the chemical separation of ^{155}Tb and ^{155}Gd and the start of the collection is of great importance to reduce the presence of ^{155}Gd resulting from the decay process of ^{155}Tb . In this way, a purer beam of ^{155}Tb will be implanted, leading to less self-sputtering. Moreover, for online collections, no chemical preparation can be performed to remove

contaminants. Consequently, collections with a target that has been chemically purified will result in less self-sputtering. In particular, if the beam purely consists of terbium, self-sputtering occurs much later as a function of the Tb fluence, as everything that is implanted with a mass $\approx 155u$ contributes similarly to the self-sputtering. More specifically, for the conditions corresponding to the collection of 2021, self-sputtering starts at an incoming fluence of terbium of $0.001\text{\AA}^{-2}$, whereas if implantation had been performed with a pure terbium beam, self-sputtering had been only important at an incoming fluence of $0.48\text{\AA}^{-2}$. Hence, it is necessary to explore methods to enhance the purity of the Tb beam.

Moreover, it was shown that the total incoming fluence impinging on the foil should be reduced, e.g., by broadening the implantation spot. The latter can be done by broadening the beam before implantation, nevertheless, this diminishes the quality of the beam. Alternatively, wobbling the beam in the directions perpendicular to the beam can be implemented to increase the implantation spot. Since the fluence is defined as the number of particles per area, increasing the area 4 times implies that 4 times more particles are collected at the moment that self-sputtering becomes significant.

In addition, it is crucial to systematically replace the collection foil, which is likely the least drastic measure to implement. We found that the fluence at which self-sputtering starts for a pure terbium beam can be used to obtain an estimate/lower bound for the total current if the beam consists also of gadolinium or other contaminants: $c_{\text{int.}}^{\text{crit}} = 2\pi e\psi_{\text{pure}}^{\text{crit}}\sigma_r^2$. So, given the standard deviation of the beam, the integrated current at which self-sputtering becomes important (at the center of the beam) can be calculated from the critical fluence for the implantation of a pure beam. Hence, a summarizing table with the critical fluence for implantation of a pure beam of different medical isotopes is given in Table 9.1. It is concluded that self-sputtering occurs earlier for heavier isotopes. Actinides do not follow the trend, most likely because the lattice binding energies are not very well known (as no macroscopic amount exists). Moreover, experimental confirmation of TRIDYN in the actinide region is not feasible, as all actinides are radioactive. Additionally, in Appendix D, the corresponding self-sputtering curves are shown.

Considering the importance of changing the foils, the target changing system implemented in ISOLPHARM at SPES (see Section 1.5) could be very promising.

Lastly, further investigation into self-sputtering at MEDICIS is considered by investigating placing an aluminum foil with a hole on the back of the collimator to try to collect the self-sputtered particles.

ψ^{crit} [Å ⁻²]	Metal				Salt				Sugar			
	Al		Zn		NaCl		NaNO ₃		C ₅ H ₅ O ₆		C ₆ H ₁₀ O ₅	
	30keV	60keV	30keV	60keV	30keV	60keV	30keV	60keV	30keV	60keV	30keV	60keV
Sc	4.32 ⁺⁽¹³⁾ ₋₍₁₃₎	10.080 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	0.600 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	1.440 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	2.400 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	5.040 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	4.080 ⁺⁽¹³⁰⁾ ₋₍₆₀₎	8.640 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	11.280 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	12.000 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	12.000 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	12.000 ⁺⁽⁶⁰⁾ ₋₍₆₀₎
Cu	3.000 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	5.520 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	0.360 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	0.840 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	1.680 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	3.000 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	2.760 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	5.160 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	7.560 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	12.000 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	9.240 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	12.000 ⁺⁽⁶⁰⁾ ₋₍₆₀₎
Sm	2.040 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	2.88 ⁺⁽¹³⁾ ₋₍₁₃₎	0.240 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	0.480 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	1.080 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	1.560 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	2.160 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	3.000 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	5.160 ⁺⁽¹³⁰⁾ ₋₍₆₀₎	7.440 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	6.600 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	9.480 ⁺⁽⁶⁰⁾ ₋₍₆₀₎
Tb	2.160 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	3.120 ⁺⁽¹³⁰⁾ ₋₍₆₀₎	0.360 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	0.480 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	1.200 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	1.680 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	2.160 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	3.000 ⁺⁽¹³⁰⁾ ₋₍₆₀₎	5.760 ⁺⁽¹³⁰⁾ ₋₍₆₀₎	8.280 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	7.440 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	10.320 ⁺⁽⁶⁰⁾ ₋₍₆₀₎
Tm	2.040 ⁺⁽¹³⁰⁾ ₋₍₆₀₎	2.880 ⁺⁽¹³⁰⁾ ₋₍₆₀₎	0.240 ⁺⁽¹³⁰⁾ ₋₍₆₀₎	0.480 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	1.080 ⁺⁽¹³⁰⁾ ₋₍₆₀₎	1.560 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	2.160 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	3.000 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	5.400 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	7.560 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	6.720 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	9.480 ⁺⁽⁶⁰⁾ ₋₍₆₀₎
Ac	2.040 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	2.640 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	0.240 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	0.360 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	1.200 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	1.440 ⁺⁽¹³⁰⁾ ₋₍₆₀₎	2.280 ⁺⁽¹³⁰⁾ ₋₍₆₀₎	3.120 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	7.800 ⁺⁽¹³⁰⁾ ₋₍₆₀₎	10.200 ⁺⁽⁶⁰⁾ ₋₍₁₃₀₎	10.200 ⁺⁽⁶⁰⁾ ₋₍₆₀₎	12.000 ⁺⁽⁶⁰⁾ ₋₍₆₀₎

Table 9.1: Fluence at which self-sputtering starts to be important (ψ^{crit}) and the foil should be changed for the isotopes of interest for medical applications. $c_{int.}^{crit} = 2\pi e\psi^{crit}\sigma_r^2$ can be used to determine the corresponding critical integrated current, i.e., self-sputtering will start at the center of the beam.

PART III:

Recoil sputtering for alpha emitters

‘If you knew Time as well as I
do,’ said the Hatter, ‘you
wouldn’t talk about wasting it.’

Lewis Carroll

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10

Recoil sputtering

In this chapter, the sputtering subsequently to the implantation due to the recoiling daughter nucleus originating from an α -decay will be investigated. Both TRIDYN and SRIM will be combined to get a first estimate on the significance of recoil-sputtering in the context of medical isotope collections. Moreover, the dependence on the energy of the incoming beam and the implantation spot will be examined. Throughout this chapter, ^{149}Tb will be used as the incoming radionuclide, but the simulations can be performed for any α -decaying radionuclide.

10.1 Context

^{149}Tb is an interesting medical isotope as it has an α -decay branch towards ^{145}Eu (branching ratio = 17%, see Figure 10.1). However, the half-life is rather small (4.1h), therefore the time between the collection of ^{149}Tb and the therapy must be minimized in order to have a sufficient number of energetic α -particles remaining to treat the patient.

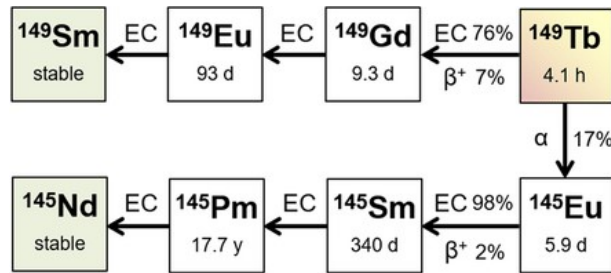


Figure 10.1: Decay scheme of ^{149}Tb . (figure retrieved from [65])

For α -decaying isotopes, apart from the decay-induced decrease in the activity, one of the additional phenomena that needs to be taken into account is *recoil-sputtering*. If a mother nucleus decays via α -decay, the daughter nucleus obtains a significant recoil energy ($\approx 0.1 - 1\text{MeV}$). Hence, the daughter nucleus traverses the foil and transfers its energy to surrounding particles, creating nuclear collision cascades. Consequently, earlier implanted ^{149}Tb particles or foil particles can be sputtered away (see Figure 10.2). The

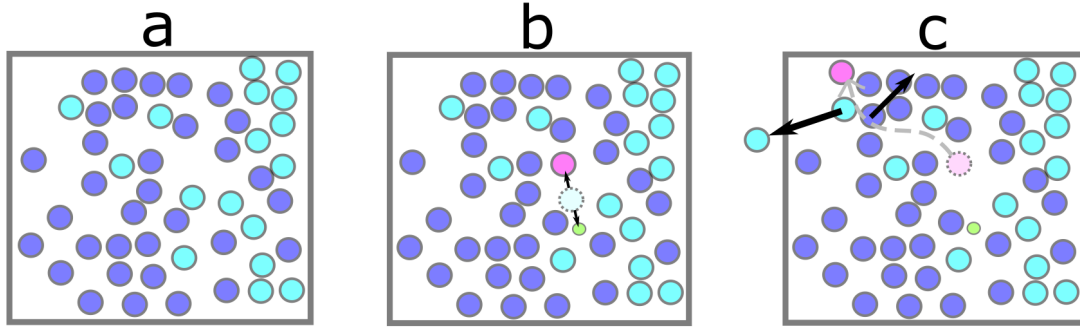


Figure 10.2: A simplified visualization of the recoil-sputtering process: **a)** starting from a foil implanted with α -decaying nuclei (light blue), **b)** a mother nucleus can decay into the daughter nucleus (pink) and an α -particle (green). **c)** The daughter nucleus obtains a recoil-momentum and traverses the foil while colliding with foil particles. Consequently, a nuclear collision cascade emerges and if a particle near the surface obtains a momentum in the outward direction, it can be ejected from the foil.

energy of the ^{145}Eu nucleus can be calculated according to Equation 1.5:

$$E(^{145}\text{Eu}) = \frac{Q}{1 + \frac{m_{^{145}\text{Eu}}}{m_{\alpha}}} = \frac{4.0775\text{MeV}}{1 + \frac{145}{4}} = 0.10946\text{MeV}. \quad (10.1)$$

In conclusion, increasing the time between collection and treatment time results in a decrease of the amount of terbium present in the foil due to decay, but also due to sputtering induced by recoiling daughter nuclei, i.e., recoil-sputtering. The aim of this chapter is to obtain a measure for the importance of this process and its dependence on the implantation energy and implantation spot. In order to maximize the reliability, both SRIM [57] and TRIDYN [53, 54, 55] are used. A typical view of the SRIM interface is shown in Figure 10.3.

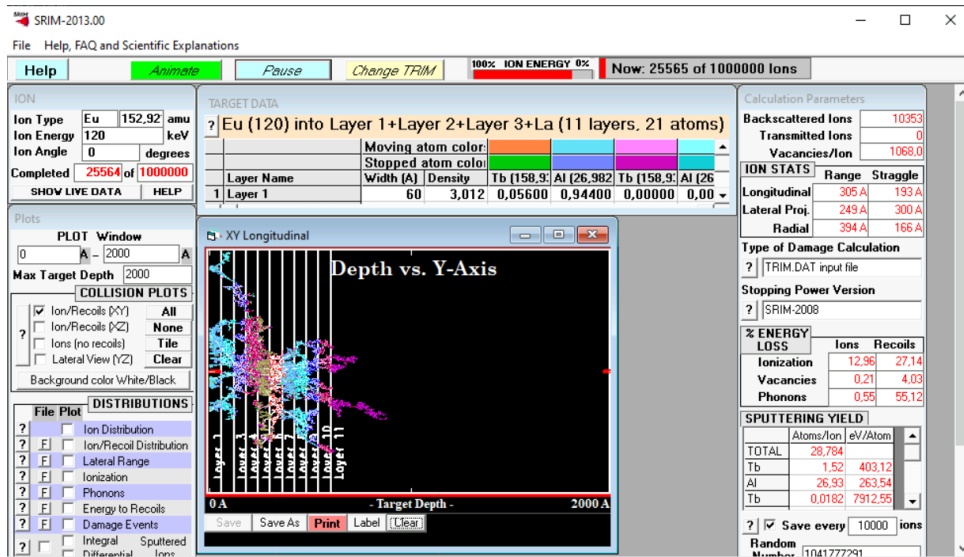


Figure 10.3: Interface of SRIM. During the simulation, the recoiling ions can be tracked on several graphs, for example the XY projection. Moreover, the sputtering yield is indicated during the simulation for each of the different layers of the target.

10.2 Simulation

In order to simulate recoil-sputtering, first, a simulation for terbium implantation is performed using TRIDYN (energy of the terbium beam = 60keV). Simulations for different fluences (around the value where self-sputtering just starts to have an effect: $1.5\text{\AA}^{-2}$, $2.01\text{\AA}^{-2}$, $2.49\text{\AA}^{-2}$, $3.48\text{\AA}^{-2}$ and $4\text{\AA}^{-2}$) are performed for implantation in aluminum. From these TRIDYN simulations, an implantation profile is obtained as can be seen in Figure 10.4. This figure illustrates why TRIDYN simulations are preferred over SRIM simulations for obtaining depth profiles: if SRIM simulations were used, an increase in the number of Tb particles would not change the distribution, only the fraction of Tb would increase.

These implantation profiles are used to generate both the target that is used for the SRIM simulations and the recoiling europium particles. The target that is used for the SRIM simulation is generated from the implantation profile obtained from the TRIDYN simulation, which is divided into multiple bins. The recoiling europium ions¹ are generated at different depths (determined by the depth profile) and with a velocity vector that is isotropically distributed in space. These ions are then simulated by the use of SRIM².

SRIM is a stationary simulation package, which implies that the target is not updated, and thus, each of the generated ions recoils from the same target. Therefore, performing numerous simulations (≈ 90000 recoiling particles) can provide an estimate for the initial sputtering yield.

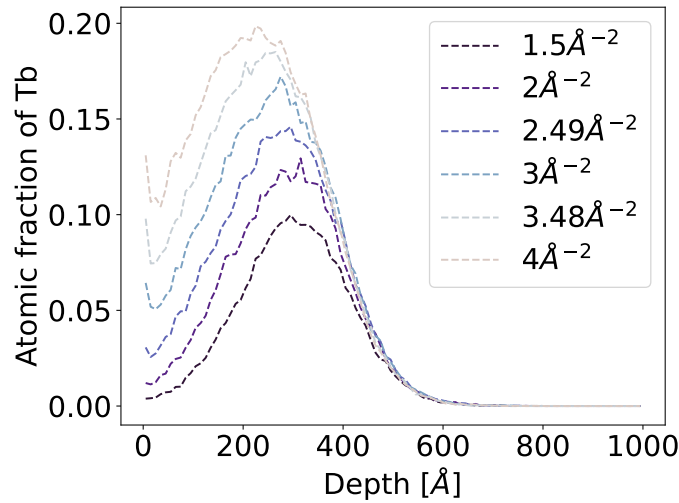


Figure 10.4: Depth profile (atomic fraction of terbium at a specific depth in the foil) for terbium upon implantation in Al for different implanted fluences.

¹Only the recoiling ^{145}Eu particles are simulated as the recoiling α -particles can be neglected due to their smaller mass: a simulation was performed and the recoil-sputtering yield was found to be 0.006653 for Al and 0.000667 for Tb, which is indeed negligible compared to the recoil-sputtering yield due to the recoiling europium particles (18 for Al and 1 for Tb).

²This was done by using a code to generate the SRIM inputfiles from SRIM outputfiles that was provided by Michael Heines and adapted towards TRIDYN inputfiles.

In order to evaluate the importance of these recoil-sputtering events, the activity that is lost after 1 hour due to recoil-sputtering as well as the amount of aluminum that is recoil-sputtered will be calculated for each of the performed simulations. As an example, the calculations are performed for an incoming fluence equal to $3\text{\AA}^{-2}$. The results for the sputtering yields due to the recoiling europium particles for an incoming fluence of $3\text{\AA}^{-2}$ are summarized in Figure 10.5 and Table 10.1. For each α -decay, on average, 18 aluminum particles and 1 terbium particle are sputtered out of the collection foil. Additionally, it is noticed that the sputtering yield for Tb is the largest close to the surface. The latter indicates why it is important to also track the amount of aluminum that is sputtered from the surface: as Al particles are removed from the foil, Tb particles move closer towards the surface, i.e., the region where the sputter yield of Tb is the largest.

For an incoming fluence of $3\text{\AA}^{-2}$ the retained fluence after implantation is equal to $2.9\text{\AA}^{-2}$, assuming that the implantation spot is $2\pi\sigma_r = 0.05\text{cm}^2$ (implantation in the center of the beam), this corresponds to $1.4 \cdot 10^{15}$ particles. Thus, initially, an activity of 65124.88MBq is present³. The number of particles that are lost after one hour due to the decay is given by:

$$\begin{aligned} N_{\text{lost}}(1\text{h}) &= N(t_0) - N(1\text{h}) \\ &= 1.387 \cdot 10^{15} (1 - \exp(-4.696 \cdot 10^{-5}\text{s}^{-1} \cdot 60 \cdot 60\text{s})) = 2.15732 \cdot 10^{14}, \end{aligned} \quad (10.2)$$

which corresponds to an activity lost due to the decay of 10130.8MBq. From the branching ratio, it is concluded that 17% of the decays goes via the α -decay channel. Thus, $3.7 \cdot 10^{13}$ recoil events need to be taken into account. The sputtering yield for terbium is equal to 1.0398(34)⁴. Therefore, an additional activity of 2.7517(90)% compared to the retained fluence after implantation is lost upon recoil-sputtering.

