Assesment of the isotopic inventory built-up in ISOL targets operated with 100-MeV protons and the migration of the volatile species in the ISOL system.

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Abstract

ISOL (Isotopic Seperation On-Line) is a technique for generating, extracting and separating specific isotopes while the production process is ongoing (online). In this thesis the isotopic built-up in different volumes in the ISOL-system by means of migration of the volatile and non-volatile isotopes created through the irradiation of a target with 100 MeV protons will be assessed through a Python3 model. The model’s purpose is to predict the built-up of specific isotopes and to determine the risk these isotopes pose. Due to the absence of sensible data regarding sticking behaviour of elements on different substrates this data has been estimated using the vapour pressures. The model shows that the radioactivity within the system is mostly dependent on the short-lived isotopes and to a lesser degree on the medium- and long-lived isotopes. This is an expected result caused by the near constant production of these isotopes which will lead to those being in the system for an extended period of time because of a reached equilibrium. The mass separator slits will receive a fraction of the produced isotopes and as such will become highly radioactive. In order to safely dispose of the slits care should be taken. Using the legal limit of 20 mSv per year and the initial dose-rate to be below 10µSv/h, the first year of interim storage will result in an absorbed dose of 4.4 mSv for a worker standing 5 metre from the storage pool. In case of an accident involving a leak from the casserole with adequate warning systems and handling the dose to personnel could be limited to 0.9 mSv. More research to determine the adsorption enthalpies of elements on the relevant materials is advised for future continued research.
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1 Introduction

Both in sciences and fundamental nuclear physics there is a high demand for radioactive particles. In the former this is often related to medical isotopes which can be used as tracer or for treatments of different cancers. In nuclear physics radioactive nuclei far from the stability line are in high demand as they might unveil the true nature of material, or even the universe itself. A possible method is by application of the ISOL-process on a target material under proton irradiation. ISOL systems are currently in high demand[1].

1.1 What is ISOL?

ISOL (Isotopic Separation On-Line) is a process by which newly created isotopes are extracted and separated during operation. The creation of these isotopes (many of which are far from the stability line and thus radioactive) is done by bombarding a target with charged particles as seen in figure[1]. The isotopes are then extracted from the target, typically through high temperature diffusion (in the target material) and molecular flow (in the target container). The constant flux of the primary particle beam ensures a constant flux of isotopes. When the particles leave the target container they experience a chance to become ionised after which they can be mass-separated. The complete system of extraction of these isotopes during the constant particle impact on the target is called online extraction, hence the name Isotopic Separation On-Line.

![Schematic process by which ISOL works. Seen here is the formation, molecular flow, ionisation and separation of isotopes.][2]

1.2 ISOL@MYRRHA

MYRRHA (Multi-purpose hYbrid Research Reactor for High-tech Applications) will be an accelerator driven nuclear reactor cooled with lead-bismuth eutectic. The reactor, operational in 2037[3], will be driven by a 4-mA, 600-MeV proton

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[1]: [Link to source]
[2]: [Link to source]
[3]: [Link to source]
linear accelerator (LINAC). The first phase of the project, a 100-MeV LINAC should become operational in 2026, a receiving proton target facility in 2027. The latter will contain an ISOL-facility, a first step towards ISOL@MYRRHA which intends to use a fraction of the proton beam on an ISOL target for production of high purity radioactive ion beams (RIBs)[4]. In the first phase ISOL@MYRRHA will be running at a maximum of 500μA proton current. From 2037 onwards ISOL@MYRRHA will use up to 5% (200 μA) of the 600-MeV proton beam while working in parallel with the MYRRHA reactor.

1.3 The goal of this work

Due to the high energy interaction between the proton and the target material radioactive nuclei far from the stability line are formed. Due to the the poor nuclear stability these nuclei will decay quickly. The aim of the current work is to model the transport of the radioactive volatile and non-volatile species formed using the 100-MeV protons and to determine the built-up of the radioactive isotopes in the different volumes of the ISOL-system. Being able to determine the concentration of radioactive isotopes at different positions in the system is important to determine the risk in case of an accident and can given an indication for the extraction of certain isotopes and thus the extraction efficiency of this isotope. In order to build an effective model the ISOL-system is simplified to a set of volumes which will be considered to contain the isotopes. These volumes will be discussed here shortly and a schematic representation can be seen in figure 2.

1. discs
Thorium carbide sheets in the target will be impacted with 100-MeV protons from the linear accelerator. The target material can be swapped for other materials. Thorium carbide was used in this work as a worst case scenario.

2. Target container
The target container will contain the discs and together they form the target. The whole target will be heated to 2200 K in order to release the volatile isotopes from the discs to the target volume where they will collide with the heated walls until they reach the ion source.

3. Ion source
The ion source is a heated tantalum tube in which elements have different chances to get ionised on impact due to surface ionisation. Non-ionised particles will be deposited in the casserole and front-end after diffusing out of the ion source.

4. Casserole
The casserole will contain the target and will have holes towards the front-end which will be used to evacuate neutrals that ended up in the casserole to the front-end.
5. Front-end
The ions leaving the ion source will be transported through the front-end. The front-end is the vacuum volume containing the mass-separator, collimators and the optics used to shape the RIB. The volume contains pumps in order to move the contaminants to the decay tank volume. A fraction of the neutrals released from the ion source will be deposited in the front-end as well.

6. Collimators
Part of the ion beam will be 'implanted' on the collimators. Depending on the volatility the collimators may lose some of their inventory again due to evaporation towards the front-end volume.

7. Mass separator
The mass separator is used to filter out ions with a certain mass-number. The mass separator will have its walls and slits contaminated because of its use. Where the inventory of the mass separator’s slits are required the whole mass separator inventory will be used. Similar to the collimators the inventory can evaporate to the front-end again assuming the volatility is high enough. The RIB leaving the mass separator is considered to be lossless.

8. Extraction station / experimental area
At the extraction station or experimental area the desired RIB arrives. For this work it shall be named extraction station.

9. Decay tank
The decay tank will receive the isotopes that were pumped away but for the purpose of this model it also contains all the pipes after the pumps. The decay tank is meant to contain the radioactive inventory until the decay process has cooled down significantly enough.
2 Theory

The ISOL-process involves different processes which all have their role to play. There is the decay of isotopes which results in a change element, the volatility of the elements which determines the evaporation of the inventories, the molecular flow which determines the flight time inside of the volume, the sticking times which determines how long atoms will stick on certain surfaces and the ionisation which is important for the extraction of the isotopes. All these processes will be discussed in this section.

2.1 Nuclear decay

Unstable nuclei will decay until they decay to a stable nucleus. The most important decay options are:

- $\alpha$-decay.
  A nucleus decay through emitting a $^4\text{He}$ nucleus.

- $\beta$-decay.
  A nucleus decays through either emitting a positron or an electron.

- $\gamma$-decay.
A nucleus will decay to a lower excited state or the ground state by emitting a $\gamma$-photon.

- **Electron capture.**
  A nucleus will capture an electron which will result in the emission of an electron neutrino.

- **Spontaneous fission.**
  A nucleus will fission and break apart into two smaller nuclei.