Additionally, the effect of recoil-sputtering on the aluminum foil is calculated. After 1 hour, the thickness of the aluminum layer that is lost due to recoil-sputtering ($t_{\text{lost}}(\text{Al})$) is given by:

$$\begin{aligned} t_{\text{lost}}(\text{Al}) &= Y_{\text{sp.,Al}} \cdot \frac{\text{Number of recoil events}}{N(\text{Al}) \cdot \text{surface of the foil}} \\ &= 18.2562(143) \cdot \frac{3.67 \cdot 10^{13}}{0.06\text{\AA}^{-3} \cdot 0.048 \cdot 10^{16}\text{\AA}^2} = 23.2640(183)\text{\AA}, \end{aligned} \quad (10.3)$$

where the implantation spot is equal to 0.048cm^2 .

³Note that this is a rather large activity, however, a pure beam of ^{149}Tb was used rather than a beam that consists only for 0.2% of ^{155}Tb as in the collection of 2021. Moreover, the half lifetime of ^{149}Tb is smaller than for ^{155}Tb which also implies a larger activity.

⁴The error on the sputtering yields corresponds to the statistical uncertainty, which is determined using Poisson statistics: $\sigma(Y_{\text{sp.}}) = \sigma\left(\frac{\text{Number of sputtered particles}}{\text{Number of incoming particles}}\right) = \frac{\sqrt{\text{Number of sputtered particles}}}{\text{Number of incoming particles}} = \frac{\sqrt{Y_{\text{sp.}} \cdot \text{Number of incoming particles}}}{\text{Number of incoming particles}} = \sqrt{\frac{Y_{\text{sp.}}}{\text{Number of incoming particles}}}$.

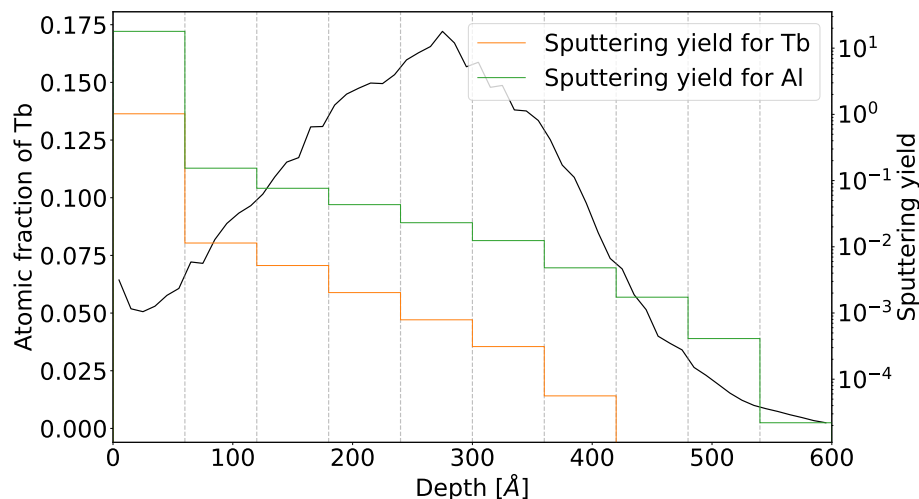


Figure 10.5: Sputtering yield of Tb and Al, together with the atomic fraction of Tb for the target used in SRIM as a function of the depth of the implantation foil.

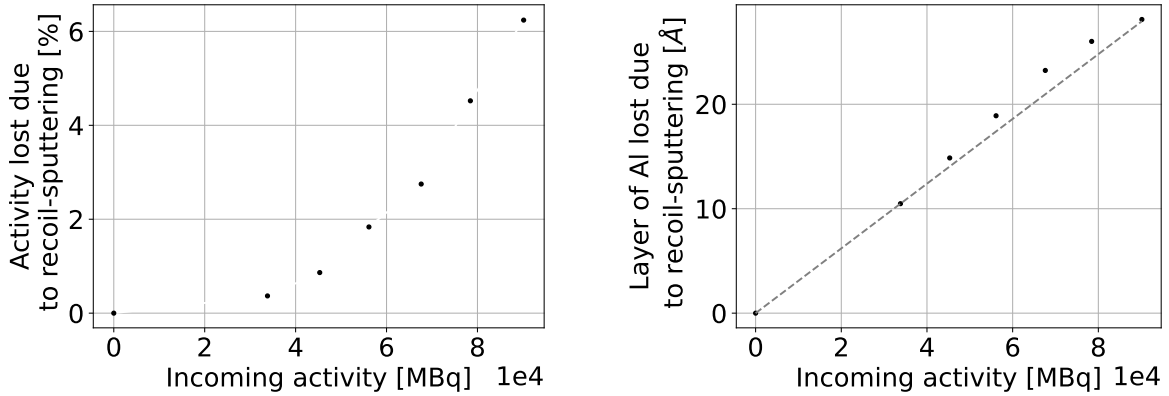
	Sputtering yield of Al	Sputtering yield of Tb
Total	18.2562(143)	1.0398(34)
0-60Å	98.27%	98.1%
60-120Å	0.84%	1.10%
120-180Å	0.42%	0.50%
180-240Å	0.24%	0.1955%
240-300Å	0.13%	0.0759%
300-360Å	0.07%	0.0299%
360-420Å	0.03%	0.0054%
420-480Å	0.0095%	0%
480-540Å	0.0023%	0%
540-600Å	0.00012%	0%
600-2000Å	0%	0%

Table 10.1: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = 3Å^{-2} , $E(^{149}\text{Tb}) = 60\text{keV}$, implantation spot = 0.048cm).

These calculations were repeated for each of the simulations that have been performed and the results are summarized in Table 10.2 and Figure 10.6. The fraction of the activity corresponding to ^{149}Tb that is lost due to recoil-sputtering increases as the incoming activity is increased. Two main reasons have been identified, namely that for increasing incoming activities, more ^{149}Tb particles are present in the foil and thus if a recoil occurs, the chance of interacting with a ^{149}Tb particle increases. Secondly, if the incoming activity increases, more sputtering of the aluminum has occurred (upon implantation, preliminary to the recoil-sputtering). Therefore, the ^{149}Tb particles are located closer to the surface and are consequently easier sputtered away due to the recoiling europium particles. While the first effect is approximately linear with the implanted fluence, the second introduces a major non-linearity.

Moreover, as the incoming activity increases, the layer of aluminum that is lost increases as well. The main reason is that for larger incoming activities, more ^{149}Tb is present in the foil and thus more recoil events can cause recoil-sputtering of Al. Also, the sputtering yield of aluminum compared to the sputtering yield of terbium decreases for increasing incoming terbium fluences. The main reason is that more terbium, and thus relatively less aluminum, is present in the foil. This is also reflected in Figure 10.6b, where the dashed line shows the expected behavior if the recoil-sputtering yield of aluminum would stay constant. Initially, as the incoming activity of ^{149}Tb increases, more aluminum is recoil-sputtered as for increasing incoming activities of ^{149}Tb , ^{149}Tb is located closer to the surface and thus, on average, the recoils reach closer to the surface and therefore more aluminum particles are recoil-sputtered away. However, if the incoming activity increases more, the relative amount of aluminum compared to terbium decreases and thus, per decaying mother nucleus, less aluminum will be sputtered from the foil.

Upon comparison of the fraction lost due to recoil-sputtering with the activity lost due to the decay, it is concluded that the loss due to the decay dominates. However, the branching ratio for the α -decay channel is rather small (17%).



(a) Fraction of the activity of ^{149}Tb lost due to recoil-sputtering [%] as a function of the incoming activity.

(b) Width of the aluminum layer lost due to recoil-sputtering as a function of the incoming activity.

Figure 10.6: Activity of ^{149}Tb and width of the aluminum layer lost due to recoil-sputtering, for an incoming energy = 60keV and an implantation spot of 0.048Å.

Incoming activity [MBq]	Sputtering yield		Activity lost due to recoil-sputtering		Thickness of aluminum layer that is lost [Å]
	Al	Tb	f_i [%]	f_r [%]	
33811.2	15.8795(133)	0.13880(125)	0.3665(34)	0.3670(34)	10.4813(88)
45307	16.8980(137)	0.32679(191)	0.8576(51)	0.8641(51)	14.8553(121)
56126.592	17.5223(140)	0.6942(28)	1.8042(72)	1.8356(74)	18.8982(151)
67622.4	18.2562(143)	1.0398(34)	2.6478(87)	2.7494(90)	23.2441(183)
78441.98	18.1600(142)	1.7100(44)	4.2247(109)	4.5215(117)	26.022(21)
90163.2	17.8500(141)	2.3600(52)	5.577(123)	6.2402(138)	28.118(23)

Table 10.2: Summarizing table on recoil-sputtering for implantation of ^{149}Tb in aluminum for an implantation energy = 60keV and an implantation spot = 0.048cm^2 . f_i and f_r are the fractions compared to the incoming activity and retained activity after implantation of ^{149}Tb respectively. A more detailed table can be found in Table E.6.

10.2.1 Dependence on the energy

For self-sputtering, a large improvement for increasing implantation energies was found. Therefore, it is examined how increasing the energy affects recoil-sputtering. Simulations have been performed for the same incoming activities of ^{149}Tb as previously, but for an implantation energy of 100keV. Again, the implantation of terbium in aluminum is simulated with TRIDYN (implantation profile, see Figure 10.7), thereafter, SRIM was used for the simulation of the recoils.

The results are summarized in Figure 10.8 and Table 10.3, indicating that the fraction of the activity of ^{149}Tb that is lost, decreases as the energy increases. The main reason for this behavior is that for increasing energies, terbium is implanted deeper in the foil and therefore self-sputtering limits are reached later. Thus, after implantation of the same activity, the ^{149}Tb atoms are located deeper into the foil than for implantation at 60keV. As a result, the collision cascades caused by the recoiling daughter nucleus end at greater distances away from the surface, leading to less recoil-sputtering. For the same reason, the thickness of the layer of Al that is removed due to recoil-sputtering decreases as the implantation energy increases.

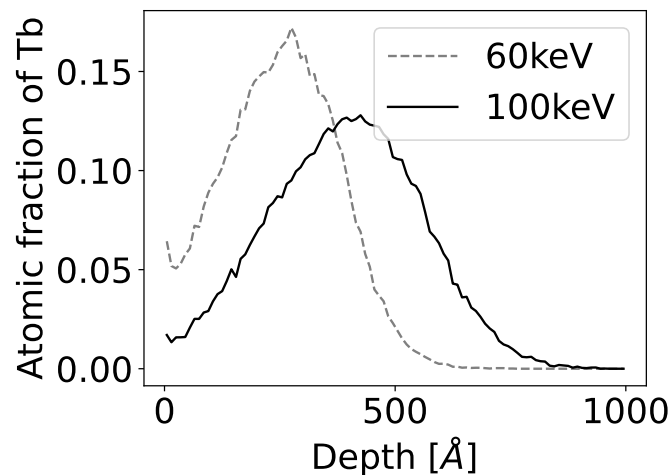
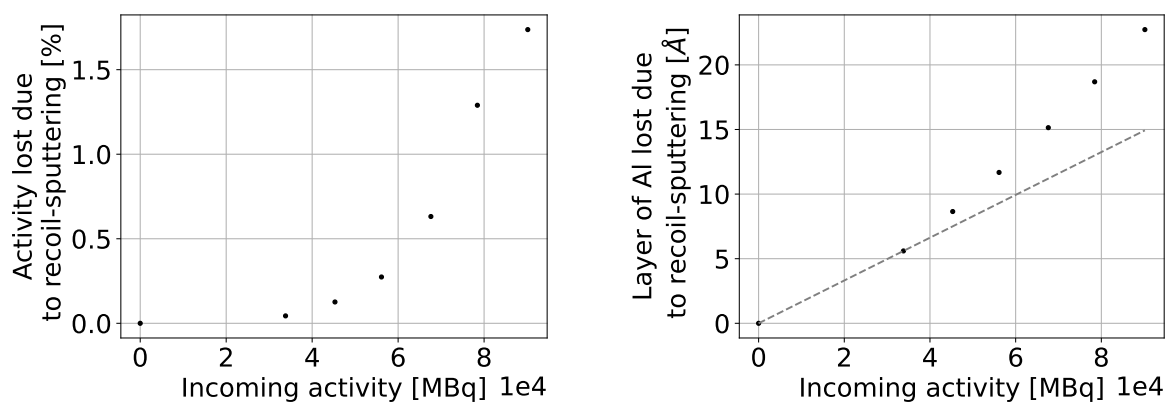


Figure 10.7: Depth profile (atomic fraction of terbium at a specific depth in the foil) for the implantation of 100keV terbium in aluminum with an incoming fluence of $3\text{\AA}^{-2}$ compared to the depth profile for 60keV implantation.



(a) Fraction of the activity of ^{149}Tb lost due to recoil-sputtering [%] as a function of the incoming activity. **(b)** Width of the aluminum layer lost due to recoil-sputtering as a function of the incoming activity.

Figure 10.8: Activity of ^{149}Tb and width of the aluminum layer lost due to recoil-sputtering, for an incoming energy = 100keV and an implantation spot of $0.048\text{\AA}$.

Incoming activity [MBq]	Sputtering Yield		Activity lost due to recoil-sputtering		Thickness of aluminum layer that is lost [Å]
	Al	Tb	f_i [%]	f_r [%]	
33811.2	8.4939(98)	0.01660(42)	0.04378(111)	0.04385(111)	5.6008(65)
45307	9.7973(105)	0.04770(73)	0.12576(193)	0.12613(194)	8.6527(93)
56126.592	10.6960(109)	0.10360(108)	0.2725(29)	0.2739(29)	11.6759(119)
67622.4	11.5826(113)	0.23881(163)	0.6244(43)	0.6316(44)	15.1426(148)
78441.98	12.4379(118)	0.4877(23)	1.2896(61)	1.2896(61)	18.6940(178)
90163.2	13.3800(122)	0.6703(28)	1.7086(72)	1.737(75)	22.73(21)

Table 10.3: Summarizing table on recoil-sputtering for implantation of ^{149}Tb in aluminum for an implantation energy = 100keV and an implantation spot = 0.048cm^2 . f_i and f_r are the fractions compared to the incoming activity and retained activity after implantation of ^{149}Tb respectively. A more detailed table can be found in Table E.7.

10.2.2 Dependence on the implantation spot

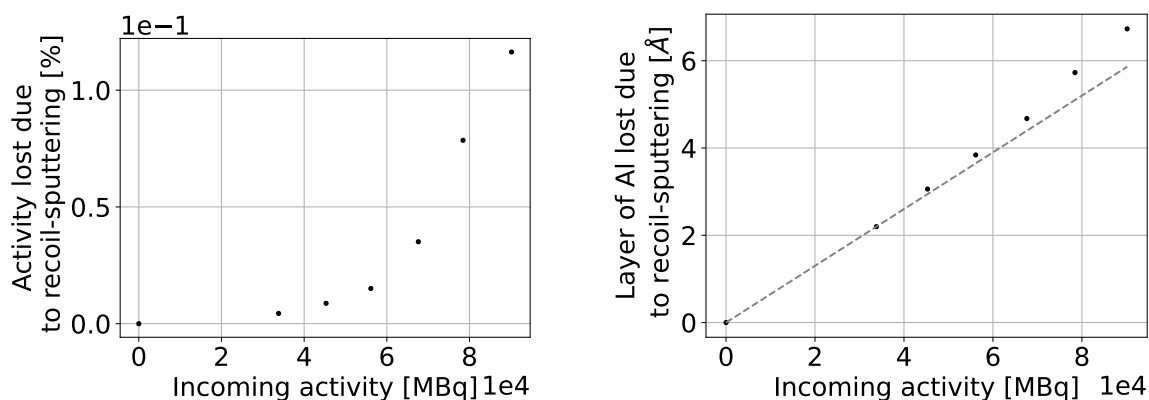
Lastly, the improvement that is obtained by increasing the implantation spot is examined. As the implantation spot is increased, the amount of ^{149}Tb is more spread (in the plane of the surface) across the Al foil. This results in a smaller fluence and therefore less self-sputtering. If the area of implantation is increased by a factor 4, the fluence will decrease by a factor 4. In other words, a fluence $\frac{\psi_0}{4}$ implanted in an area $4A_0$ will contain the same amount of particles as a fluence ψ_0 implanted in an area A_0 . The importance of this effect on recoil-sputtering will now be examined by performing simulations for the same incoming activity as before, but for a 4 times larger implantation spot.

The results are summarized in Figure 10.9 and Table 10.4. For an increasing implantation spot, the activity that is lost due to recoil-sputtering as well as the thickness of the Al layer that is removed decrease. This is mainly because self-sputtering limits are reached later for increasing implantation spots and thus the recoiling daughter nuclei are located deeper into the foil. Additionally, less ^{149}Tb per surface area is present in the foil to initiate recoils and to interact with.

When comparing the effect of increasing the energy of the impinging beam to increasing the implantation spot, a huge reduction in the activity of terbium that is lost is observed for the latter (≈ 18 -fold). However, the decrease in the thickness of the aluminum layer that is removed is relatively smaller (≈ 3 -fold), which is explained by the fact that for an increasing implantation spot, the amount of Al to Tb increases and therefore the sputtering yield of Al increases.

Incoming activity [MBq]	Sputtering Yield		Activity lost due to recoil-sputtering		Thickness of aluminum layer that is lost [$\text{\AA}$]
	Al	Tb	f_i [%]	f_r [%]	
33811.2	13.3000(122)	0.001679(137)	0.00444(37)	0.00444(37)	2.1979(21)
45307	13.8300(124)	0.003300(192)	0.00873(51)	0.00873(51)	3.0624(28)
56126.592	14.0040(125)	0.00570(26)	0.01507(69)	0.01507(69)	3.8414(35)
67622.4	14.1360(126)	0.01328(39)	0.03510(104)	0.03510(104)	4.6701(42)
78441.98	14.9436(129)	0.02970(58)	0.07852(154)	0.07853(154)	5.7282(50)
90163.2	15.270(131)	0.04400(70)	0.116302(186)	0.116343(186)	6.726(58)

Table 10.4: Summarizing table on recoil-sputtering for implantation of ^{149}Tb in aluminum for an implantation energy = 60keV and an implantation spot = 0.192cm^2 . f_i and f_r are the fractions compared to the incoming activity and retained activity after implantation of ^{149}Tb respectively. A more detailed table can be found in Table E.14



(a) Fraction of the activity of ^{149}Tb lost due to recoil-sputtering [%] as a function of the incoming activity.

(b) Width of the aluminum layer lost due to recoil-sputtering as a function of the incoming activity.

Figure 10.9: Activity of ^{149}Tb and width of the aluminum layer lost due to recoil-sputtering, for an incoming energy = 60keV and an implantation spot of $0.192\text{\AA}$.

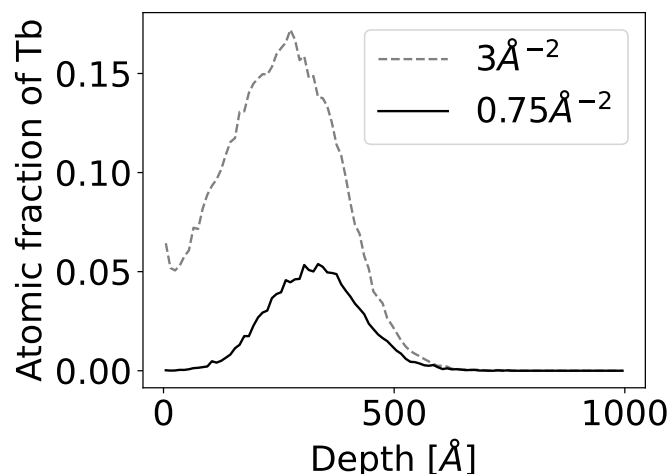


Figure 10.10: Depth profile (atomic fraction of terbium at a specific depth in the foil) for the implantation of terbium in aluminum for an implantation spot equal to 0.192Å compared to the depth profile for an implantation spot of 0.048Å .

10.3 Conclusion

In this chapter, a first approximation for the importance of recoil-sputtering has been proposed. Already for an α -decaying particle with a branching ratio of 17%, recoil-sputtering has some importance at large impinging activities. Recoil-sputtering, although causing a minor loss in activity (0.001%-10%), exhibits a notable dependence on the characteristics of the implantation process. In particular, further away from the self-sputtering limits, the occurrence of recoil-sputtering diminishes. To be precise, when the incoming activity decreases, the beam energy increases or the implantation spot increases, recoil-sputtering is minimized. Additionally, for increased waiting times, more recoil-sputtering will occur. Lastly, a comparison with experiments is provided in Appendix F for ^{225}Ac .

General conclusion & future research

Goodbye..?? Oh no, please.
Can't we go back to page one
and do it all over again?

Winnie The Pooh

Limitations and future research

This thesis has contributed to further investigations into the optimization of the efficiency of radionuclide collections. However, further research is still needed to improve this efficiency. To ensure reliable analysis of future MEDICIS collections, it is crucial to obtain an accurate efficiency fit, as the activity determination relies on this fit. Hence, it is necessary to conduct new efficiency measurements. Additionally, there is a need for further investigations into the ISOL method. For externally irradiated targets, elution is employed for chemical purification prior to the ISOL method. However, systematic research should be conducted on the different eluents that can be used and on the collection of terbium via its molecules to optimize the vaporization from the target. Regarding self-sputtering, further investigation into implantation in sugars is needed. Furthermore, additional research is required to conclude on the experimental confirmation of the TRIDYN simulations. The implantations were carried out simultaneously with the deadline of this master's thesis. The results available at the time of submission seem to point to a different picture than originally anticipated and will require further analysis beyond the timeline of this thesis. Lastly, for the recoil-sputtering simulations, TRIDYN and SRIM were combined, resulting in semi-static simulations. To improve the accuracy and reliability of the results, it is recommended to investigate methods to extend these simulations towards dynamic simulations.

Conclusion

Throughout this thesis, the collection of radioisotopes, specifically ^{155}Tb , was analyzed in order to enhance the collection efficiency. The study covered three distinct steps of the radionuclide collection process: the ISOL mass separating method for externally irradiated targets, self-sputtering during implantation of the mass-separated particles in the collection foil and recoil-sputtering during the transport of this collection foil.

To conclude on this thesis, Equation 1.9 will be revisited:

$$\varepsilon_{\text{tot}} = \varepsilon_{\text{diff}} \cdot \varepsilon_{\text{eff}} \cdot \varepsilon_{\text{ion}} \cdot \varepsilon_{\text{sep}} \cdot \varepsilon_{\text{transport}} \cdot \varepsilon_{\text{implant}} \cdot C_{\text{decay}} \quad (10.4)$$

which can be regrouped together in two main terms:

$$\varepsilon_{\text{tot}} = \varepsilon_{\text{ISOL}} \cdot \varepsilon_{\text{implant}}. \quad (10.5)$$

In Part I, $\varepsilon_{\text{ISOL}}$ was considered by analyzing the collections of 2022. It was concluded that investigating the laser ON-OFF effect systematically and that acquiring activity spectra every 5 minutes was a beneficial choice. In that way, a measure for the amount of terbium with respect to its contaminants is obtained. However, no successful collections were performed in 2022. The main hypothesis is that the type and the concentration of the eluent that was used during the target preparation influences the collection efficiency. Indicating that for future collections, the chemistry of the target should be investigated prior to the collection. Once the separation of the terbium beam and its contaminants is obtained, terbium is implanted in a collection foil and self-sputtering needs to be considered ($\varepsilon_{\text{implant}}$), which was discussed in Part II of this thesis. It was concluded that

self-sputtering is more pronounced for heavier implantation foils as was expected based on typical self-sputtering behavior. For example, implantation in Al is preferred over implantation in Zn. Furthermore, implantation at higher energies should be considered at the facilities that are currently being commissioned, as well as beam wobbling or foil replacement techniques. Additionally, it was shown that the amount of impurities should be minimized to minimize self-sputtering, which is also crucial when considering the use in hospitals. Hence, collections where chemical preparation is conducted prior to the ISOL method demonstrate substantial potential. However, the added value of chemical separation prior to the collection should be evaluated against the associated logistical challenges, including increased personal radiation exposure, management of nuclear waste and longer waiting times. Besides, the total current on the foil was found to be a useful indicator for estimating the onset of self-sputtering or thus for when the foil is preferably removed. Furthermore, the feasibility for experimental confirmation was demonstrated. Lastly, recoil-sputtering was investigated. The main conclusion was that recoil-sputtering is responsible for a rather small (0.001%-10%) additional loss during the transport of α -decaying nuclei that is dependent on the implantation conditions. Additionally, recoil-sputtering is minimized in those conditions where self-sputtering is minimal.

In conclusion, a thorough analysis of the entire collection process, spanning from target preparation to the transport towards the hospital, has been performed. In that way, this thesis contributed to the optimization of future medical isotope collections.

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Appendices

A

Analysis of the Kromek spectra

In this appendix, an elaboration on the determination of the activity corresponding to a radionuclide is provided. Moreover, both the statistical and systematic errors are considered. The first section will focus on determining the activity corresponding to a single peak in a spectrum such as ^{139}Ce , while the second section will cover the calculation for a radionuclide with multiple peaks in a spectrum such as ^{155}Tb . Lastly, the activity that is collected in between two activity measurements will be discussed.

A.1 Activity corresponding to one peak

The activity corresponding to one peak in a γ -spectrum is determined by:

$$A = \frac{N(E)}{I(E) \cdot \varepsilon_{\text{abs}}(E) \cdot t}, \quad (\text{A.1})$$

where $N(E)$ is the number of counts in the peak corresponding to an energy E , $I(E)$ is the intensity of the γ -line of the corresponding energy (which can be found in, e.g., [66]) and $\varepsilon_{\text{abs}}(E)$ is the corresponding absolute detection efficiency. The lifetime, t , is calculated by the Kromek detector software as the measurement time minus the detector's dead time.

Efficiency measurements for the Kromek detector were performed in [39] using four different sources: ^{152}Eu , ^{137}Cs , ^{133}Ba and ^{60}Co . A fit for the efficiencies was made, based on the expected behavior of the efficiency as a function of the energy:

$$\varepsilon_{\text{abs}}(E) = \exp \left(\sum_{i=0}^4 a_i \log(E)^i \right). \quad (\text{A.2})$$

Moreover, the uncertainties on the activities were considered and propagated towards an uncertainty on the efficiencies. This was achieved by performing multiple fits for different activities taken from each source's activity distributions. In that way, for each energy, a distribution of efficiencies is obtained. For each energy, the mean of the corresponding efficiency distribution is taken as the efficiency for that energy and the uncertainty ($\delta\varepsilon$) is taken to be the 1σ -value of that distribution¹. The results are shown in Figure A.1. When

¹The code to perform this type of fit was provided by Michael Heines and adapted towards returning the mean value instead of the value corresponding to the first fit.

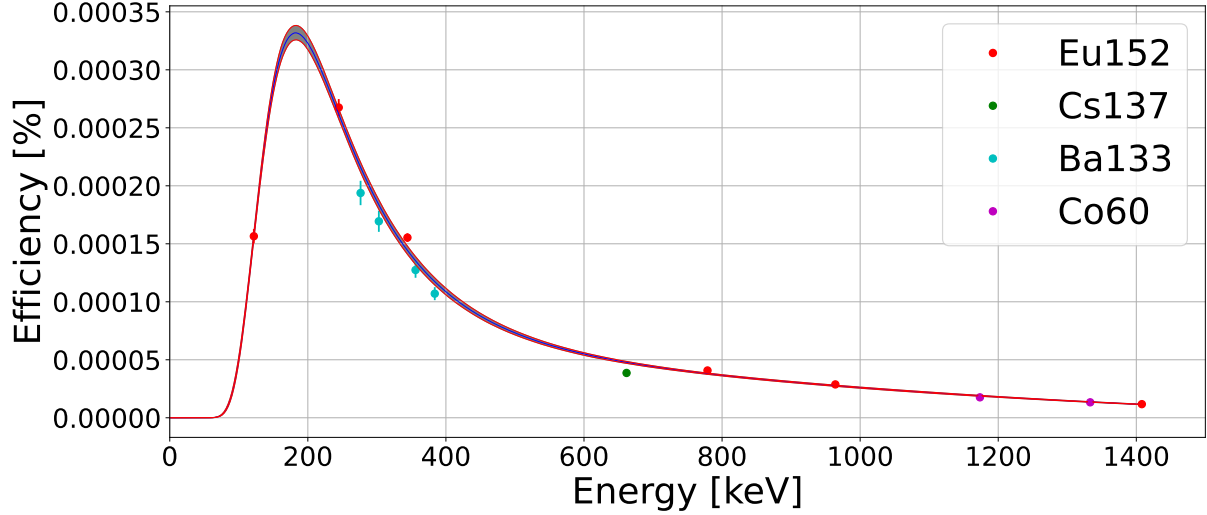


Figure A.1: Efficiency as a function of the energy for the different sources used in [39] together with the fitted value and the error.

determining the activity, the uncertainty on the efficiency contributes to a systematic error on the activity, as it is a consistent error for the setup and thus for every measurement.

However, as can be noted from Figure A.1, no data-points are present in the region of the peak, the fit is thus not very reliable. Therefore, it is advised to repeat the efficiency measurements by using sources with energy peaks between 100keV and 300keV, such as ^{133}Ba , ^{152}Eu , ^{241}Am , ^{57}Co , ^{226}Ra , ^{137}Cs , ^{158}Tb and ^{229}Th . Additionally, for example for the ^{133}Ba source, a systematic offset is found, which indicates an inaccurate quoted source activity.

In order to obtain $N(E)$, the peak is fitted by a Gaussian peak by the use of the `lmfit` package in python. The inverse of the statistical error is taken as a weight. $N(E)$ is then determined by the fit for the amplitude, the corresponding statistical error (Δc) is also determined by the fit. Nonetheless, as a substantial amount of background is present, therefore a SNIP algorithm was used to eliminate the background.

Concluding, the activity corresponding to one peak is given by:

$$A = \frac{N(E)(\pm\Delta c)}{I(E) \cdot \varepsilon(E)[\pm\delta\varepsilon] \cdot t} = \frac{N(E)}{I(E) \cdot \varepsilon(E) \cdot t} \left(\pm \frac{\Delta c}{I(E) \cdot t \cdot \varepsilon(E)} \right) \left[\pm \frac{N(E)}{I(E) \cdot t \cdot \varepsilon(E)} \frac{\delta\varepsilon(E)}{\varepsilon(E)} \right], \quad (\text{A.3})$$

A.2 Activity corresponding to an isotope with multiple peaks in the spectrum

The activity corresponding to an isotope is calculated by taking the weighted sum of the activity corresponding to each of the peaks corresponding to γ -lines of the isotope at stake:

$$A = \frac{\sum_{\text{peaks}} A_i \cdot w_i}{\sum_{\text{peaks}} w_i} \quad \text{with } w_i = \frac{1}{\sigma(A_i)^2} = \frac{1}{\left(\frac{\Delta c(E_i)}{I(E_i) \cdot t \cdot \varepsilon(E_i)} + \frac{N(E_i)}{I(E_i) \cdot t \cdot \varepsilon(E_i)} \frac{\delta\varepsilon(E_i)}{\varepsilon(E_i)} \right)^2} \quad (\text{A.4})$$

The corresponding error is given by:

$$\frac{1}{\sqrt{\sum_{\text{peaks}} \frac{1}{\sigma(A_i)}}} \quad (\text{A.5})$$

A.3 Activity collected within two measurements

The activity that is collected in between 2 Kromek measurements can be calculated as follows:

$$A_2 - A_1 e^{-\lambda t}. \quad (\text{A.6})$$

However, as the same peaks are used to calculate both activities, the uncertainties on both activities cannot be treated independently. Therefore, the collected activity within two measurements is first calculated for each of the peaks separately. Thereafter, the weighted mean of the collected activities corresponding to the different peaks is used:

$$A_{\text{collected}} = \frac{\sum_{\text{peaks}} A_{\text{collected},i} \cdot w_i}{\sum_{\text{peaks}} w_i}$$

with $A_{\text{collected},i} = A_{2,i} - A_{1,i} e^{-\lambda t}$

$$\text{and } w_i = \frac{1}{\sigma(A_{\text{collected},i})^2} = \frac{1}{\left[\left(\frac{\Delta c_2(E_i)}{I(E_i) \cdot t_2 \cdot \varepsilon(E_i)} - e^{-\lambda t} \frac{\Delta c_1(E_i)}{I(E_i) \cdot t_1 \cdot \varepsilon(E_i)} \right) + (A_{2,i} - A_{1,i} e^{-\lambda t}) \frac{\delta \varepsilon(E_i)}{\varepsilon(E_i)} \right]^2} \quad (\text{A.7})$$

The corresponding uncertainty is given by $\frac{1}{\sqrt{\sum_{\text{peaks}} w_i}}$.

B

Distribution of the beam profile

In this appendix, a summary on the distribution of the beam impinging on the foil is given. It is known that the number of particles in the beam is Gaussian distributed and thus the probability density function is given by:

$$g(x, y) = \mathcal{N} \exp\left(-\frac{1}{2} \begin{pmatrix} x & y \end{pmatrix} \begin{pmatrix} \frac{1}{\sigma_x^2} & 0 \\ 0 & \frac{1}{\sigma_y^2} \end{pmatrix} \begin{pmatrix} x \\ y \end{pmatrix}\right) \quad (\text{B.1})$$

$$= \mathcal{N} \exp\left(-\frac{1}{2} \left(\frac{x^2}{\sigma_x^2} + \frac{y^2}{\sigma_y^2}\right)\right), \quad (\text{B.2})$$

where $\mathcal{N}$ is the normalization factor, σ_x the standard deviation on the x -component of the beam and σ_y the standard deviation on the y -component of the beam. Moreover, it was assumed that the x and y direction of the beam are uncorrelated.