The decay-rate is quantified as

$$\frac{dN_i}{dt} = \dot{N}_i = -\lambda_i N_i$$

where $N_i$ is the number of isotopes of type i and $\lambda_i$ is the decay constant for isotope i in $\frac{1}{s}$. For any stable isotope $\lambda_i = 0$.

### 2.2 Production

In order to produce the radioactive isotopes as desired for the ISOL-system high energy particle interactions are used. In this case this is done using high energy proton collisions. When a high energy proton interacts with a target nucleus there is a possibility of several processes happening. The most notable processes are spallation,[5] fragmentation and fission which will be discussed in more detail.

![Figure 3: Simplified representation of spallation.[6]](image)

Spallation is the evaporation of neutrons and $\alpha$-particles from the nucleus. After absorption of a high energy particle the target nucleus is left in a highly excited state. In order to lose the energy of this state the nucleus will release neutrons and $\alpha$-particles. This will leave a new daughter nuclei and multiple neutrons.
In the case of fragmentation, a high energy particle will collide with the target nucleus, resulting in a small fraction of the nucleus being released from the mother nucleus. Protons and neutrons can also be released during this process. Both daughter nuclei will significantly differ in size. This is a near instant process where the incident particle does not become part of the whole nucleus before splitting off.

Fission occurs after a particle enters the nucleus and the nucleus eventually falls apart in two daughter nuclei of similar mass and possibly releasing neutrons in the process. Due to the nature of these processes the isotopes that are being formed are formed far away from the stability line. On average every decay should bring the nuclei closer to the stability line however the daughter nuclei can still be far from the stability line. Through decay the newly chemical properties of the nuclei will change which will impact the behaviour as will be described later in this chapter.
Figure 6: The line of stability. The further away from the stability line the shorter the half-life of the isotope is.\[8\]

A more detailed breakdown of the differences between these three processes around the stability line can be found in the appendix.

2.3 Volatility

In order for the isotopes to evaporate from the disc the isotopes need to be volatile. The more volatile the isotopes are, the higher the rate at which the isotopes will leave the medium. The transport of the particle through the substrate is defined by the diffusion while the evaporation from the target is defined by effusion. Diffusion is the random movement of particles due to collisions with other particles. The diffusion flux inside of a material is defined by Fick’s first law as

$$J = -D \frac{\partial c}{\partial x}$$

(2)

where $J$ is the flux in $\text{mol/m}^2\text{s}$, $D$ is the diffusion coefficient in $\text{m}^2\text{s}$, $c$ is the concentration of the isotope in $\text{mol/m}^3$ and $x$ is the direction in m. The diffusion coefficient is based on the interaction between the diffusing atoms, the material and the temperature at which this process happens. This coefficient is dependent on the temperature and the activation energy following

$$D = D_0 e^{-\frac{Q}{RT}}$$

(3)

where $D_0$ is the diffusion coefficient at infinite temperatures in $\text{mol/m}^2\text{s}$, $Q$ is the activation energy, $R$ is the universal gas constant and $T$ is the temperature in
The method used to determine the evaporation / effusion out of the target discs is using the vapour pressures of an elements at the target’s temperature. A vapour pressure is the pressure exerted by a vapour in a thermodynamical equilibrium with its own solid or liquid state. It is an indication of the rate of evaporation of a substance. A substance that has a higher evaporation rate is more volatile and therefore more likely to evaporate or evaporate faster. The vapour pressure is dependent on the temperature and on the isotope of the substance to evaporate. The temperature dependency of the vapour pressure is determined using the empirical formula

$$\log(P_{vap}) = A + \frac{B}{T} + C \log(T) + DT^3$$

where $A$, $B$, $C$ and $D$ are experimentally determined through a fit of data with known pressures.

### 2.4 Ionisation

The preferred way to extract isotopes is to ionise them. Because of the particle being charged it can be extracted using an electrode which will attract the ions through the electrostatic force. After the extraction electrode the ion beam will head towards the mass separator. In the mass separator a magnetic field is present which because of the charged particle will change the path of the particle. Since different isotopes have different masses their paths will be different which will be used to separate the isotopes by mass-number. There are multiple methods possible to ionise a particle. The three main methods used for ISOL will be discussed here:

- **Hot-surface ion source.**
  In the case of hot-surface ion ionisation the particle makes contact with a hot surface upon which there is a chance for an electron to be released to the surface material, effectively ionising the particle. The chance of successful ionisation is dependent on a number of factors and can be calculated using the Saha-Langmuir equation which is as follows:

$$\epsilon = \frac{1}{1 + \frac{g^+}{g^-} e^{\frac{E_{ion} - W}{k_b T}}}$$

where $g^+$ is the degeneracy in the ionised state while $g^-$ is the degeneracy in the neutral state, $k_b$ is the Boltzman constant, $E_{ion}$ its ionisation energy and $W$ the work function of the surface. The desorbed electrons will linger in the ion source and this electron cloud will pull the particles away from the wall if they are ionised lowering the chance of adsorbing another electron and becoming neutral again.

- **Resonant ionisation laser ion source (RILIS).**
  In the case of ionisation using RILIS a specific isotope is ionised through
the step-wise excitation and ultimately liberation of the atoms ’outer’ electron. This is done by bombarding the atom with photons at wavelengths that resonate with these excited energy levels. As such the desired isotopes can be efficiently extracted into a relatively pure RIB.

- Forced electron beam induced arc discharge (FEBIAD).
  In the case of FEBIAD neutrals are guided to a volume into which electrons (emitted through application of a strong electric field) are accelerated to energies of around 150 eV. The energies cause the ionization of the desired isotopes, the abundant presence of electrons prevent recombination of ions against the wall due to electric field interaction between the electron cloud and the ions.

2.5 Molecular flow

Due to the low pressure in the volumes the chance of a free particle colliding with another free particle are negligible. A free particle that is not bound to a surface will move in straight lines through the volume. Due to the collisions with the walls the particles will reach the temperature of the wall. The average kinetic energy of the particle is defined as:

\[ E_{\text{kin}} = \frac{3}{2} k_b T \]  

(6)

at the size and energy levels of the particle this classical equation holds. The means that the velocity of the particles is found to be

\[ v = \sqrt{\frac{3 k_b T}{m}} \]  

(7)

assuming no sticking. If the mean free path length is known the velocity can be used to determine the collisions a particle has per second. The equation

\[ C = \frac{v}{x_{\text{mfp}}} \]  

(8)

describes this where \( C \) is the number of collisions per second and \( x_{\text{mfp}} \) is the mean free path. Thus the mean flight time between collisions is

\[ \tau_f = \frac{x_{\text{mfp}}}{v} \]  

(9)

2.6 Sticking time

The particles moving through the system have a chance to chemically interact with material surfaces in the system and stick to these surfaces for certain lengths of time. The interaction is the adsorption of an atom or molecule on the surface[9]. The average time a molecule will stick on a surface is determined by
the temperature, the surface structure and the material of the adsorbent and chemistry of the molecule. The sticking time can be determined using:

\[ \tau_s = \tau_0 e^{\frac{\Delta H_0}{k_B T}} \]  

where \( \Delta H_0 \) is the adsorption enthalpy, \( \tau_0 \) is the sticking time at infinite temperatures. \( \tau_0 = \frac{1}{\nu_m} \) where the Debye frequency \( (\nu_m) \) represents with which the atom/molecule tries to leave the surface. This frequency should be of the vertical vibration of the absorbed atom / molecule but due to the absence of this data and the fact that it is not sensitive due to being linear dependent the maximum vibrational frequency of the bulk material is used as an approximation. The adsorption enthalpy changes from element to element and also differs depending on the adsorbent’s chemical and structural make up. Accuracy however is very important because of the exponential nature of equation 10 small deviations will have a large impact.