B.1 Calculation of the normalization factor

In this section, the normalization factor N for the distribution of the beam impinging on the foil will be calculated. The total number of particles N in the beam can be calculated

as follows:

$$\begin{aligned}
N &= \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \mathcal{N} \exp\left(-\frac{1}{2}\left(\frac{x^2}{\sigma_x^2} + \frac{y^2}{\sigma_y^2}\right)\right) dx dy \\
&\quad \downarrow \begin{cases} x = \frac{\sigma_x}{\sigma_r} r \cos(\theta) \\ y = \frac{\sigma_y}{\sigma_r} r \sin(\theta) \end{cases} \quad \text{with } \sigma_r = \sqrt{\sigma_x \sigma_y} \\
&= \mathcal{N} \int_0^{2\pi} \int_0^{+\infty} \exp\left(-\frac{1}{2}\left(\frac{r}{\sigma_r}\right)^2\right) r dr d\theta \\
&= 2\pi \mathcal{N} \int_0^{+\infty} \exp\left(-\frac{1}{2}\left(\frac{r}{\sigma_r}\right)^2\right) r dr \\
&\quad \downarrow u = \left(\frac{r}{\sigma_r}\right)^2 \\
&= 2\pi \mathcal{N} \int_0^{+\infty} \exp\left(-\frac{1}{2}u\right) \frac{\sigma_r^2}{2} du \\
&= \pi \mathcal{N} \sigma_r^2 \left[-2 \exp\left(-\frac{1}{2}u\right) \right]_0^{+\infty} \\
&= 2\pi \mathcal{N} \sigma_r^2.
\end{aligned} \tag{B.3}$$

Therefore $\mathcal{N} = \frac{N}{2\pi\sigma_r^2}$ and thus:

$$\begin{aligned}
g(x, y) &= \frac{N}{2\pi\sigma_r^2} \exp\left(-\frac{1}{2}\left(\frac{x^2}{\sigma_x^2} + \frac{y^2}{\sigma_y^2}\right)\right) \\
g(r) &= \frac{N}{2\pi\sigma_r^2} \exp\left(-\frac{1}{2}\left(\frac{r}{\sigma_r}\right)^2\right)
\end{aligned} \tag{B.4}$$

B.2 Fluence implanted in a ring

In this section, it will be proven that the fluence implanted in a ring defined by the radii r and $r + \Delta r$ ($R(r, r + \Delta r)$) is given by:

$$\psi_{R((r, r+\Delta r))} = \frac{N}{\pi((r + \Delta r)^2 - r^2)} \left(\exp\left(-\frac{r^2}{2\sigma_r^2}\right) - \exp\left(-\frac{(r + \Delta r)^2}{2\sigma_r^2}\right) \right), \tag{B.5}$$

with N the total number of particles and $\sigma_r = \sqrt{\sigma_x \sigma_y}$, the standard deviation of the Gaussian beam profile.

First, the number of particles in a ring with radius r and $r + \Delta r$, $N(R(r, r + \Delta r))$, is

calculated. With the use of Equation B.4, it is found that:

$$\begin{aligned}
 N(R(r, r + \Delta r)) &= \int_{x=r}^{x=r+\Delta r} \frac{N}{2\pi\sigma_r^2} \exp\left(\frac{-x^2}{2\sigma_r^2}\right) x dx \cdot 2\pi \\
 &= \frac{N}{\sigma_r^2} \int_{x=r}^{x=r+\Delta r} \exp\left(\frac{-x^2}{2\sigma_r^2}\right) x dx \\
 &= \frac{N}{\sigma_r^2} \left[\frac{1}{2} \int_{u=r^2}^{u=(r+\Delta r)^2} \exp\left(\frac{-u}{2\sigma_r^2}\right) du \right] \\
 &= \frac{N}{2\sigma_r^2} \left[-\exp\left(\frac{-u}{2\sigma_r^2}\right) (2\sigma_r^2) \right]_{u=r^2}^{u=(r+\Delta r)^2} \\
 &= N \left[\exp\left(\frac{-r^2}{2\sigma_r^2}\right) - \exp\left(\frac{-(r+\Delta r)^2}{2\sigma_r^2}\right) \right], \tag{B.6}
 \end{aligned}$$

where $u = x^2$ was used. The area corresponding to this ring is given by $\pi[(r + \Delta r)^2 - r^2]$ and thus the fluence in this ring is equal to:

$$\psi_{R((r, r+\Delta r))} = \frac{N}{\pi[(r + \Delta r)^2 - r^2]} \left[\exp\left(\frac{-r^2}{2\sigma_r^2}\right) - \exp\left(\frac{-(r + \Delta r)^2}{2\sigma_r^2}\right) \right]. \tag{B.7}$$

B.3 Worst case scenario fluence that is implanted

Now, a derivation for the maximal possible fluence that can be implanted, i.e., all the particles impinge at radius zero, is provided. More specifically, it is proven that the fluence for this case is given by:

$$\psi = \frac{N}{2\pi\sigma_r^2}, \tag{B.8}$$

with N the total number of particles and $\sigma_r = \sqrt{\sigma_x \sigma_y}$, the standard deviation of the Gaussian beam profile.

First, the number of particles implanted with a radius in $[0, \epsilon]$ is calculated by the use of Equation B.4:

$$\begin{aligned}
 N(\epsilon) &= \int_{r=0}^{r=\epsilon} \frac{N}{2\pi\sigma_r^2} \exp\left(\frac{-r^2}{2\sigma_r^2}\right) r dr \cdot 2\pi \\
 &= \frac{N}{\sigma_r^2} \int_{r=0}^{r=\epsilon} \exp\left(\frac{-r^2}{2\sigma_r^2}\right) r dr \\
 &= \frac{N}{\sigma_r^2} \left[-\frac{1}{2} \exp\left(\frac{-x}{2\sigma_r^2}\right) \cdot (2\sigma_r^2) \right]_{x=0}^{x=\epsilon^2} \\
 &= N \left(1 - \exp\left(\frac{-\epsilon^2}{2\sigma_r^2}\right) \right). \tag{B.9}
 \end{aligned}$$

The corresponding surface is given by $\varepsilon^2\pi$ and thus the fluence is given by:

$$\begin{aligned}
 \psi &= \lim_{\varepsilon \rightarrow 0} \frac{\text{number of particles within a radius } \varepsilon}{\text{surface area of circle with radius } \varepsilon} \\
 &= \lim_{\varepsilon \rightarrow 0} \frac{N \left(1 - \exp \left(\frac{-\varepsilon^2}{2\sigma_r^2} \right) \right)}{\varepsilon^2 \pi} \\
 &= \frac{N}{2\pi\sigma_r^2}.
 \end{aligned} \tag{B.10}$$

C

RADIATE proposal

Experimental confirmation of the simulations in TRIDYN was performed by means of high-current implantation and subsequent RBS analysis. A proposal for the high-current implantation was made and submitted to RADIATE. The proposal can be found on the next pages. An overview of the setup and results can be found in [Section 7](#).

RADIATE Transnational Access Proposal



Main applicant (name, institution, country)

Charlotte Duchemin, CERN, Switzerland

Proposal title

Implanted targets for self-sputtering in the context of MEDICIS

Motivation & justification of beam time, and scientific background

Collections performed at MEDICIS, using the ISOL method, are of large importance as they provide radionuclides for radiopharmaceutical research and cancer treatment. It is therefore important to maximize the amount that is retained during a collection. During the collections at MEDICIS, it was observed that the activity measured in the collection chamber was larger than the activity collected on the foil [1]. This indicates that some of the radionuclides were removed from the foil upon implantation. A proposed explanation for this behavior is self-sputtering, an effect where the fluence is sufficiently large such that incoming particles can remove particles that were previously implanted. In order to get a better understanding on this effect, simulations with TRIDYN [2] have been performed. These simulations showed that self-sputtering is indeed relevant for the collections performed at MEDICIS and that under the current conditions, about one third of the activity is lost due to self-sputtering.

Moreover, the self-sputtering behavior is strongly dependent on the energy of the incoming beam (Figure 2), the retained fluence increases a lot if the energy is increased from 30keV to 60keV. Investigation in the behavior as a function of the energy is therefore important in order to minimize the self-sputtering.

We want to examine the reliability of TRIDYN simulations in the lanthanide region. Until now, no reliable estimates for the self-sputtering are available (SRIM [3] cannot handle self-sputtering). In order to benchmark the results from the simulations, implantations of Yb in zinc and aluminum foils are proposed. This requires a number of samples with different implantation fluences, different implantation energies, and subsequent RBS measurements.

If applicable, previous results on this topic (mainly related to the proposer's work)

For the CERN MEDICIS radioisotope collection of July 2021, 2.26(21)[18]MBq of radioactivity was lost due to self-sputtering, which is about 27% of the full sample. This is quite an important part. Moreover, once the self-sputtering limit is reached, implanting more isotopes in the foil will no longer increase the retained activity. This can be seen clearly in Figure 1: increasing the incoming fluence beyond $2\text{\AA}^{-2}$ does no longer yield an increase in the retained fluence, i.e., the amount that is implanted is equal to the amount that is self-sputtered.

These limits were already reached at a few MBq during the aforementioned collection of 2021 (implantation in zinc). As the prospects of MEDICIS are to go up to activities of several GBq, investigation on self-sputtering is needed.

Simulations for both implantation in zinc and aluminum are performed; the results are shown in Figure 1 and 2. The retained fluence deviates from the incoming fluence due to the self-sputtering of Yb. Moreover, it is clear that more self-sputtering occurs for implantation in zinc compared to aluminum.

Also, for implantation in aluminum, the self-sputtering drastically decreases for increasing energies as the atomic number of aluminum is smaller than for zinc.

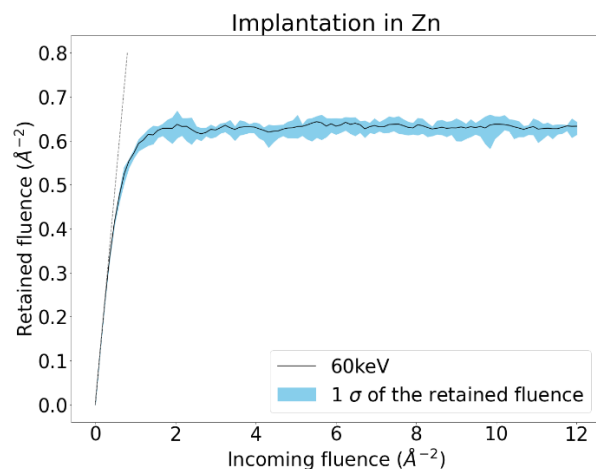


Figure 1: Summarizing graph for the implantation of terbium in zinc.

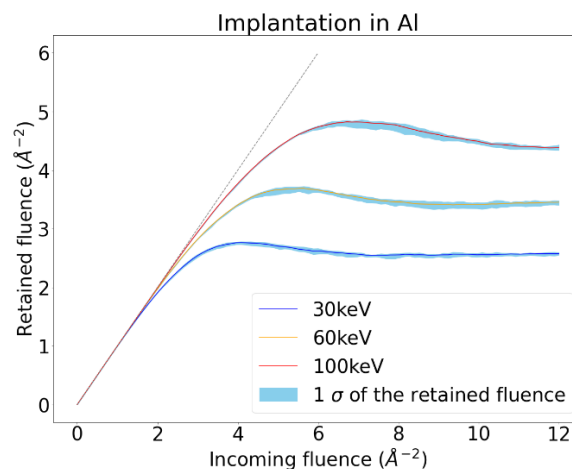


Figure 2: Summarizing graph for the implantation of terbium in aluminum.

Description of the proposed experiment including most relevant technical data

We propose to perform a series of implantations in order to verify the TRIDYN simulations. By performing RBS on the resulting samples, the retained fluence can be plotted as a function of the incoming fluence. A first part of these implantations would focus on the comparison between zinc and aluminum samples. Subsequently, the effect of a changing energy will be investigated. The proposed ion species is $^{173}\text{Yb}^+$ but it can be substituted for any other lanthanide, should this provide less pressure on the existing machine schedule.

The proposed experiments would require a large amount of sample implantations. However, the small size requirement of the samples ($\sim 5 \times 5 \text{ mm}$) will allow for simultaneous implantation for the same beam energy, thereby severely reducing the total required fluence and implantation time.

Next steps after beam time

After the beamtime, RBS measurements will be performed at IMBL (KU Leuven) in order to determine the retained fluences in each of the samples. Based on these results, a comparison with the results from the TRIDYN simulations will be conducted. By doing so, the reliability of TRIDYN can be benchmarked and used for new simulations in the lanthanide region. In that sense, by performing these implantation experiments, the TRIDYN simulations will be verified and can be used in order to research methods to retain more activity. If more activity is retained, more patient doses can be produced per collection, which decreases the cost of the treatment and increases the accessibility.

Ion implantation / irradiation conditions, if known and relevant to experiment

sample name	material	size	side	ion species	energy (keV)	fluence (cm ⁻²)	Temperature (°C)	tilt angle (°)	twist angle (°)
Zn60 - 1	zinc	5x5 mm	1	$^{173}\text{Yb}^+$	60	0.5E16	RT	7	NA
Zn60 - 2	zinc	5x5 mm	1	$^{173}\text{Yb}^+$	60	1E16	RT	7	NA

Zn60 - 3	zinc	5x5 mm	1	$^{173}\text{Yb}^+$	60	1.5E16	RT	7	NA
Zn60 - 4	zinc	5x5 mm	1	$^{173}\text{Yb}^+$	60	2E16	RT	7	NA
Zn60 - 5	zinc	5x5 mm	1	$^{173}\text{Yb}^+$	60	2.5E16	RT	7	NA
Al60 - 1	aluminum	5x5 mm	1	$^{173}\text{Yb}^+$	60	1E16	RT	7	NA
Al60 - 2	aluminum	5x5 mm	1	$^{173}\text{Yb}^+$	60	2E16	RT	7	NA
Al60 - 3	aluminum	5x5 mm	1	$^{173}\text{Yb}^+$	60	3E16	RT	7	NA
Al60 - 4	aluminum	5x5 mm	1	$^{173}\text{Yb}^+$	60	4E16	RT	7	NA
Al60 - 5	aluminum	5x5 mm	1	$^{173}\text{Yb}^+$	60	5E16	RT	7	NA
Al60 - 6	aluminum	5x5 mm	1	$^{173}\text{Yb}^+$	60	6E16	RT	7	NA
Al60 - 7	aluminum	5x5 mm	1	$^{173}\text{Yb}^+$	60	7E16	RT	7	NA
Al60 - 8	aluminum	5x5 mm	1	$^{173}\text{Yb}^+$	60	8E16	RT	7	NA
Al30 - 1	aluminum	5x5 mm	1	$^{173}\text{Yb}^+$	30	1E16	RT	7	NA
Al30 - 2	aluminum	5x5 mm	1	$^{173}\text{Yb}^+$	30	2E16	RT	7	NA
Al30 - 3	aluminum	5x5 mm	1	$^{173}\text{Yb}^+$	30	3E16	RT	7	NA
Al30 - 4	aluminum	5x5 mm	1	$^{173}\text{Yb}^+$	30	4E16	RT	7	NA
Al30 - 5	aluminum	5x5 mm	1	$^{173}\text{Yb}^+$	30	5E16	RT	7	NA
Al30 - 6	aluminum	5x5 mm	1	$^{173}\text{Yb}^+$	30	6E16	RT	7	NA
Al30 - 7	aluminum	5x5 mm	1	$^{173}\text{Yb}^+$	30	7E16	RT	7	NA
Al30 - 8	aluminum	5x5 mm	1	$^{173}\text{Yb}^+$	30	8E16	RT	7	NA

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[2]: W Möller and W Eckstein. Tridyn—a trim simulation code including dynamic composition changes. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 2(1-3):814–818, 1984.

[3]: James F Ziegler, Matthias D Ziegler, and Jochen P Biersack. Srim—the stopping and range of ions in matter (2010). Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 268(11-12):1818–1823, 2010. doi: 10.1016/j.nimb.2010.02.091.

D

Self-sputtering figures for different isotopes

In this appendix, the figures corresponding to Table 9.1 are given. Implantation of different isotopes in different substrates are considered for both implantation at 30keV and 60keV. Each time, the retained fluence is shown as a function of the incoming fluence.