3 Implementation of the model

3.1 The main model

In order to model the inventory in different volumes the system is constructed as a system of coupled differential equations. The equation for isotope i in volume l would be:

\[ \frac{dN^l_i}{dt} = \dot{N}^l_i = P^l_i + \sum_{j \neq i} \lambda^j_i N^l_j + \sum_{m \neq l} \left( \delta^l_{i,m} N^m_i - \delta^m_{i,l} N^l_i \right) - \lambda^i_i N^l_i \]  

which is inspired by the work of Kim Rijpstra[12].

- \( N^l_i \) is the number of atoms of isotope i in volume l.
- \( P^l_i \) is the production of isotope i in volume l in particles per second. This will be zero for all volumes except for the target volume.
- \( \lambda^j_i \) is the decay constant of isotope j to isotope i. \( (\lambda^j_i = f_i \lambda_j \) where \( \lambda_j \) is the decay constant for isotope j while \( f_i \) is the branching ratio resulting in a decay to i).
- \( \delta^l_{i,m} \) is the factor determining the movement rate between volumes. \( l \) is the volume the isotope moves towards, \( m \) is the volume it moves from and \( i \) is the isotope including the deposition / extraction of ionised particles.

In order to solve the system inventories for the different volumes a system of coupled differential equations for the system being active is constructed. The matrix can be found in the appendix as equation [A1] where all the elements correspond to 3852x3852 matrices. The symbols correspond to:

- \( D \) as the decay matrix.
• $V_H$ as the volatility matrix in the target system in the hot parts of the system (target).

• $V_C$ as the volatility matrix in the target system in the cold parts of the system.

• $M$ as the molecular flow matrix in the target container and casserole.

• $\epsilon$ as the ionisation matrix.

• $F$ as the mass separator matrix.

• $U$ as the pumping matrix for the extraction station and the front end.

Multiplication between different matrices is not matrix multiplication but multiplication of the elements in the matrices on the same positions. The factor 0.95, 0.04 and 0.01 are parameters in the model and are based on percentages in other ISOL-facilities they correspond to the amount of the total RIB making it to the mass separator, getting stuck on the mass separator system and getting stuck on the collimators respectively. $A$ determines the fraction of neutral isotopes leaving the ion source that are deposited in the casserole (in the model $A$ is taken as 0.5). The named sections within the vectors correspond themselves to vectors representing the number of atoms or its time derivative for each of the 3852 known isotopes within the named volume. The production vector is defined as it is calculated using FLUKA applied to the disc section of the vector. The initial condition is a 8x3852 elements long vector with all values set to zero except for the discs containing thorium carbide (this corresponds to the thorium carbide in the discs at beginning of life).

The target volume is 20 cm long and the targets have a diameter of 4 cm while the volume has an inner diameter of 5 cm as seen in figure 7. The volume contains 10 thorium carbide discs (figure 8) with an excess of carbon (ThC$_2$ + C$_2$) and the discs have a thickness of 0.5 mm and a density of 4 g/cm$^3$. The temperature of the target volume, discs and ioniser is 2200 K while the rest of the system is considered to be at 323 K.

![Figure 7: Target volume with ioniser-tube.](image1)

![Figure 8: Target volume with discs visible.](image2)
3.2 Cooling down

During the cool down period the beam is turned off and segments of the system are closed down to avoid atoms moving between these segments. This means there is no transport possible between the target container and the casserole and from the casserole to the front end. The matrix describing the cooling down period can be found in the appendix as equation A2. There is no transient period in this model. The temperature of the target will go from 2200 K to 373 K in an instant. Due to the large difference in temperature and the fact that the model will be at the lower temperature for a far longer period of time this assumption is considered as reasonable. Due to these circumstance the system of differential equations changes quite drastically. The lower temperatures will result in a reduction in volatility. This means the volatility matrix for the high temperatures is replaced by the volatility at lower temperatures. The individual matrices used in both larger matrices will be discussed in more detail in the following subsections.

3.3 Decay matrix

With the formation of unstable isotopes radioactive decay will significantly influence the system. Unstable isotopes will continue decaying until they become stable. The amount of every decaying nucleus lost will be equal to the number of daughter nuclei created in the volume. For example:

\[
\begin{pmatrix}
\dot{N}_A \\ \dot{N}_B \\ \dot{N}_C \\ \dot{N}_D 
\end{pmatrix} = 
\begin{pmatrix}
-\lambda_A & 0 & 0 & 0 \\ f_B & 0 & 0 & 0 \\ f_C & -\lambda_C & 0 & 0 \\ 0 & 0 & \lambda_C & 0 
\end{pmatrix} 
\begin{pmatrix}
N_A \\ N_B \\ N_C \\ N_D 
\end{pmatrix}
\] (12)

is a decay matrix for a theoretical universe which contains only four different isotopes called A, B, C and D. A can decay into both B and C with corresponding branching ratios \( f_B \) and \( f_C \) where \( f_B + f_C = 1 \) and C decays to D. Both B and D are stable. A schematic representation of this can be seen in figure 9. \( \lambda_i \) refers to the decay constant for isotope i.
For the real world matrix a similar procedure was used containing 3852 known isotopes resulting in a 3852x3852 matrix where the half-life times were extracted JEFF-3.1/RDD.

### 3.4 Volatility matrix

Due to the fact that the diffusion coefficient is dependent on a lot of factors such as lattice structure, the traversed material and the atomic structure of the diffusing molecule as well as the temperature. The number of possible molecule-material combinations is enormous, and hardly explored when it comes to diffusion (and adsorption). This is especially true for thorium carbide. A possible solution for future researchers would be the creation of a diffusion coefficient database containing measured diffusion coefficients of materials in different materials cited with the circumstances under which the experiment was executed.

Due to the absence of diffusion values and the thinness of the target discs the assumption is made that diffusion through the target can be neglected in favour of just taking into account the evaporation from the target discs to the target container this approximation works best for very mobile isotopes and to a lesser extent for very immobile isotopes. In order to determine the evaporation rate the evaporation pressures were used. As shown in equation [4] the vapour pressure will generally increase for increasing temperatures at high temperatures (where $\frac{Q}{T}$ becomes negligible compared to $DT^3$). As per U. Köster [13] and shown in figure [10] the volatility of the elements will be divided into five different groups. The groups are made based on the temperature at which the vapour pressure exceeds 0.01 mbar as this will be the pressure at which the evaporation pressure is higher than the vessel pressure and this should thus lead to significant evaporation. The five groups are defined as follows:

1. Vapour pressure of 0.01 millibar is reached at a temperature lower than 100°C.
2. Vapour pressure of 0.01 millibar is reached at a temperature between 100°C and 400°C.

3. Vapour pressure of 0.01 millibar is reached at a temperature between 400°C and 1000°C.

4. Vapour pressure of 0.01 millibar is reached at a temperature between 1000°C and 2000°C.

5. Vapour pressure of 0.01 millibar is reached at a temperature higher than 2000°C.

The evaporation out of a volume is represented in equation 11 by the term $\delta_{i, 1}$, where this term can be responsible for different processes resulting in a transport between volumes. In this subsection only the volatility aspect is taken into account. Volatility is only used in this model for the evaporation of the inventory in the discs, mass separator and collimators. The volatility dependent part of this term is defined as $\frac{\ln(2)}{t_{1/2}}$ where $t_{1/2}$ is the time it takes for half of the nuclei to evaporate out of the system.

Figure 10: Visual representation of the five groups with their corresponding elements.[13]
In the case of lower temperatures a lot of elements will be significantly below their evaporation pressures and these elements are assumed not to be evaporated at all and will therefore remain in the volume unless they possibly decay. The times in the lower temperature case are taken by taking the same times from the heated groups when the difference between the group temperature and the volume temperature are similar.

### 3.5 Ionisation matrix

While laser ionisation should be implemented as fast as possible ISOL@MYRRHA will initially work with surface ionisation and even after the implementation of laser ionisation surface ionisation is still a very real possibility in which case surface ionisation can be considered for conservative estimates of what gets ionised. For determining the ionisation matrix equation 5 (Saha-Langmuir) is used to calculate the chance for all of the isotopes to ionise. In this model the ionisation is considered to be a time independent process. A particle will either be ionised or not but doesn’t spend time in the ioniser. Equation 5 accounts for the probability to ionise upon a single collision. If an atom does not become ionised it might collide with the ioniser again and has another chance to become ionised again. An ionised atom that collides with the ioniser again has the chance to lose its ionisation. In order to mitigate this effect a solution was implemented as follows:

1. An ionised particle will be extracted due to the extraction field and the plasma working of a hot cavity and will therefore not lose its ionisation.
2. An unionised particle will continue colliding with the ioniser until:
   a) The particle has left the ioniser.
   b) The particle becomes ionised and is extracted.

The chance to ionise a particle would thus become \( P = 1 - (1 - \epsilon)^C \) where \( \epsilon \) is determined using equation 5, \( C \) is the average number of collisions in the ioniser before the particle leaves the ioniser and is determined to be 2009 using MolFlow+ [15]. For the ioniser itself it is noted that the hot-surface ioniser is made out of tantalum which has a work function 4.25 eV it will be attached to a 5 cm long transfer line with an inner diameter of 8 mm and the ionier itself was 3 cm long with a diameter of 3 mm it will be used at a temperature of 2200K.
3.6 Molecular flow matrix

FreeCAD\[14\] was used to model the target container with sheet after which this was exported to MolFlow+\[15\]. For the molecular flow simulation the discs are selected as particle desorbers and the end of the ionisation tube is defined as the particle adsorber. After a MolFlow+ simulation of one hour real-world time the average number of collisions was determined to be 86041 in the target container and 713 in the casserole. The model used is shown in figure\[11\]. Since the dimensions of the front-end and the extraction station are unknown they are set to the same values as the casserole as parameters in the model. As the adsorption enthalpy is unknown the evaporation enthalpy can be as
\[
\frac{d \ln \frac{P_{\text{sat}}}{P_0}}{dT} = \frac{\Delta H_{\text{sub}}}{RT^2}
\]

Since evaporation is adsorption of an element surrounded by the same element it is able to give an indication of the adsorption enthalpy. In order to determine the Debye frequency
\[
\nu_0 = \frac{k_B}{\theta_D \hbar}
\]

where \(\theta_D\) is the Debye temperature and \(\hbar\) is the reduced Planck constant will be used to determine the Debye frequency. Using equation\[9\] and \[10\] will result in the flight time between collisions while using equation\[10\] and \[13\] will result in the sticking time after every collision. The residence time therefore will be
\[
\tau_{\text{res}} = N(\tau_f + \tau_s)
\]

where \(N\) is average number of collisions in the target container. In equation\[11\] the collision matrix is part of the \(\delta\) being responsible for the transport between volumes. For noble gasses and for elements which are boiling at these temperatures an adsorption enthalpy of 21 kJ/mol as well as a sticking time of 0 for noble gasses\[16\].
Figure 11: Casserole in red with the target container in white. The holes leading to the front end are shown.

3.7 Filter matrix

The mass separator separates atoms based on their mass. The mass-number of the desired isotope is thus chosen and therefore the matrix is designed with a 99% chance of letting the desired mass-number go through and 1% chance of allowing an isotope that only differs with the desired mass-number by one due to imperfections in the mass-separator-system. An atom that does not go through the mass-separator is thus considered as an atom that impacts on the mass separator slits and will be considered as having moved to the mass-separator volumes.

3.8 Pumping matrix

At the near-vacuum condition of the system pumping away molecules inside of the system is no longer a process that is dependent on pressure differences but becomes a problem similar to the molecular flow problem similar as with the molecular flow matrix. A particle will only get pumped away if it happens to collide with the pumping area. Because the dimensions of the systems are not yet known the pumping matrices were created using the number of collisions and the free path length of the casserole. These are parameters in the model which can easily be changed after dimensions are known and modelled using MolFlow+. The total residence time is once more calculated using equation [15]. Similar as to the molecular flow matrix in the casserole the pumping matrices will only affect the elements from volatility groups 1 and 2 since the other elements will not be volatile at the temperatures in the volumes where the pumping happens.
3.9 Solution method

In order to solve this system of coupled differential equations python3 was used. A list of all the packages used can be found in the appendix. The scripts are tasked with constructing all the matrices as described above and combining these into one large matrix as seen in equation A1. The production vector is created using FLUKA. For the initial conditions all volumes are empty of any isotopes except for the thorium content in the discs which is set to the target thorium mass. The system is solved using script generate_data_stick.py for the beam being turned on and then for the beam being turned off (switching to the cooling down matrix equation) for several cycles with every cycle being saved to the hard disk to save computer memory. After all the cycles are done a cool down calculation is done (extended period using equation A2). After every cycle except for the last one the casserole is replaced resetting the discs, target vacuum and casserole back to their initial states. After generating the inventories for the different volumes in time the script process_data.py is used to calculate the activity in all the volumes in time and plotting the results. The solver being used to solve the differential equations is solve ivp which is an initial value problem solver. Due to the stiffness of the system being spread out over many orders of magnitudes the backwards differentiation formula method was used.

4 Validation of the model

The model as constructed with the previously mentioned set of instructions is based on a set of fundamental assumptions which need to be validated. These assumptions are:

1. Simplifying the evaporation from the target discs to five different groups based on the vapour pressure and the effect this has on the inventories of isotopes is small enough to still consider the data to be valid.

2. The amount of particles leaking out of the target through the window is low enough to be irrelevant.

The assumptions as described and the accuracy of the matrices as constructed have been tested through a variety of methods as shall be discussed in this section.