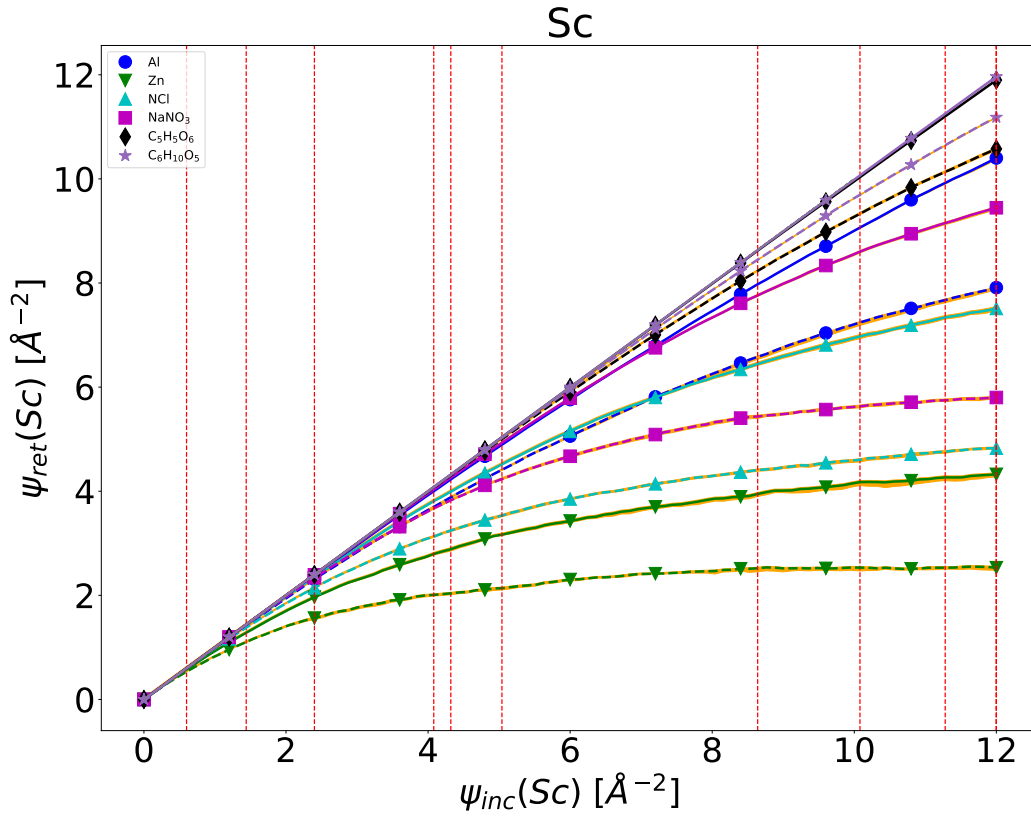


Figure D.1: Implantation of Sc in the different substrates for a beam energy of 30keV (dashed lines) and 60keV (full lines).

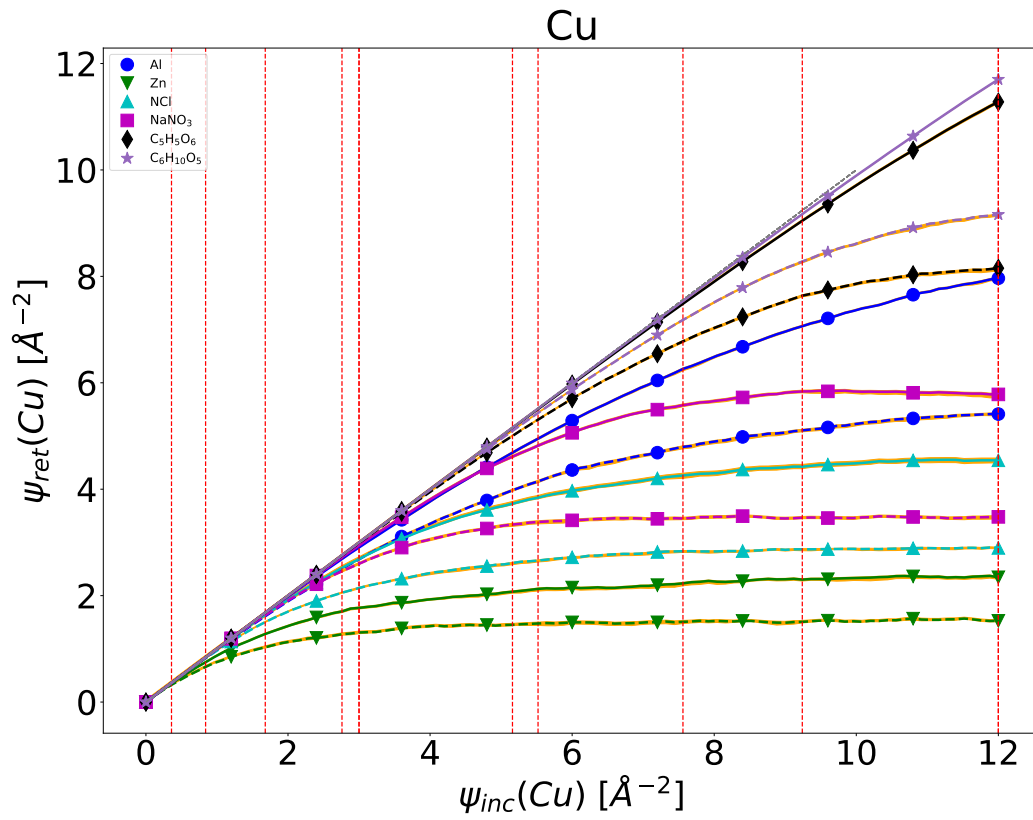


Figure D.2: Implantation of Cu in the different substrates for a beam energy of 30keV (dashed lines) and 60keV (full lines).

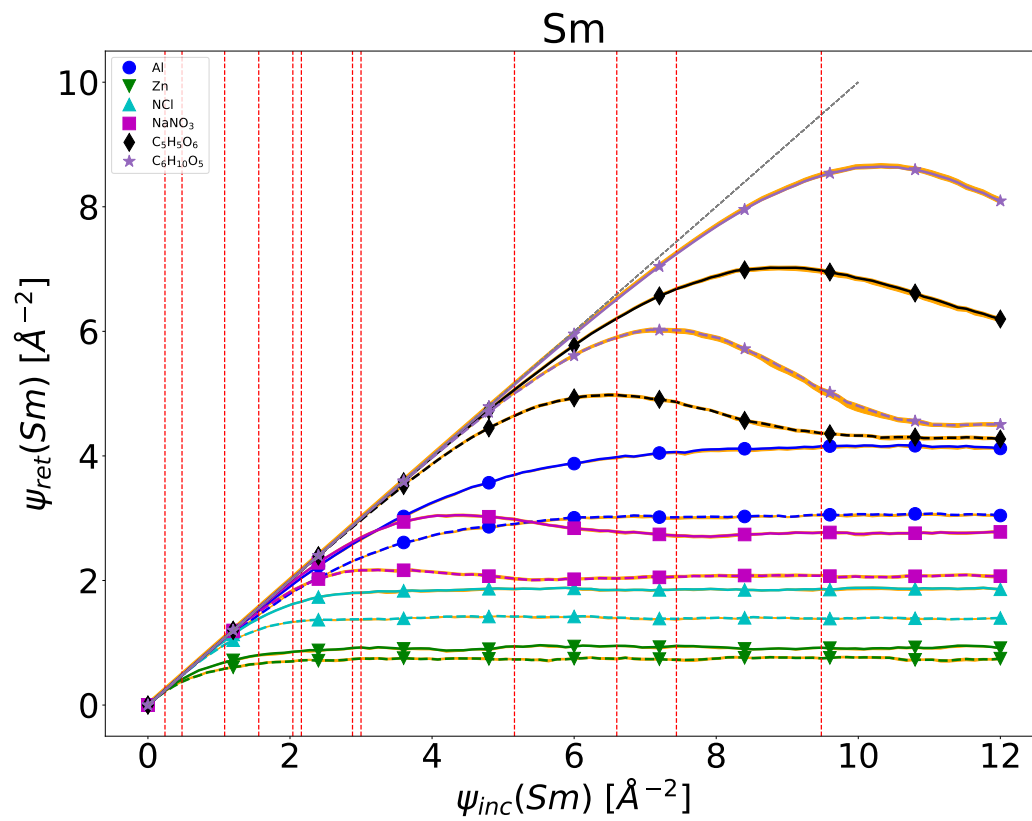


Figure D.3: Implantation of Sm in the different substrates for a beam energy of 30keV (dashed lines) and 60keV (full lines).

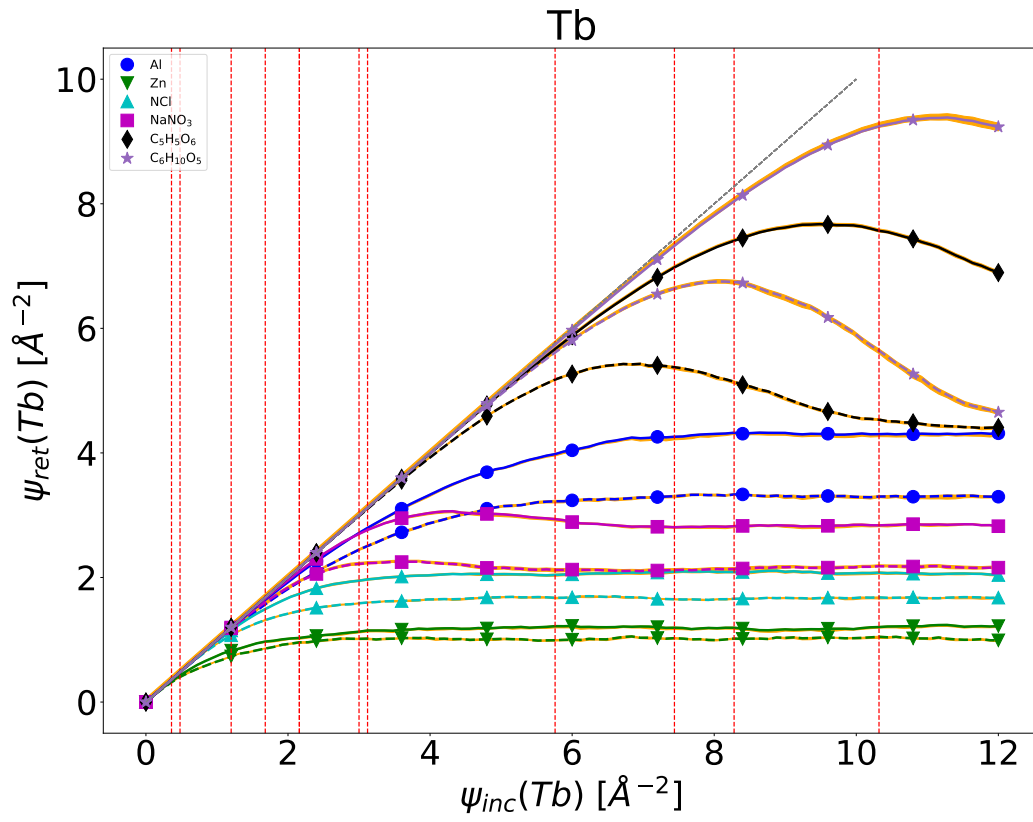


Figure D.4: Implantation of Tb in the different substrates for a beam energy of 30keV (dashed lines) and 60keV (full lines).

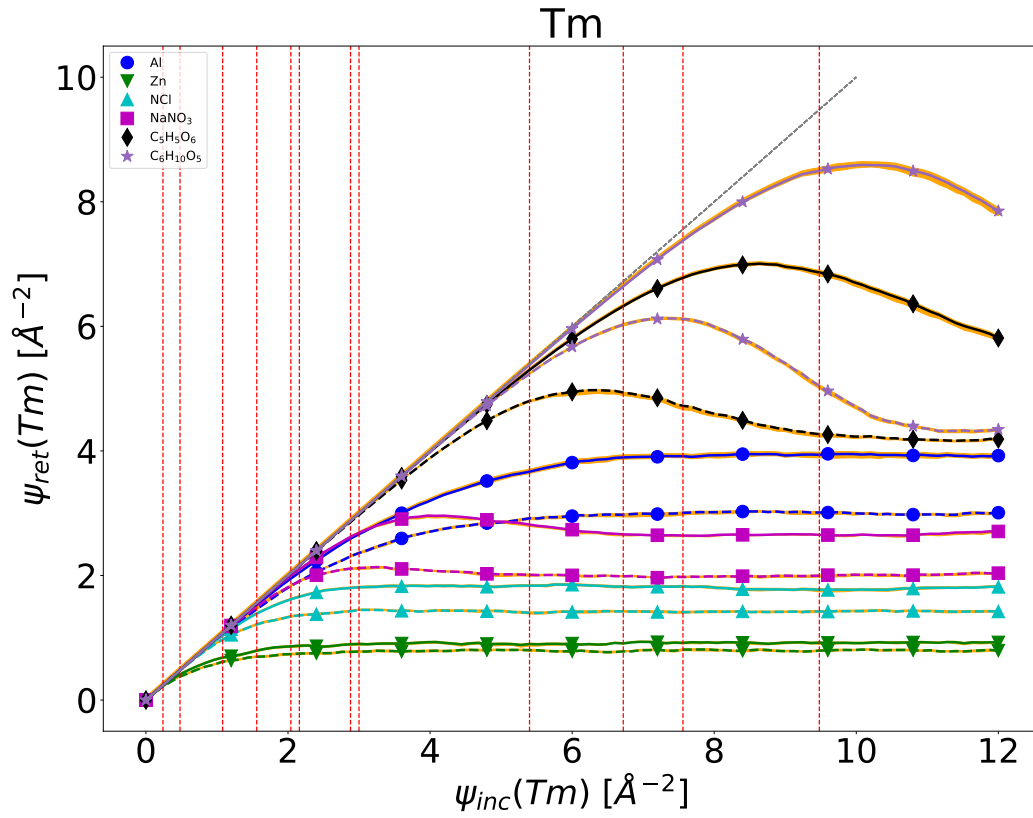


Figure D.5: Implantation of Tm in the different substrates for a beam energy of 30keV (dashed lines) and 60keV (full lines).

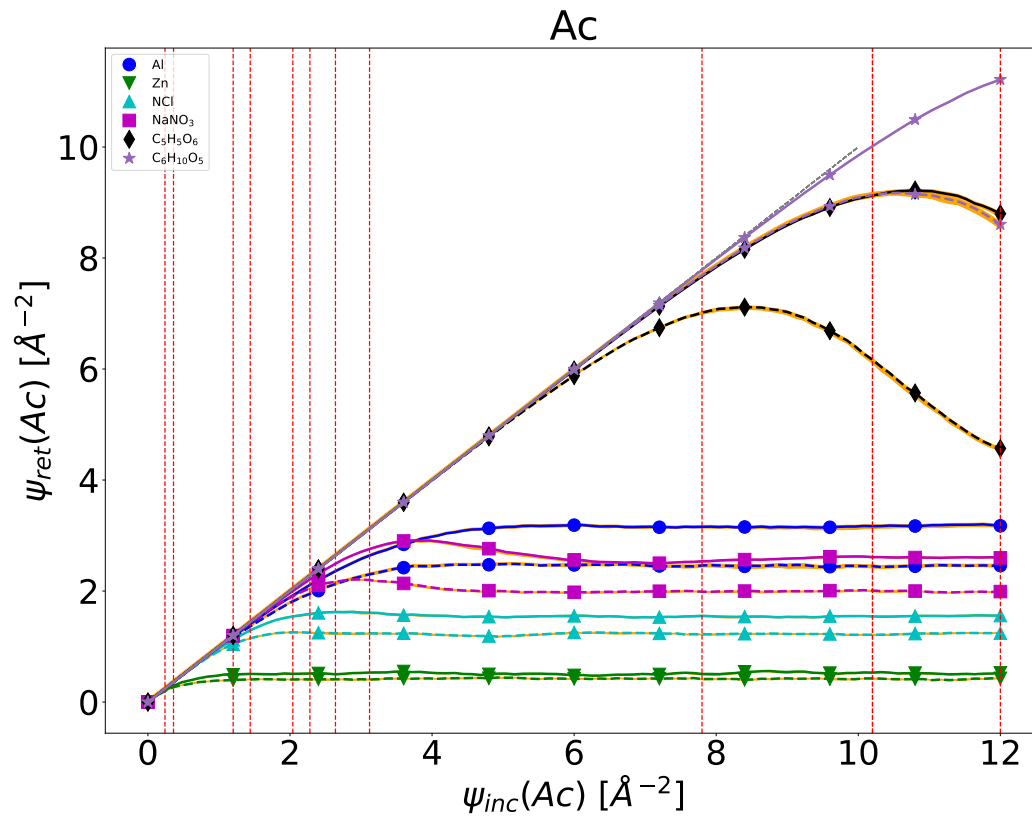


Figure D.6: Implantation of Ac in the different substrates for a beam energy of 30keV (dashed lines) and 60keV (full lines).