4.1 Decay matrix validation

For validation of the decay matrix a scenario was created containing one million $^{90}\text{Sr}$ in a single volume without any form of transport. The script decaytest.py uses the decay matrix to evaluate the change in isotopes within the volume. The resulting plot of the isotopes over a period of 60 years is then compared to Nucleonica’s Decay Engine++.
From the comparison it seems that the decay matrix is operating as expected.
The values and behaviour seems the same using both the Decay Engine++ as the matrix as constructed. The half-life of $^{90}\text{Sr}$ 28.79 years is much higher than the half life of $^{90}\text{Y}$s half-life of 2.671 days therefor an equilibrium is created where the amount of $^{90}\text{Y}$ decaying is equal to the amount of $^{90}\text{Sr}$ decaying into $^{90}\text{Y}$ this equilibrium remains until a significant enough amount of time has passed to affect the amount of $^{90}\text{Sr}$ decaying and thus breaking the equilibrium conditions. From its similar behaviour to that of the Decay Engine++ and its physically expected behaviour the decay matrix can be considered as valid for the rest of this report.

4.2 Volatility matrix validation

For validation of the volatility matrix a demo-system was created containing two hypothetical volumes where material could evaporate out of volume 1 into volume 2 but not the other way around. Volume 1 contains $10^{11}$ nuclei of $^{222}\text{Rn}$ ($t_\frac{1}{2}=3.8235$ days) which decays to $^{218}\text{Po}$ ($t_\frac{1}{2}=3.098$ minutes) which in turn decays to $^{214}\text{Pb}$ ($t_\frac{1}{2}=27.06$ minutes) which also means that the isotope will change volatility group from group 1 to 2 and 3 respectively meaning it will become less and less volatile the further it decays. The script `decay_vol_test.py` managed the inventory of both these volumes and plots graphs containing the volumes in time.

![Graphs showing decay and transport of $^{222}\text{Rn}$, $^{218}\text{Po}$ and $^{214}\text{Pb}$](image)

Figure 14: The decay and transport of $^{222}\text{Rn}$, $^{218}\text{Po}$ and $^{214}\text{Pb}$. The total loss of isotopes is due to the instability of $^{214}\text{Pb}$ whose daughter isotopes are not taken into account.

It is clearly shown that the radon atoms leave volume 1 at a significantly faster pace than the other two isotopes while $^{218}\text{Po}$ is far less common in volume 1 due to the fast evaporation of the mother nuclei and its shorter half life it still...
remains in volume 1 for a longer period of time than \(^{222}\text{Rn}\). The same can be said for \(^{214}\text{Pb}\) which’s half life is closer to that of \(^{218}\text{Po}\) but still remains in the system for a much longer period of time due to its low volatility. The reason for the total number of atoms diminishing is because of the instability of \(^{214}\text{Pb}\) meaning it will decay on into a daughter isotope that is not being followed.

Figure 15: Decay of \(^{222}\text{Rn}\), \(^{218}\text{Po}\) and \(^{214}\text{Pb}\) as per Decay Engine++[20]

Due to all of the isotopes of interested leaving volume 1 very quickly it is possible to compare the decay of \(^{222}\text{Rn}\) to the inventory in volume 2 using Decay Engine++[20] again. This further validates the decay matrix. The behaviour of the volatility matrix seems to express the behaviour as expected. Due to the exact rates of evaporation not being known the matrix can be validated as reasonable but the exact values can not be validated. Therefore it is important to evaluate the importance of the exact group times’ influence.

4.3 Volatility group times validation

With the evaporation group times being generalisations over a large number of elements it needs to be validated to what degree these generalisations affect the built-up of inventories in different parts of the system. In a real-world scenario the evaporation times likely differ from the used times given to the groups thus an assessment of the stability of these times is required. This is done by varying these times and determining the built-ups of inventories based on these times. If the results do not change significantly the system can be considered stable around these values. In which case the evaporation most likely holds up reasonably well for a real-world scenario, assuming the elemental evaporation times are close enough to the group times used. The validation is done by
determining the build-up in all the volumes for four different isotopes from four
different volatility groups.

<table>
<thead>
<tr>
<th>Combination</th>
<th>t1</th>
<th>t2</th>
<th>t3</th>
<th>t4</th>
<th>t5</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>10^{-2}</td>
<td>1</td>
<td>10^4</td>
<td>10^8</td>
<td></td>
</tr>
<tr>
<td>B</td>
<td>10^{-3}</td>
<td>1</td>
<td>10^4</td>
<td>10^8</td>
<td></td>
</tr>
<tr>
<td>C</td>
<td>10^{-4}</td>
<td>1</td>
<td>10^4</td>
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<tr>
<td>D</td>
<td>10^{-2}</td>
<td>0.1</td>
<td>10^4</td>
<td>10^8</td>
<td></td>
</tr>
<tr>
<td>E</td>
<td>10^{-2}</td>
<td>10</td>
<td>10^3</td>
<td>10^8</td>
<td></td>
</tr>
<tr>
<td>F</td>
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</tr>
<tr>
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<tr>
<td>I</td>
<td>10^{-2}</td>
<td>1</td>
<td>10^5</td>
<td>10^8</td>
<td></td>
</tr>
<tr>
<td>J</td>
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<td>1</td>
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<td>10^8</td>
<td></td>
</tr>
<tr>
<td>K</td>
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<td>10^{-1}</td>
<td>10</td>
<td>10^3</td>
<td>10^8</td>
<td></td>
</tr>
</tbody>
</table>

Table 2: The combinations being execute in order to check the validity of the
model.

The isotopes being followed are $^{141}$Xe (half-life 1.73 s), $^{94}$Rb (half-life 2.702
s), $^{114}$In (half-life 71.9 s) and $^{225}$Ac (half-life 9.920 d) which correspond to group
1, 2, 3 and 4 respectively. The mass separator will be set to their respective
mass-numbers in order to separate out these specific isotopes. The run-time
with the beam on will be set to 28 days there will be no cooling down period.
In order to follow the isotope of interest the script follow_isotope.py[21] is used
which loads the data relative to the isotope in question and plots it in time for
all volumes while also printing the final amount of the isotope in each volume.
The data being used is created using generate_data_stick.py[21].
Table 3: Different iterations with the set-up configured to extract the isotope in question. X means that all the results were similar to the reference (A). The number is related to the volumes respectively: discs, target container, extraction station, mass separator, front-end, collimators, casserole and decay tank. The amount of pluses is related to the amount of orders of magnitude the value is higher than the reference and the minuses signify the amount of orders of magnitude the value is below the reference.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
<th>E</th>
<th>F</th>
<th>G</th>
<th>H</th>
<th>I</th>
<th>J</th>
<th>K</th>
<th>L</th>
<th>M</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{131}$Xe</td>
<td>X</td>
<td>1-</td>
<td>1+</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>1-</td>
<td>1+</td>
</tr>
<tr>
<td>$^{94}$Rb</td>
<td>X</td>
<td>X</td>
<td>7-</td>
<td>1-</td>
<td>3+</td>
<td>2-</td>
<td>5-</td>
<td>6-</td>
<td>8-</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>5+</td>
</tr>
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<td>3+</td>
<td>4+</td>
<td>5-</td>
<td>6+</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7-</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^{115}$In</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>1+</td>
<td>2-</td>
<td>3+</td>
<td>4+</td>
<td>5-</td>
<td>6+</td>
</tr>
<tr>
<td>$^{229}$Ac</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>X</td>
<td>1-</td>
<td>1+</td>
<td>5+</td>
<td>6+</td>
<td>7-</td>
<td>7+</td>
<td>1-</td>
</tr>
</tbody>
</table>

4.4 Leakage effects

Due to very low production rates of certain isotopes of interest it is not only important to filter out the right isotope in order to prevent dilution it is also very important to make sure as little of the isotope of interest is lost as possible. A significant losing factor could be the leakage of formed isotopes through the window / plug. In order to validate this the target was designed with a plug using freeCAD\[14\] and exported to MolFlow\[15\]. For the desorbers the target discs were chosen while the particles could only leave the system by being absorbed in either the tip of the ioniser or due to leaking out of the system through the plug. The total number of particles leaving the system are determined and the fraction leaving through the ioniser is determined for different opacities. Due to the graphite being used to fill up the the sides between the plug and the target vessel different values for the opacity were used The plug being used in
this model has a gap of 0.5 mm.