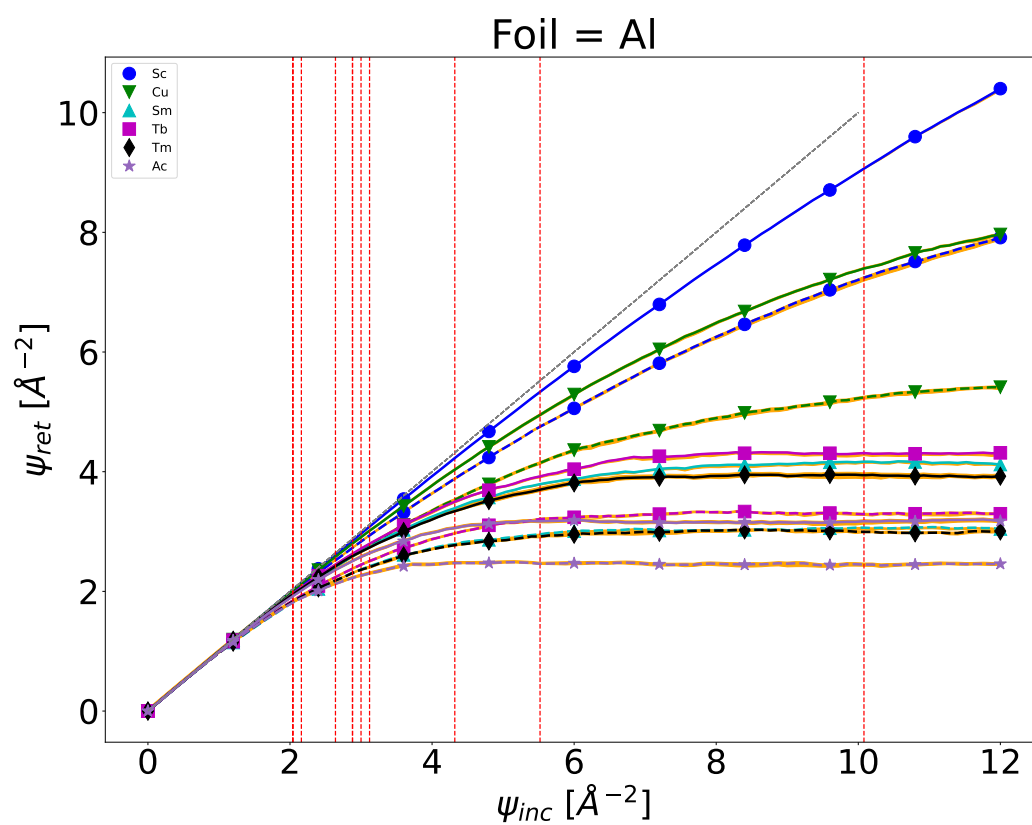


Figure D.7: Implantation of different medical isotopes in Al for a beam energy of 30keV (dashed lines) and 60keV (full lines).

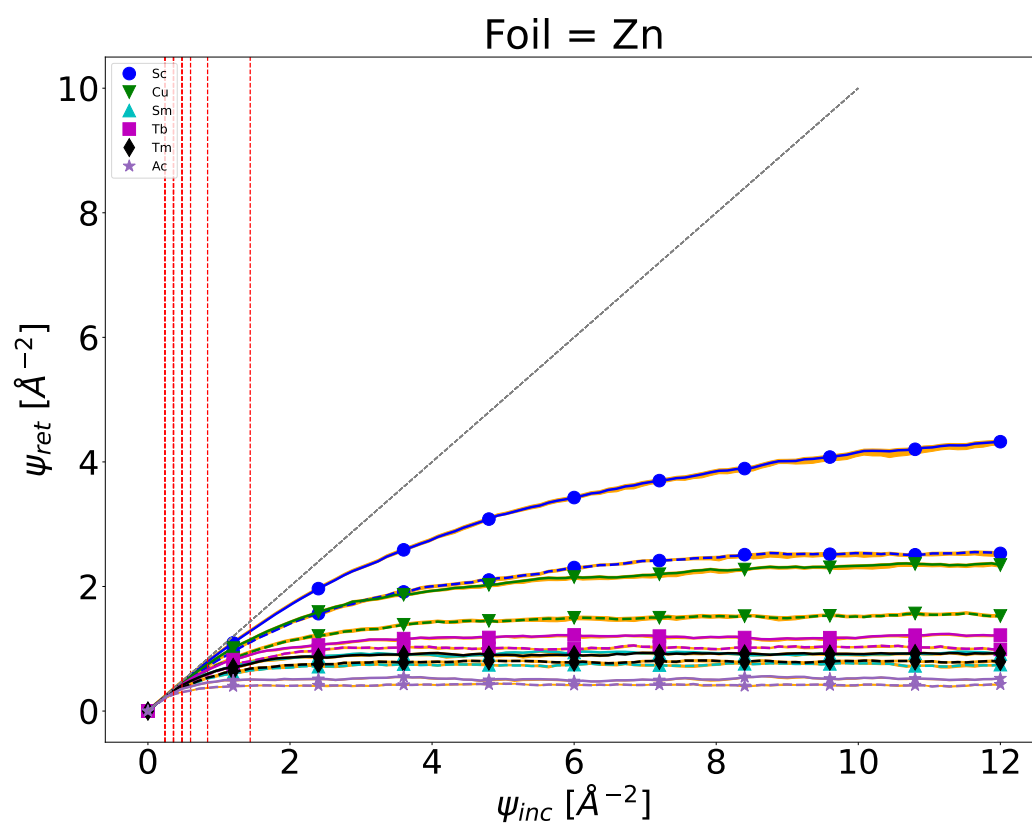


Figure D.8: Implantation of different medical isotopes in Zn for a beam energy of 30keV (dashed lines) and 60keV (full lines).

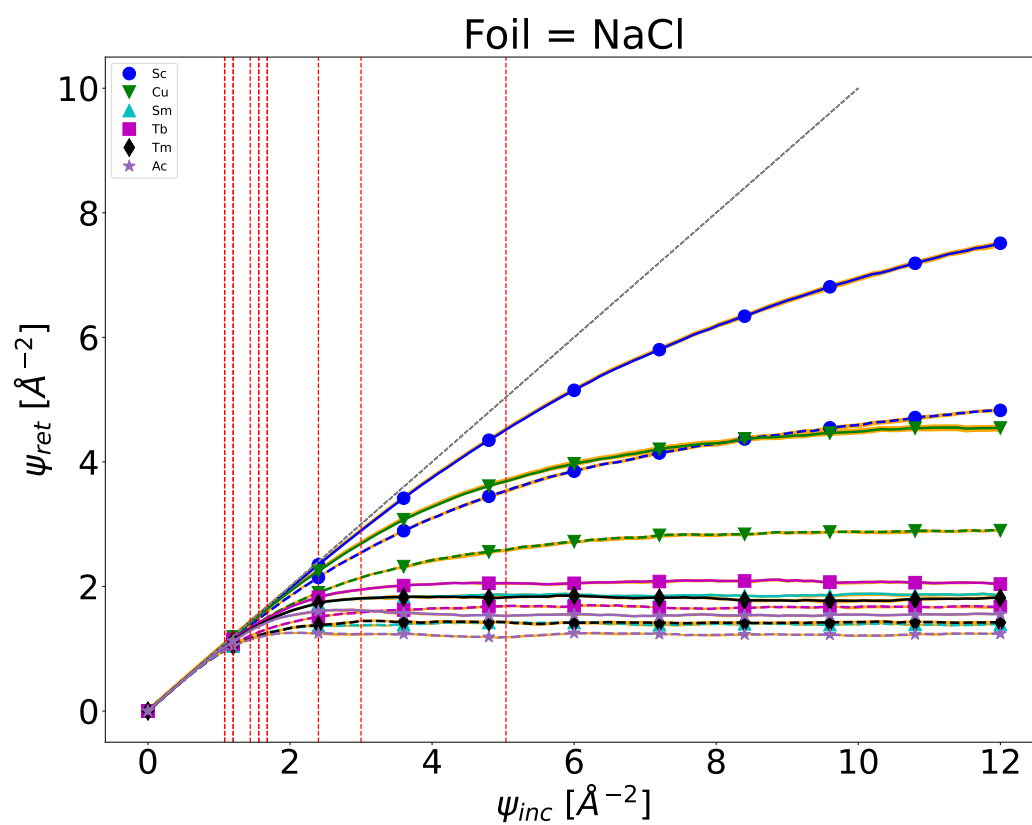


Figure D.9: Implantation of different medical isotopes in NaCl for a beam energy of 30keV (dashed lines) and 60keV (full lines).

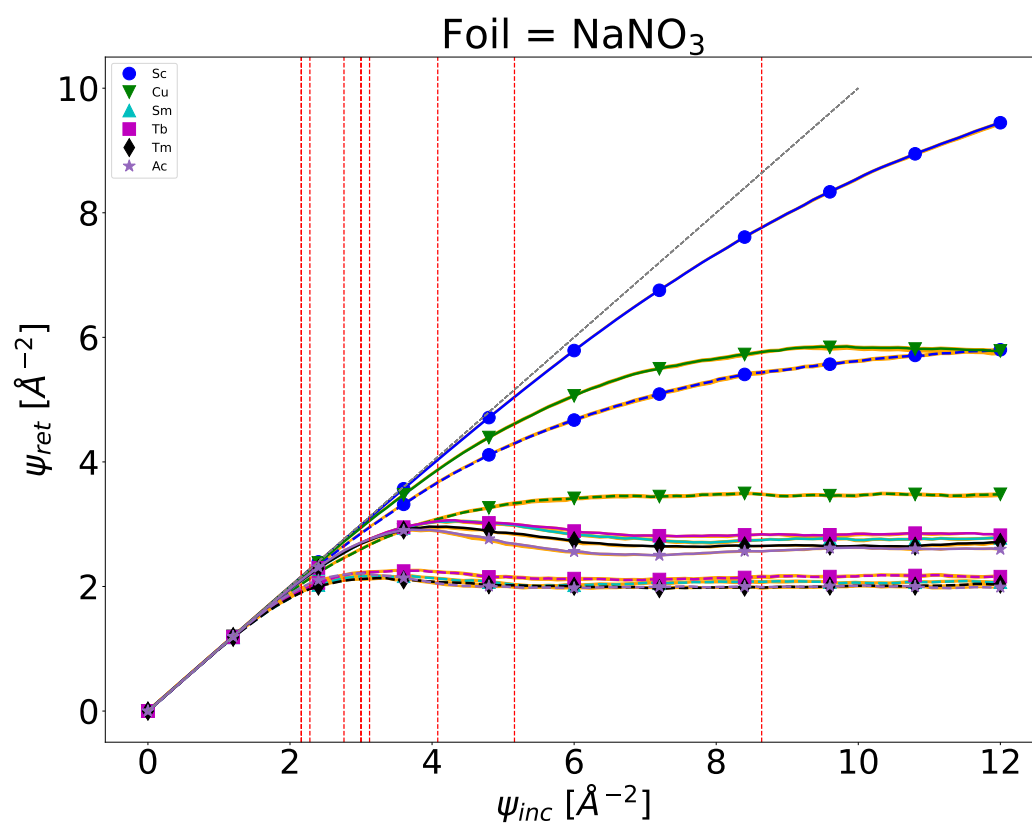


Figure D.10: Implantation of different medical isotopes in NaNO_3 for a beam energy of 30keV (dashed lines) and 60keV (full lines).

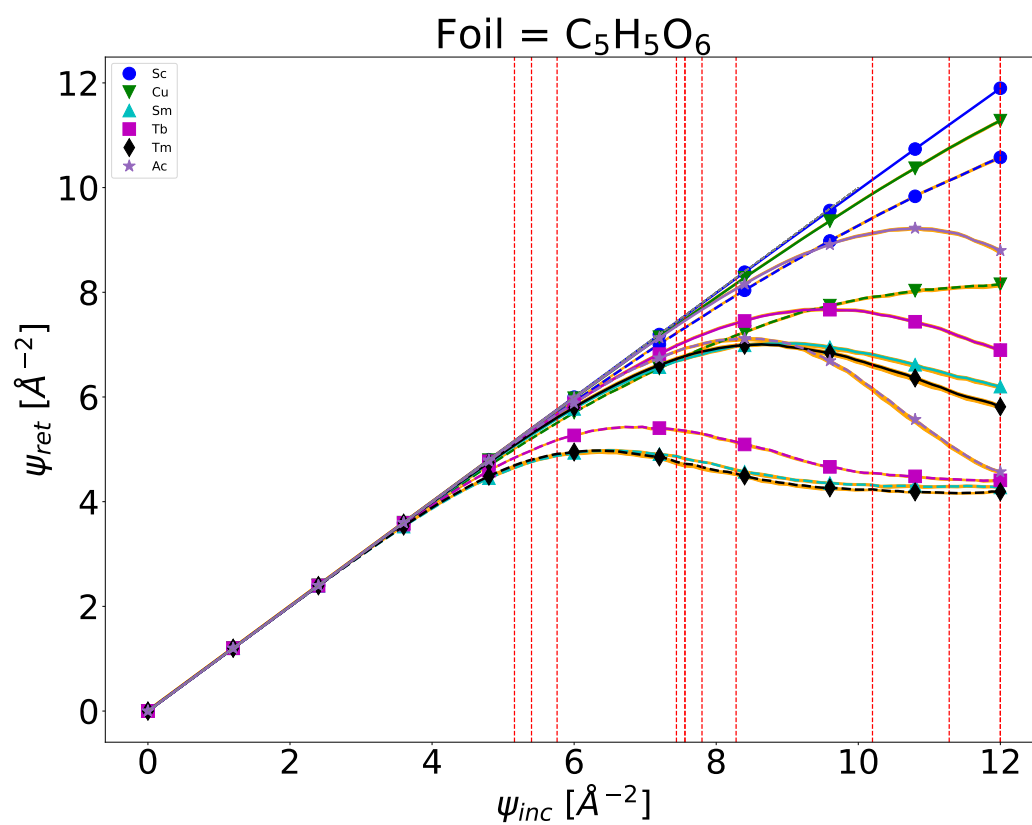


Figure D.11: Implantation of different medical isotopes in dextrates for a beam energy of 30keV (dashed lines) and 60keV (full lines).

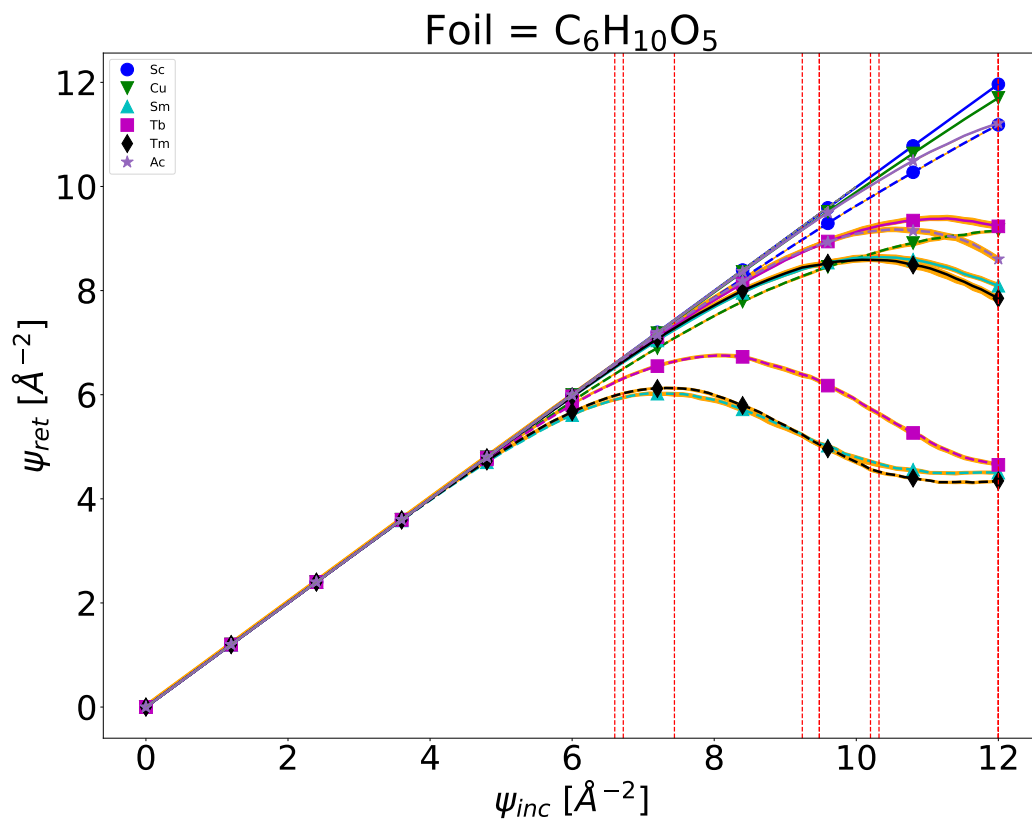


Figure D.12: Implantation of different medical isotopes in cellulose for a beam energy of 30keV (dashed lines) and 60keV (full lines).

E

Recoil-sputtering tables

In this appendix, the tables corresponding to Chapter 10 are provided.

E.1 Energy = 60keV, implantation spot = 0.048cm²

	Sputtering yield of Al	Sputtering yield of Tb
Total	15.8795(133)	0.13880(125)
0-100Å	98.68%	97.62%
100-200Å	0.81%	1.64%
200-300Å	0.36%	0.64%
300-400Å	0.12%	0.10%
400-500Å	0.03%	0.02%
500-600Å	0.003%	0%
600-700Å	0.0001%	0%
700-2000Å	0%	0%

Table E.1: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $1.5\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 60\text{keV}$, implantation spot = 0.048cm).