Figure 16: Green marks the gap.

Figure 17: The relative fraction lost depending on the opacity of the gap.

From the simulation it is shown that due to the significantly smaller area of the hole to the ioniser than the gap between the plug and the target vessel. This
difference in size results in more collisions on the gap than particles leaving the
vessel through the ioniser. This means that a high opacity of the gap is required
in order to prevents as much loss as possible or a smaller gap if possible. It is
shown that a small change in opacity can have significant consequences. Due
to the optimalisation options available and the unknown nature of the opacity
as of now it will be assumed for the remainder of this work that the gap has an
opacity of 100% and there shall thus be no leakage.

5 Model results

5.1 Production

Using FLUKA the production per 100-MeV proton impacting on the thorium-
carbide target was determined. The production vector can then be divided in
around three different clusters.

1. Hydrogen up till oxygen (atom number 1 till 8)
2. Manganese up till terbium (atom number 25 till 65)
3. Francium up till protactinium (atom number 87 till 91)
The production of isotopes in the target per proton. The production is visualised using a base 10 logarithm and smaller white dots are used to show the stability line.

The resulting production of isotopes is in line with the expected production. Most of the isotopes being created are the result of a fission event in the $^{232}\text{Th}$ which as discussed in the production subsection results in the large group with atom numbers between 25 and 65 and the two smaller groups are the result of fragmentation and proton capture mostly. Spallation is not that significant of a contributor at these proton energies. The overwhelming majority of the isotopes produced are below the stability line as is expected due to the excess of neutrons being available in $^{232}\text{Th}$ and these remaining after the nucleus splits into lighter nuclei further away from the stability line. The production of isotopes...
for different targets and energies can be found in the appendix.

5.2 Volume based activity

In order to determine the activity in different volumes the whole system is simulated for a whole year consisting of 12 cycles of a month. The scripts `generate_data_stick.py` and `process_data.py` are used for this.

<table>
<thead>
<tr>
<th>Setting</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volume of target</td>
<td>6.283 cm³</td>
</tr>
<tr>
<td>Density ThC₂+ C₂</td>
<td>4 g/cm³</td>
</tr>
<tr>
<td>Beam intensity</td>
<td>$3.125 \times 10^{15}$ protons/second (500 µA)</td>
</tr>
<tr>
<td>Duration active cycle</td>
<td>28 days</td>
</tr>
<tr>
<td>Cooling between cycles</td>
<td>1 day</td>
</tr>
<tr>
<td>Cooling after last cycle</td>
<td>14 days</td>
</tr>
<tr>
<td>Number of cycles</td>
<td>12</td>
</tr>
<tr>
<td>Temperature target</td>
<td>2200 K</td>
</tr>
<tr>
<td>Temperature vacuum system</td>
<td>323 K</td>
</tr>
<tr>
<td>Mass-number for extraction</td>
<td>225</td>
</tr>
</tbody>
</table>

Table 4: Variables as set for this iteration

In this iteration the mass separator was set to extract isotopes with mass-number 225 (this could be used as a way to extract $^{225}$Ac). At beginning of life the system starts out completely clean (new system) without any contaminants.

A year of operations for this system would consist of 12 cycles of 28 days between which cycles it is cooling down for 1 day. During the cooling down period the system is colder and the system is closed off at the ion-source and between the front end and the casserole. After the cooling down period the casserole containing the target itself is replaced with a clean new casserole. This process is repeated until 12 cycles have been done. After the 12 cycles the system is powered down for a two week period for large scale maintenance.
Figure 19: Activity in the discs, target vacuum, mass separator, transport tube, collimator and casserole volume in time.
As shown in figure 19 there are peaks after beam shutdown in the target container. These peaks are caused by the evaporation of isotopes from the discs while the target container is unable to remove the isotopes from its volume due to the container being closed off. It is clearly shown that the activity increases the most in the beginning of every cycle due to the short lived isotopes and then builds up more slowly for the remainder of the time due to the longer lived isotopes building up. This is behaviour as expected since short lived isotopes will reach an equilibrium the fastest and will be the largest contributing factor to the activity. Longer lived isotopes can also reach an equilibrium but the ones who reach an equilibrium within the time-span of the cycle, however this equilibrium is reached not by just decay but also by the entering or leaving of volumes through processes like molecular flow and pumping. Longer lived isotopes that don’t leave volumes or don’t leave volumes fast enough will not reach an equilibrium within the time-span of the experiment or, depending on the half-life not even within the lifetime of the ISOL system and thus they will continue to build up. When the proton beam is turned off the equilibrium is broken and especially the effect on short lived isotopes is shown by their quick decay resulting in the sudden drop of activity.
Figure 21: The amount of isotopes of $^{225}$Ac and its daughter nuclei in the discs, target vacuum, mass separator, transport tube, collimater and casserole volume in time. The numbers are isotope names using the ZZAAAM convention. ZZ being the proton number AAA the mass-number and M the excited state.
As can be seen in figure 21 and 22 the isotope $^{209}$Bi will slowly build up in several volumes. This can be mostly attributed to the fact that this isotope is stable and the most likely end-point for the decay-chain. $^{225}$Ac reaches an equilibrium in most volumes within several days if there is no option to leave the volume and faster if there is the possibility to leave the volume without decaying. This is in line with the expectation of an isotope in volatility group 4. $^{205}$Ti is also a stable isotope but is decay through this chain is less likely to occur which makes the production small enough not to be noticed on these graphs. The same problem exists for $^{221}$Fr, $^{217}$Ac, $^{217}$Rn, $^{213}$Po and $^{209}$Pb which do not show up due to short half-lives and lower production rates. Due to the lower volatility of especially bismuth in the lower temperature areas bismuth will mostly remain in the colder volumes once there. This will result in a higher build up through many years of production. The fact that bismuth is the most common at the extraction station is due to $^{225}$Ac remaining there to decay. If the isotope of interest is extracted and used the build-up will not happen at the extraction station which might also prevent further dilution of the isotope of interest after a longer period of time.

5.3 The importance of sticking time

In order to demonstrate the impact sticking time has on the inventory of isotopes two different simulations were also done using `generate_data_stick.py`. One without any sticking time and one with a sticking time ten times longer. The
remainder of the conditions are equal to those in the subsection volume based activity.