	Sputtering yield of Al	Sputtering yield of Tb
Total	16.8980(137)	0.32679(191)
0-100Å	98.77%	98.50%
100-200Å	0.78%	1.10%
200-300Å	0.31%	0.32%
300-400Å	0.11%	0.07%
400-500Å	0.02%	0%
500-600Å	0.002%	0%
600-700Å	0%	0%
700-2000Å	0.00007%	0%

Table E.2: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $2.01\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 60\text{keV}$, implantation spot = 0.048cm).

	Sputtering yield of Al	Sputtering yield of Tb
Total	17.5223(140)	0.6942(28)
0-100Å	98.85%	98.88%
100-200Å	0.74%	0.82%
200-300Å	0.30%	0.25%
300-400Å	0.10%	0.04%
400-500Å	0.02%	0.002%
500-600Å	0.002%	0%
600-700Å	0.00006%	0%
700-2000Å	0%	0%

Table E.3: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $2.49\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 60\text{keV}$, implantation spot = 0.048cm).

	Sputtering yield of Al	Sputtering yield of Tb
Total	18.1600(142)	1.7100(44)
0-100Å	98.95%	99.42%
100-200Å	0.71%	0.65%
200-300Å	0.25%	0.13%
300-400Å	0.08%	0.01%
400-500Å	0.01%	0.001%
500-600Å	0.001%	0%
600-700Å	0%	0%
700-2000Å	0%	0%

Table E.4: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $3.48\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 60\text{keV}$, implantation spot = 0.048cm).

	Sputtering yield of Al	Sputtering yield of Tb
Total	17.8500(141)	2.3600(52)
0-100Å	98.94%	99.15%
100-200Å	0.71%	0.57%
200-300Å	0.25%	0.10%
300-400Å	0.08%	0.006%
400-500Å	0.01%	0%
500-600Å	0.001%	0%
600-700Å	0%	0%
700-2000Å	0%	0%

Table E.5: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $4\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 60\text{keV}$, implantation spot = 0.048cm).

Incoming activity [MBq]	Sputtering Yield		Retained activity after implantation of ^{149}Tb		Activity lost due to decay			Activity lost due to recoil-sputtering			Thickness of aluminum layer that is lost [$\text{\AA}$]
	Al	Tb	[MBq]	f_i [%]	[MBq]	f_i [%]	f_r [%]	[MBq]	f_i [%]	f_r [%]	
33811.2	15.8795(133)	0.13880(125)	33761.61	99.85	5251.23	15.53	15.55	123.91(112)	0.3665(34)	0.3670(34)	10.4813(88)
45307	16.8980(137)	0.32679(191)	44966.64	99.25	6994.04	15.44	15.55	388.6(23)	0.8576(51)	0.8641(51)	14.8553(121)
56126.592	17.5223(140)	0.6942(28)	55166.35	98.29	8580.49	15.29	15.55	1012.6(41)	1.8042(72)	1.8356(74)	18.8982(151)
67622.4	18.2562(143)	1.0398(34)	65124.88	96.31	10129.42	14.98	15.55	1790.5(59)	2.6478(87)	2.7494(90)	23.2441(183)
78441.98	18.1600(142)	1.7100(44)	73293.67	93.44	11399.98	14.53	15.55	3314.0(86)	4.2247(109)	4.5215(117)	26.022(21)
90163.2	17.8500(141)	2.3600(52)	80574.34	89.36	12532.40	13.90	15.55	5028.0(111)	5.577(123)	6.2402(138)	28.118(23)

Table E.6: Summarizing table on recoil-sputtering for implantation of ^{149}Tb in aluminum for an implantation energy = 60keV and an implantation spot = 0.048cm². f_i and f_r are the fractions compared to the incoming activity and retained activity after implantation of ^{149}Tb respectively.

E.2 Energy = 100keV, implantation spot = 0.048cm²

Incoming activity [MBq]	Sputtering Yield		Retained activity after implantation of ¹⁴⁹ Tb		Activity lost due to decay			Activity lost due to recoil-sputtering			Thickness of aluminum layer that is lost [Å]
	Al	Tb	[MBq]	f_i [%]	[MBq]	f_i [%]	f_r [%]	[MBq]	f_i [%]	f_r [%]	
33811.2	8.4939(98)	0.01660(42)	33727.80	99.75	5245.97	15.52	15.55	14.80(38)	0.04378(111)	0.04385(111)	5.6008(65)
45307	9.7973(105)	0.04770(73)	45174	99.71	7026.29	15.51	15.55	56.98(88)	0.12576(193)	0.12613(194)	8.6527(93)
56126.592	10.6960(109)	0.10360(108)	55835.82	99.48	8684.61	15.47	15.55	152.95(160)	0.2725(29)	0.2739(29)	11.6759(119)
67622.4	11.5826(113)	0.23881(163)	66853.76	98.86	10401.03	15.38	15.56	422.3(29)	0.6244(43)	0.6316(44)	15.1426(148)
78441.98	12.4379(118)	0.4877(23)	76877.65	98.01	11957.43	15.24	15.55	991.4(47)	1.2896(61)	1.2896(61)	18.6940(178)
90163.2	13.3800(122)	0.6703(28)	86919.58	96.40	13519.33	14.99	15.55	1540.4(65)	1.7086(72)	1.737(75)	22.73(21)

Table E.7: Summarizing table on recoil-sputtering for implantation of ¹⁴⁹Tb in aluminum for an implantation energy = 100keV and an implantation spot = 0.048cm². f_i and f_r are the fractions compared to the incoming activity and retained activity after implantation of ¹⁴⁹Tb respectively.

	Sputtering yield of Al	Sputtering yield of Tb
Total	8.4939(98)	0.01660(42)
0-100Å	98.31%	97.59%
100-200Å	0.79%	1.40%
200-300Å	0.47%	1.01%
300-400Å	0.27%	0.20%
400-500Å	0.12%	0%
500-600Å	0.05%	0%
600-700Å	0.01%	0%
700-800Å	0.0009%	0%
800-900Å	0.0001%	0%
900-2000Å	0%	0%

Table E.8: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $1.5\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 100\text{keV}$, implantation spot = 0.048cm).

	Sputtering yield of Al	Sputtering yield of Tb
Total	9.7973(105)	0.04770(73)
0-100Å	98.39%	97.91%
100-200Å	0.80%	1.28%
200-300Å	0.42%	0.49%
300-400Å	0.23%	0.26%
400-500Å	0.11%	0.05%
500-600Å	0.04%	0.02%
600-700Å	0.009%	0%
700-800Å	0.0009%	0%
800-900Å	0.0001%	0%
900-1000Å	0%	0%
1000-2000Å	0%	0%

Table E.9: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $2.01\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 100\text{keV}$, implantation spot = 0.048cm).

	Sputtering yield of Al	Sputtering yield of Tb
Total	10.6960(109)	0.10360(108)
0-100Å	98.54%	98.26%
100-200Å	0.76%	0.94%
200-300Å	0.39%	0.56%
300-400Å	0.19%	0.22%
400-500Å	0.09%	0.03%
500-600Å	0.03%	0%
600-700Å	0.007%	0%
700-800Å	0.001%	0%
800-900Å	0%	0%
900-1000Å	0%	0%
1000-2000Å	0%	0%

Table E.10: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $2.49\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 100\text{keV}$, implantation spot = 0.048cm).

	Sputtering yield of Al	Sputtering yield of Tb
Total	11.5826(113)	0.23881(163)
0-100Å	98.60%	95.98%
100-200Å	0.75%	0.79%
200-300Å	0.35%	0.25%
300-400Å	0.18%	0.08%
400-500Å	0.08%	0.005%
500-600Å	0.03%	0.005%
600-700Å	0.005%	0.005%
700-800Å	0.001%	0%
800-900Å	0.0001%	0%
900-1000Å	0%	0%
1000-2000Å	0%	0%

Table E.11: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $3\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 100\text{keV}$, implantation spot = 0.048cm).

	Sputtering yield of Al	Sputtering yield of Tb
Total	12.4379(118)	0.4877(23)
0-100Å	98.65%	99.18%
100-200Å	0.75%	0.60%
200-300Å	0.35%	0.17%
300-400Å	0.16%	0.04%
400-500Å	0.07%	0.007%
500-600Å	0.02%	0%
600-700Å	0.003%	0%
700-800Å	0.0009%	0%
800-900Å	0%	0%
900-2000Å	0%	0%

Table E.12: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $3.48\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 100\text{keV}$, implantation spot = 0.048cm).

	Sputtering yield of Al	Sputtering yield of Tb
Total	13.3800(122)	0.6703(28)
0-100Å	98.73%	99.10%
100-200Å	0.72%	0.67%
200-300Å	0.31%	0.18%
300-400Å	0.15%	0.05%
400-500Å	0.06%	0.005%
500-600Å	0.02%	0%
600-700Å	0.004%	0%
700-800Å	0.0005%	0%
800-900Å	0.0002%	0%
900-2000Å	0%	0%

Table E.13: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $4\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 100\text{keV}$, implantation spot = 0.048cm).

E.3 Energy = 60keV, implantation spot = 0.192cm²

Incoming activity [MBq]	Sputtering Yield		Retained activity after implantation of ¹⁴⁹ Tb		Activity lost due to decay			Activity lost due to recoil-sputtering			Thickness of aluminum layer that is lost [Å]
	Al	Tb	[MBq]	f_i [%]	[MBq]	f_i [%]	f_r [%]	[MBq]	f_i [%]	f_r [%]	
33811.2	13.3000(122)	0.001679(137)	33811.20	100	5258.94	15.55	15.55	1.501(123)	0.00444(37)	0.00444(37)	2.1979(21)
45307	13.8300(124)	0.003300(192)	45305.20	99.996	7046.70	15.55	15.55	3.95(24)	0.00873(51)	0.00873(51)	3.0624(28)
56126.592	14.0040(125)	0.00570(26)	56122.99	99.99	8729.28	15.55	15.55	8.46(39)	0.01507(69)	0.01507(69)	3.8414(35)
67622.4	14.1360(126)	0.01328(39)	67613.01	99.99	10513.41	15.55	15.55	23.74(70)	0.03510(104)	0.03510(104)	4.6701(42)
78441.98	14.9436(129)	0.02970(58)	78427.56	99.98	12198.50	15.55	15.55	61.59(121)	0.07852(154)	0.07853(154)	5.7282(50)
90163.2	15.270(131)	0.04400(70)	90131.64	99.97	14018.93	15.55	15.55	104.86(167)	0.116302(186)	0.116343(186)	6.726(58)

Table E.14: Summarizing table on recoil-sputtering for implantation of ¹⁴⁹Tb in aluminum for an implantation energy = 60keV and an implantation spot = 0.192cm². f_i and f_r are the fractions compared to the incoming activity and retained activity after implantation of ¹⁴⁹Tb respectively.

	Sputtering yield of Al	Sputtering yield of Tb
Total	13.3000(122)	0.001679(137)
0-100Å	98.42%	75.46%
100-200Å	0.8624%	9.9464%
200-300Å	0.49%	11.26%
300-400Å	0.18%	3.36%
400-500Å	0.04%	0%
500-600Å	0.0039%	0%
600-700Å	0.0001%	0%
700-2000Å	0%	0%

Table E.15: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $0.375\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 60\text{keV}$, implantation spot = 0.192cm).

	Sputtering yield of Al	Sputtering yield of Tb
Total	13.8300(124)	0.003300(192)
0-100Å	98.48%	85.18%
100-200Å	0.85%	7.06%
200-300Å	0.46%	5.73%
300-400Å	0.17%	2.03%
400-500Å	0.03%	0%
500-600Å	0.03%	0%
600-700Å	0%	0%
700-2000Å	0%	0%

Table E.16: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $0.5025\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 60\text{keV}$, implantation spot = 0.192cm).

	Sputtering yield of Al	Sputtering yield of Tb
Total	14.0040(125)	0.00570(26)
0-100Å	98.47%	85.96%
100-200Å	0.87%	7.21%
200-300Å	0.46%	4.68%
300-400Å	0.16%	1.56%
400-500Å	0.03%	0%
500-600Å	0.003%	0%
600-700Å	0.0.00008%	0%
700-2000Å	0%	0%

Table E.17: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $0.6225\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 60\text{keV}$, implantation spot = 0.192cm).

	Sputtering yield of Al	Sputtering yield of Tb
Total	14.1360(126)	0.01328(39)
0-100Å	98.47%	92.63%
100-200Å	0.85%	4.19%
200-300Å	0.44%	2.93%
300-400Å	0.17%	0.24%
400-500Å	0.04%	0%
500-600Å	0.04%	0%
600-700Å	0.0002%	0%
700-2000Å	0%	0%

Table E.18: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $0.75\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 60\text{keV}$, implantation spot = 0.192cm).

	Sputtering yield of Al	Sputtering yield of Tb
Total	14.9436(129)	0.02970(58)
0-100Å	98.57%	94.28%
100-200Å	0.85%	3.44%
200-300Å	0.40%	1.95%
300-400Å	0.14%	0.37%
400-500Å	0.03%	0.07%
500-600Å	0.003%	0%
600-700Å	0.0002%	0%
700-2000Å	0%	0%

Table E.19: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $0.87\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 60\text{keV}$, implantation spot = 0.192cm).

	Sputtering yield of Al	Sputtering yield of Tb
Total	15.270(131)	0.04400(70)
0-100Å	98.62%	96.14%
100-200Å	0.84%	2.40%
200-300Å	0.41%	1.54%
300-400Å	0.14%	0.03%
400-500Å	0.02%	0%
500-600Å	0.003%	0%
600-700Å	0.0001%	0%
700-2000Å	0%	0%

Table E.20: Overview of the sputtering yield of aluminum and terbium for the different layers of the target (fluence = $1\text{\AA}^{-2}$, $E(^{149}\text{Tb}) = 60\text{keV}$, implantation spot = 0.192cm).

F

Recoil-sputtering for ^{225}Ac

As was discussed in Chapter 10, recoil-sputtering is needs to be considered for α -decaying nuclei. Apart from ^{149}Tb , another promising alpha-decaying isotope for medical applications is ^{225}Ac . Four high energy α -particles are emitted in short succession in the decay chain of ^{225}Ac as can be seen in Figure F.1. Since 2020, several samples of ^{225}Ac (on zinc or aluminum foils) have been collected. Two of these samples were analyzed with the method of alpha decay recoil spectroscopy, with the main goal of determining the ^{227}Ac activity through the activity of its recoil daughters. During these campaigns, traces of ^{225}Ac were measured even when the sample was removed, indicating that recoil-sputtering had occurred. This appendix presents the method used to predict the amount of recoil-sputtering for an ^{225}Ac sample using TRIDYN and SRIM. The result from this simulation is compared to the experimentally observed recoil sputtered activity of ^{225}Ac for the collection of December 2022.

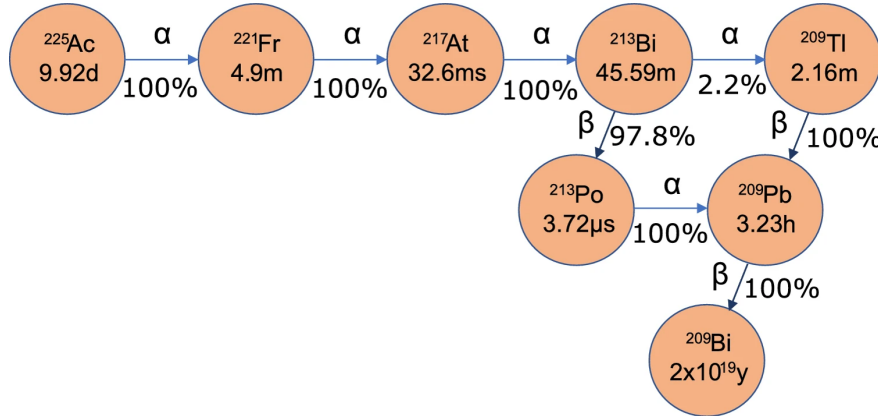


Figure F.1: Decay chain of ^{225}Ac . (figure retrieved from [67])

F.1 Experiment

In this section, a short introduction to the measurements that were performed on the actinium sample from the collection of December 2022 will be given.

Following the collection of ^{225}Ac on zinc foils at MEDICIS, the foils were shipped to KU Leuven and analyzed. Alpha-spectroscopy on the ^{225}Ac -implanted zinc foil has

been performed by the use of the ASET set-up. A schematic representation of the set-up is presented in Figure F.2. In the first part of the measurement, the foil was placed in front of the detector of the ASET set-up (a fully depleted PIPS detector with area 300mm^2) and the α -decays originating from the ^{225}Ac chain were measured for 21 days. In the α -decay, the daughter nucleus obtains recoil energy, thus some recoil daughters, within an emission cone whose angle is defined by the daughter's range in the material, are implanted on the detector. Additionally, ^{225}Ac can be implanted on the detector via recoil-sputtering. Recoil-sputtering occurs when a recoiling daughter transfers its energy to an implanted ^{225}Ac which is subsequently ejected from the foil.