Figure 23: Activity in the discs, target vacuum, mass separator, transport tube, collimator and casserole volume in time without sticking.
Figure 24: Activity in the decay tank and extraction station volume in time without sticking.
Figure 25: Activity in the discs, target vacuum, mass separator, transport tube, collimator and casserole volume in time with 10 times the sticking sticking.
As can be seen in figure 19, 20, 23, 24, 25 and 26 the most severely impacted volume is the target container. This is as expected considering that the most collisions happen here. The discs, mass separator, collimator and decay tank are the least impacted which is to be expected considering the lack of molecular flow and thus collisions on walls where atoms can stick. In other volumes the activity is not as severely impacted but it does show the importance of accurate sticking times. The impact on individual isotopes is more severe for isotopes with a short half-life while isotopes with long half-lives are barely affected.

5.4 Radiation dose slits

In order to prevent damage to the slits of the mass separator as well as the slits becoming too irradiated to handle the slits need to periodically be replaced and stored away as nuclear waste. For this purpose the inventory of the mass separator at the end on the cycles as generated using `generate_data_stick.py` is exported using `export.py` to the Nucleonica’s Dosimetry and shielding app. The main goal is to find a safe way to store the slits where safe is defined as follows:

1. For the sake of handling the dose-rate at the beginning of storage should not exceed 10 µSv/h.

2. In order to comply with the legislation regarding allowed doses for people working in the nuclear industry the yearly absorbed dose should not exceed 20 mSv (100 mSv in 5 years).
Using nucleonica for a standard case of 100 cm of distance to the source packaged in 1 cm thick lead the dose-rate comes down to 4.7 mSv/h, exceeding the maximum allowed dose-rate by several orders of magnitude. This dose can be lowered significantly using several other possible methods such as:

- **Water shielding.**
  The vessel is submersed in water for shielding until the activity has gone down enough to store the slits in dry-storage.

- **Extended cooling.**
  The slits can be left in the ISOL system until the activity has gone down enough to handle then easier.

- **Extra layer for storage.**
  The lead vessel can be stored in a building that has no people in it and shields the outside world from the radiation coming from the stored vessels.

- **Increasing the shielding material.**
  The shielding material can be made thicker so that the effective dose-rate will be lower.

For the interest of this work the increasing of the shielding material is looked at in more detail. While the absorbed doses as described would not violate any laws it does violate the ALARA principle and therefore the increased shielding would most likely be combined with the extra layered storage. In the interest of remaining conservative and giving a worst case scenario it will be assumed that an employee will be at a distance of 5 metres from the vessel for all of his/her working hours (8 hours a day, 5 days a week, 48 weeks a year). Using lead shielding with a thickness of 7.1 cm at a distance of 5 metres the initial dose-rate as determined by Nucleonica to be 9.7 $\mu$Sv/h. Since Nucleonica does not calculate an integrated dose over a longer period of time this dose is calculated using `slits_dose.py` which integrates the the normalised activity over a one year period in order to approximate the total absorbed dose. In order to calculate the integrated dose like this it is assumed that the inventory make-up remains the same. In reality this will lead to an overestimation of the absorbed dose due to high energy emissions being more common amongst more unstable isotopes. The dose for the stored slits in first year comes down to 4.4 mSv.
Figure 27: The activity of the stored slits in the first year normalised to the initial activity.

As seen in figure 28 especially in the first few days the activity will go down quite a lot. This will be due to the short lived isotopes decaying to stable or less active isotopes. The dose as absorbed in the first year will therefore also be the highest dose that a possible employee could get from the material. Every following year will result in a lower dose until everything has decayed.
Figure 28: The dose-rate through time in storage.

5.5 Leaking casserole

Due to the nature of the ISOL-process it is very important to maintain the integrity of the system in order to prevent radioactive isotopes from leaving the system. In the case of this failing despite best efforts a leak can result in the release of part of the inventory. The following incident was simulated with a leak in the front end / casserole due to a pierced casserole, a faulty weld or a casserole which is not in the proper place. The following worst case scenario is assumed:

- The inventory in the casserole is not going to the front-end.
- The target discs will be on fire due to the presence of oxygen and high temperatures.
- The leak is there since day 1 of the casserole usage.
- After one minute the radiation has been detected and the proton beam automatically gets diverted away from the ISOL-facility.
- It takes 4 minutes to evacuate the personnel from the room.
- During operation the casserole inventory gets leaked to the environment.
• After shut down the target discs will disintegrate due to the fire and all of its inventory will be released to the target volume.

• Due to the fire damage the target vacuum will keep leaking its inventory to the casserole after shutdown.

In order to simulate this scenario a modified version of \textit{generate\_data.py} is used as \textit{accident.py}. For the purpose of conservatism it was assumed that the full disk inventory will be moved to the target volume when the beam is turned off. This neglects the need for the particles to evaporate from the discs which makes the output of particles higher than it will be in reality. In the interest of conservatism it is assumed that all these particles will be volatile enough to leave the target to the casserole where they will all leave the casserole system. Once more Nucleonica’s Dosimetry and shielding\cite{20} is used to calculate the dose-rate at the point of highest activity at a distance of 100 cm with 100 cm of air for shielding. The dose-rate there is found to be 62 mSv/h. Which in the case of 5 minutes of exposure would result in an absorbed dose of 5.2 mSv assuming there is no contamination.

![Activity leaked](image)

\textbf{Figure 29:} Activity in the first 5 minutes.

In reality the dose-rate will not be at the maximum dose during the whole accident scenario. When integrating over the activity and using this to calculate the total dose rate the amount comes down to 0.9 mSv.
As seen in figure 30 after a short period of time the dose-rate has lowered enough to allow personnel back inside again in order to clean the system and make repairs and switch out the casserole again. The reason for this short time is that the system has not operated long enough to create a significant enough build-up of radioactive isotopes. If detection or evacuation were to take longer the dose received by personnel would increase as would the time required to wait before the system can be repaired.

6 Conclusion and discussion

6.1 The importance of the sticking time

As per equation 10 the sticking time is determined by the adsorption enthalpy. Due to the sticking time being exponentially dependent on the enthalpy energy it is in the interest of accuracy of the sticking time to have an enthalpy energy which is very well defined. For the ISOL-system made out of tantalum the following problems appear:

1. The enthalpy energy is dependent on the lattice structure.
   - The lattice structure is not the same everywhere
2. The enthalpy energy is dependent on both absorbate and absorbent.

- The energy can not be generalised for a chemical group.
- The values are not known for most elements.

To mitigate this the adsorption enthalpy was replaced with the evaporation enthalpy. While holding through for desorption for element A from element A it will not hold up for different elements on other elements. Because the Debye frequency is being used in order to determine the sticking time at infinite temperatures the possible introduction of errors here is not as problematic as is the case with the adsorption enthalpy since the dependence will be linear.

6.2 Formation of compounds

Due to the high temperatures in the target it would be reasonable to assume some of the formed isotopes will form compounds with other isotopes in the target or with the excess of carbon in the thorium-carbide. In this case the volatility and the velocity of the compound would be different from the particle in an unbounded state this would undermine the model as it only takes into account elements and not compounds.

6.3 Simplification of the model

The model uses a highly idealised version of the ISOL-system. The discs are not connected to any part of the target itself and the plug / windows is experiencing no leakage at all. While the assumption based on the discs seems like a reasonably assumption both assumptions may negatively affect the reliability of the model but due to a lack of data to compare the model to this can not be verified.