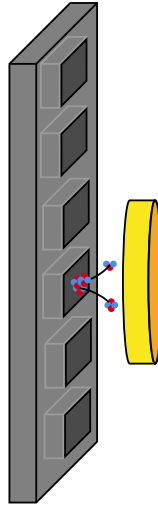


Figure F.2: ASET setup, the sample is mounted on the ladder and the detector is indicated in yellow.

Next, the ladder with the foil is removed and no more α -particles coming from the foil can be detected. However, the daughters of ^{225}Ac that were implanted in the detector cause a non-zero α -count rate. Thus, a measurement of the α -particles after foil removal can be conducted for investigation into the activity that was implanted in the foil. The results are indicated in Figure F.3, here the number of α -counts is plotted as a function of the time after removal. Both ^{221}Fr and ^{217}At decay with the decay constant of ^{221}Fr as the half-life of ^{217}At is considerably shorter than that of ^{221}Fr [67]. For the ^{213}Po decay, the α -count rate decays with the half-life of ^{213}Bi . In the absence of recoil-sputtering, all three curves would decay towards 0. However, this is not what is observed: due to the recoil-sputtering, ^{225}Ac particles are implanted in the detector and these are responsible for the plateauing of the count rate at approximately 0.4 days: the ^{225}Ac particles have a half-life of 9.92 days, thus leading to the emission of α -particles in the decay of ^{221}Fr , ^{217}At and ^{213}Po with a characteristic time that is equal to the half-life of ^{225}Ac [67].

This decay behavior can now be fitted and extrapolated back towards the time at which the foil was removed (indicated in red in Figure F.3). From this extrapolation, the count rate coming from ^{221}Fr , ^{217}At or ^{213}Po due to the recoil sputtered ^{225}Ac at the time the foil was removed is obtained. The count rate coming from ^{225}Ac for when the foil was removed is obtained by taking into account the different geometric efficiencies for each

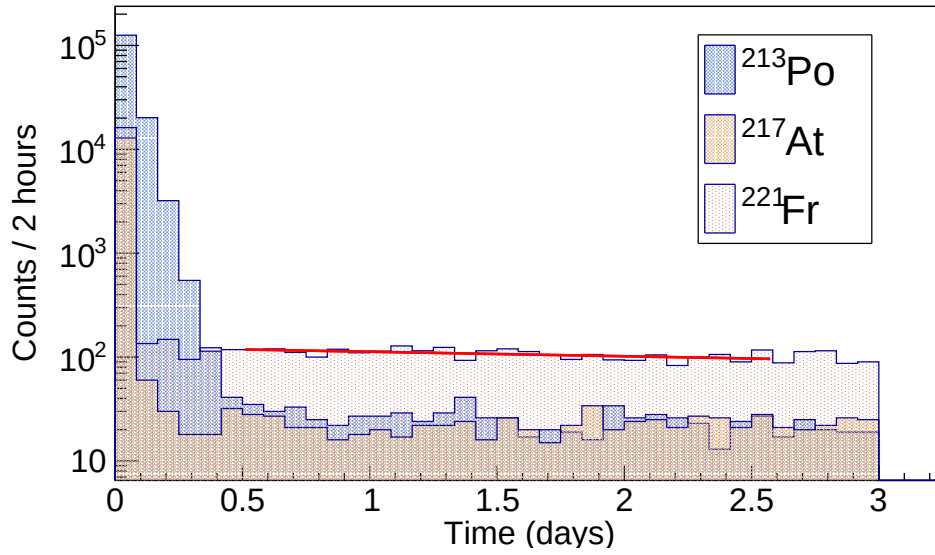


Figure F.3: The number of α counts, origination from the α -decays of the daughters of ^{225}Ac , as a function of the time after removal of the foil. (Image courtesy of Jake Johnson)

isotope following the redistribution due to recoils following α -decays. Therefore, SRIM simulations were performed to obtain the geometric efficiency. It was concluded that for a foil with an activity of 52.69(199)kBq, 0.0869(191)Bq was recoil-sputtered during the measurement at the time of foil-removal (calculated from the α -particles originating from the francium decay). In this value, the emission distribution of the actinium from the foil onto the detector, the redistribution of ^{221}Fr due to α -decay recoil, and the geometric efficiency for detection of ^{221}Fr alpha particles were accounted for, as was discussed in Chapter 10.

Even though the recoil-sputtered fraction is only 0.0008% of the total activity at that time, for collections with more particles, this effect can become more significant.

F.2 Simulation

In this section, the different steps that are performed to simulate the recoil-sputtering during the α -decay spectroscopy are discussed. Due to the presence of multiple α -decays, it is necessary to perform multiple SRIM simulations subsequently to the TRIDYN implantation simulation. Several approximations are made:

- The α -decaying branch of ^{213}Bi is neglected, as well as the recoil obtained after β decay, as almost all energy is transferred to the β particles. ^{209}Bi has a long half-life ($t_{1/2} = 2 \cdot 10^{19}\text{y}$), therefore its decay will be neglected.
- The distribution of the foil is static, in other words, the depth profile is constant throughout the simulations and equal to the depth profile after the MEDICIS collection.
- The fluence of actinium was derived from the beam current data and the spectroscopy data.

The first step in the simulation consists of the implantation simulation for ^{225}Ac , using TRIDYN. Implantation was performed in a zinc coated gold foil with the high voltage configured on 50kV and a beam consisting of ^{225}Ac and ^{139}La . The activity that was implanted during the MEDICIS collection is 52.69kBq, indicating that $6.5147 \cdot 10^{10}$ ^{225}Ac particles are impinging. The beam current on the collection foil was monitored during all times of the collection. The total number of particles implanted on the foil was calculated by integrating the beam current over the collection time and yielded that $9.095 \cdot 10^{10}$ particles were implanted. The area of the beam spot was calculated using a clipped integral to determine the mean radius. This resulted in an implantation spot of $(1.02)^2 \cdot \pi \cdot 10^{14} \text{Å}^2$ and thus the simulation was performed for a total incoming fluence equal to 0.00027Å^{-2} with 72% of the beam corresponding to ^{225}Ac ($E = 50\text{keV}$) and 28% of the beam corresponding to La ($E = 50\text{keV}$).

From the initial depth profile of ^{225}Ac , obtained using TRIDYN, all subsequent alpha decays in the ^{225}Ac chain are simulated. The first SRIM simulation is performed for the recoiling ^{221}Fr , the corresponding output is used in order to perform a simulation for the recoiling ^{217}At . Again, the output is used to simulate the recoiling ^{213}Bi . The output from simulating the ^{213}Bi is now used to generate the ^{209}Pb recoils, here the recoil obtained from the β decay is neglected. The total sputtering yield can then be calculated as:

$$Y_{\text{tot}}(\text{Ac}) = Y_{^{221}\text{Fr}, \text{recoil}}(\text{Ac}) + Y_{^{217}\text{At}, \text{recoil}}(\text{Ac}) + Y_{^{213}\text{Bi}, \text{recoil}}(\text{Ac}) + 0.978Y_{^{209}\text{Pb}, \text{recoil}}(\text{Ac}), \quad (\text{F.1})$$

where $Y_{X, \text{recoil}}(\text{Ac})$ is the sputtering yield of ^{225}Ac due to the recoiling X. The recoil-sputtering yield for ^{225}Ac is now used to obtain a prediction for the activity of the recoil-sputtered particles on the detector at the moment when the foil is removed from ASET:

$$\begin{aligned} A(\text{recoil-sputtered}, t^*) &= \lambda_{^{225}\text{Ac}} \cdot (\text{Number of } ^{225}\text{Ac} \text{ particles on the detector}) \\ &= \lambda_{^{225}\text{Ac}} \int_0^{t^*} \frac{\# \text{ } ^{225}\text{Ac} \text{ decays}}{\text{sec}} Y_{\text{tot}}(\text{Ac}) e^{-\lambda_{^{225}\text{Ac}}(t^*-t)} dt \\ &= \lambda_{^{225}\text{Ac}} \int_0^{t^*} A_0 e^{-\lambda_{^{225}\text{Ac}} t} Y_{\text{tot}}(\text{Ac}) e^{-\lambda_{^{225}\text{Ac}}(t^*-t)} dt \\ &= \lambda_{^{225}\text{Ac}} \int_0^{t^*} A_0 Y_{\text{tot}}(\text{Ac}) e^{-\lambda_{^{225}\text{Ac}} t^*} dt \\ &= \lambda_{^{225}\text{Ac}} Y_{\text{tot}}(\text{Ac}) A_0 t^* e^{-\lambda_{^{225}\text{Ac}} t^*} \end{aligned} \quad (\text{F.2})$$

where $\lambda_{^{225}\text{Ac}}$ is the decay constant of ^{225}Ac , $Y_{\text{tot}}(\text{Ac})$ the recoil-sputtering yield, A_0 the activity at the start of the α -spectroscopy and t^* the moment at which the foil was removed.

The sputtering yield that is obtained from the simulations is equal to:

$$\begin{aligned} Y_{\text{tot}}(\text{Ac}) &= Y_{^{221}\text{Fr}, \text{recoil}}(\text{Ac}) + Y_{^{217}\text{At}, \text{recoil}}(\text{Ac}) + Y_{^{213}\text{Bi}, \text{recoil}}(\text{Ac}) + 0.978Y_{^{209}\text{Pb}, \text{recoil}}(\text{Ac}) \\ &= 0.000089(32) + 0.000131(42) + 0.000171(52) + 0.978 \cdot 0.000127(48) \\ &= 0.000515(88). \end{aligned} \quad (\text{F.3})$$

And thus the activity corresponding to the recoil-sputtered ^{225}Ac is equal to 0.225(33)Bq. Considering the numerous approximations that were made in the simulation, this is in reasonable agreement with the measured value of 0.0869(191)Bq.

Contributions

Jake Johnson:

- The α -decay spectroscopy
- Determination of the count rate coming from ^{221}Fr due to the recoil sputtered ^{225}Ac
- SRIM simulations for the geometrical efficiency

Marie Deseyn:

- TRIDYN simulations for the initial distribution profile of ^{225}Ac
- SRIM simulations for the recoiling ^{221}Fr , ^{217}At , ^{213}Bi and ^{209}Bi to obtain the recoil-sputtered activity of ^{225}Ac



Poster on sputtering

To convey knowledge about self-sputtering and recoil-sputtering, a poster was made that summarizes the main conclusions of the analysis of self-sputtering and recoil-sputtering. This poster was presented at the LISA (Laser Ionization and Spectroscopy of Actinides) conference on May 22 and May 23, 2023.

Is self-sputtering worth considering for isotope collections?

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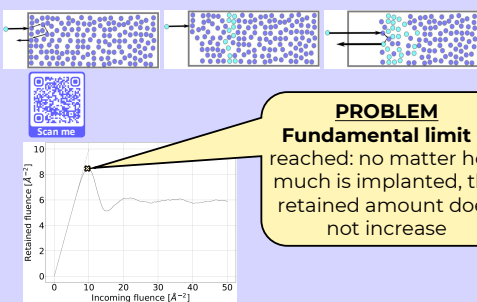
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Context

- For radionuclide collections the goal is to **maximize the amount of collected radioisotopes**
- But, during CERN-MEDICIS collections **self-sputtering was observed** which limits the amount of collected radioisotopes
- Aim of this work: investigate self-sputtering for radionuclide collections

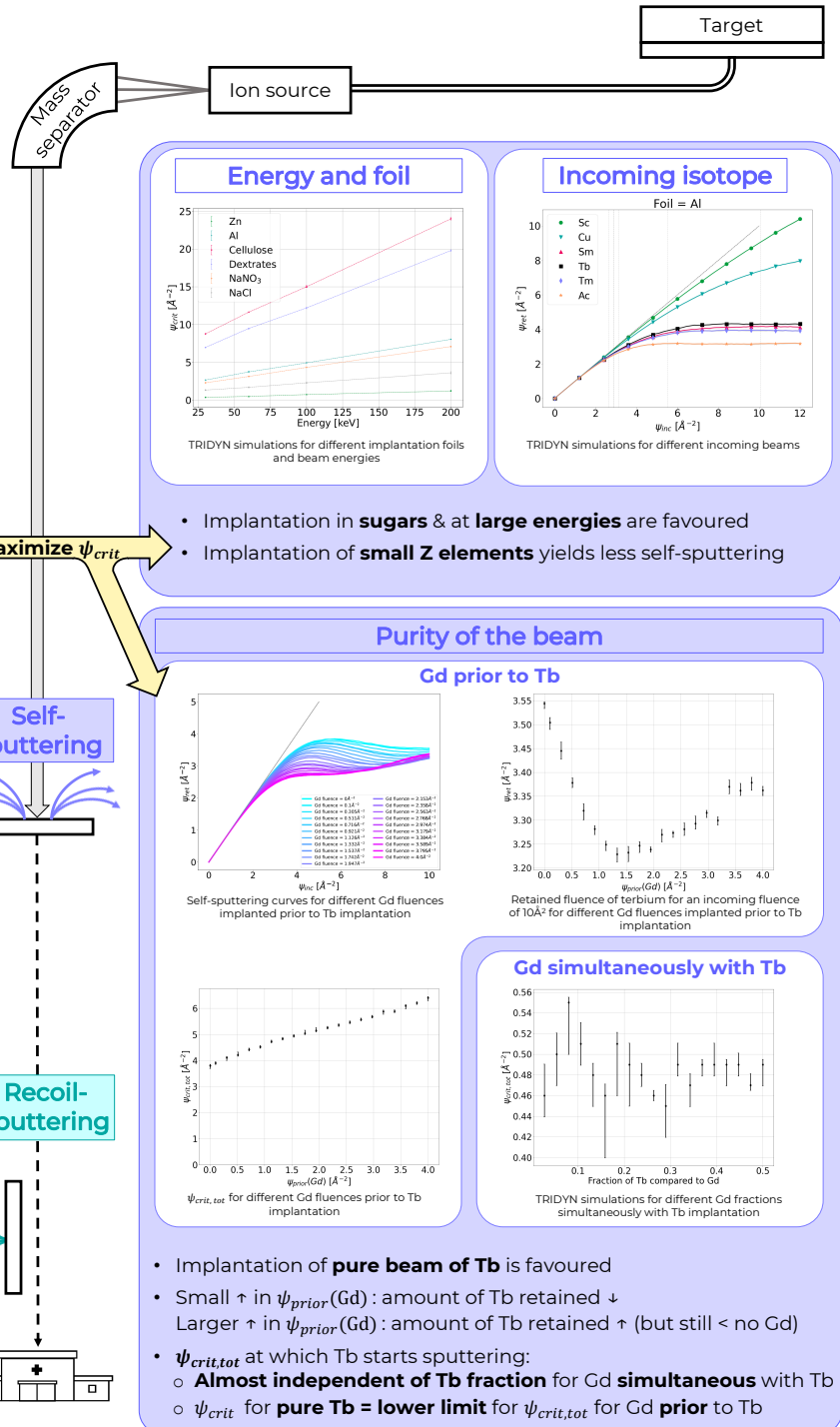
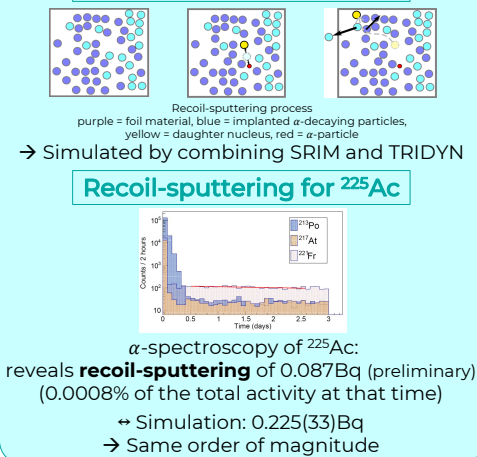
General principle



TRIDYN

- Monte Carlo based simulation package [1, 2]
- Dynamic** changes to the target $\leftrightarrow$ SRIM [3]
- Measure for when self-sputtering becomes significant: $\psi_{crit}: \psi_{ret} < 0.9\psi_{inc}$

General principle



Conclusion and outlook

- $\psi_{crit}(Al) > \psi_{crit}(Zn)$; $\psi_{crit}(metal) \approx \psi_{crit}(salt)$
- $\rightarrow$ **Experimental confirmation** of TRIDYN simulations is needed (using Yb implantations in Zn and Al + RBS and plans for sugar foils)
- Total fluence** can be used as indication for when self-sputtering starts to become significant
- Recoil-sputtering** was experimentally confirmed

References

- [1] Tridyn application examples - helmholtz-zentrum dresden-rossendorf, HZDR. <https://www.hzdr.de/db/Cms?pNid=0&pOid=65033>.
- [2] W. Möller and W. Eckstein. Tridyn—A TRIM simulation code including dynamic composition changes. Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms, 2(1-3):814–818, 1984. doi:10.1016/0168-583X(84)90321-5.
- [3] R. Stadlmayr et al. Sputtering of nanostructured tungsten and comparison to modelling with TRIDYN. Journal of Nuclear Materials, 532:152019, 2020. doi:10.1016/j.jnucmat.2020.152019.



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