6.4 Radiation doses

The radiation dose in the case of the stored slits is below the legal limit and calculated in a highly conservative worst case scenario. From this it is safe to conclude that taking into account the proper shielding the storage of the slits should not result in any problems. In the accident case the dose is relatively high for the short period but does not violate any legal limits and will only happen in the case of a severe accident. While it would affect the operations of the facility it will not result in a total loss and no long term damage to both personnel and equipment.
7 Future research proposal

7.1 Adsorption enthalpies
In order to improve on the model the sticking times should be taken into account. A future research project could consist of experimentally determining the adsorption enthalpies of elements on tantalum. This experiment can also be used to determine how significant the effect of different lattice structures on the enthalpy in this specific will be.

7.2 Molecular sidebands
While some isotopes will be very difficult to extract due to their low volatility this might be eased by using extraction through molecular sidebands. In this case non-volatile isotopes will form a compound with another available material which will be easier to extract due to a higher volatility.

7.3 Ionisation chance
The ionisation chance as calculated for this model has some problematic assumptions. In reality an ionised particle that collides with the ioniser again has a chance of losing its ionisation and ionised particles have a high chance of leaving the ioniser due to extraction via the extraction electrode. To solve this a program that is a better tool for this task should be used in the future. A possible option could be VSIM.

7.4 Comparison sample
To validate the model an experiment could be constructed with similar proton-energies with a similar target at an ISOL-site to collect data on the inventories and activities in different volumes in order to validate the similarity between the model as described here and real world data.

7.5 Advice for future researchers
A first attempt at building this model was made using SciPy for python with the *ode_solver*. While initial results were promising using this solver the solver started to crash when the system grew. This solver is dated and deprecated and should not be used for a problem such as this. *solve_ivp* has shown to work faster and more reliable.

8 Acknowledgement
This work would not have been possible without the help of a great many people who I would like to thank. Waclaw Gudowski for arranging for me to be able to do my master thesis research in Belgium at SCK•CEN. Lucia-Ana
Popescu for being my mentor and always being available for questions. Donald Houngbo for helping me get set up with FLUKA and helping me with building my model and pointing me towards the right literature when I needed to learn more, Kim Rijpstra for pushing me to try new and different things, sharing his insights on potential solutions and proofreading this work. Marc Dierckx for being the inspiration on a possible method for finding a solution and checking the mathematics used in the model as well as proofreading my thesis, Philip Creemers for always taking the time to explain parts of the ISOL system and supplying me with information required for the model, Lars Ghys for answering all my physics questions and Martin Ashford for his help with FLUKA when I ran into problems again. I am sure I kept you all away from your work for many hours and I would like to thank you for the time you spent helping me.

9 References


[4] ISOL@MYRRHA, fundamental physics at MYRRHA, SCKCEN, 2011.


[16] C. Lau et al, Recent studies to improve release properties from thick isotope separator on-line fission targets, Institut de Physique Nucléaire d’Orsay, page 247.


[21] Python scripts, The python3 programs used are not freely available but can be requested at SCKCEN.


10 Appendix

10.1 Spallation, fragmentation, fission in more detail.

Figure A1: Production simulated using FLUKA\cite{18} for 1 GeV protons. The production areas are marked based on the production process.

The first problem with the terms spallation and fragmentation is that they are not exactly defined. This is not a problem when far from the definition but in other cases debate can be had about whether or not a process falls into one of those two or another process.

- Fission:
  In the case of fission a particle is absorbed by the nucleus and forms a new nuclei before fissioning. Upon fissioning in most cases 2 daughter nuclei
are formed close in mass-number and neutrons are released. In 1 in 250 to 500 events ternary fission happens where 3 daughter nuclei are created while in 1 in a billion events quaternary fission will happen resulting in 4 daughter nuclei. Due to the nature of this process the fission products will contain an excess of neutrons which will put most of them below the stability line meaning that most of the decays will be $\beta^-$-decay.

- **Fragmentation:**
  In the case of fragmentation the incident particle breaks away from the larger nucleus. This will lead to a small fragment remaining (as can be seen in figure A1 in purple) and neutrons and protons will be released as well.

- **Spallation:** With spallation a highly energised particle enters the nuclei where the high excitation of the particle will cause $\alpha$-particles and neutrons to be evaporated from the nucleus. Due to this there will be a shortage of neutrons in the nuclei putting these isotopes above the stability line where the decays will be dominated by $\beta^+$-decay.
### 10.2 Model matrices

$$
\begin{pmatrix}
\text{Discs} \\
\text{Target container} \\
\text{Extraction station} \\
\text{Mass separator} \\
\text{Front end} \\
\text{Collimators} \\
\text{Casserole} \\
\text{Decay tank}
\end{pmatrix}
\begin{pmatrix}
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V_H \\
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0 \\
0 \\
0 \\
0 \\
0 \\
0
\end{pmatrix}
\begin{pmatrix}
D - M_{TC} \\
D - M_{TC} F \\
0.95 e M_{TC} F \\
A(1 - \epsilon) M_{TC} \\
0.01 e M_{TC} \\
(1 - A)(1 - \epsilon) M_{TC} \\
0 \\
0
\end{pmatrix}
= 
\begin{pmatrix}
D - V_C \\
V_C \\
D - U_{ext} \\
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0 \\
0 \\
0 \\
D - V_C \\
D - U_{FE} \\
V_C \\
D - V_C \\
D - V_C \\
D - V_C \\
0 \\
0 \\
0 \\
0
\end{pmatrix} \begin{pmatrix}
\text{Discs} \\
\text{Target container} \\
\text{Extraction station} \\
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\text{Front end} \\
\text{Collimators} \\
\text{Casserole} \\
\text{Decay tank}
\end{pmatrix}
+ 
\begin{pmatrix}
\tilde{P}
\end{pmatrix}
\begin{pmatrix}
D - V_H \\
V_H \\
0 \\
0 \\
0 \\
0 \\
0 \\
0 \\
D - V_C \\
V_C \\
D - U_{ext} \\
D - U_{FE} \\
V_C \\
M_{cass} \\
D - M_{cass} \\
0 \\
0 \\
0 \\
0
\end{pmatrix}
\begin{pmatrix}
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\text{Extraction station} \\
\text{Mass separator} \\
\text{Front end} \\
\text{Collimators} \\
\text{Casserole} \\
\text{Decay tank}
\end{pmatrix}
$$

(A1)

$$
\begin{pmatrix}
\text{Discs} \\
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\text{Mass separator} \\
\text{Front end} \\
\text{Collimators} \\
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\text{Decay tank}
\end{pmatrix}
\begin{pmatrix}
D - V_C \\
V_C \\
D - U_{ext} \\
0 \\
0 \\
0 \\
0 \\
0
\end{pmatrix}
= 
\begin{pmatrix}
D - V_C \\
V_C \\
D - U_{ext} \\
0 \\
0 \\
0 \\
0 \\
0
\end{pmatrix} \begin{pmatrix}
\text{Discs} \\
\text{Target container} \\
\text{Extraction station} \\
\text{Mass separator} \\
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\text{Casserole} \\
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(A2)
10.3  **Python packages**

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</tr>
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<td>decimal</td>
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<td>Overcoming the problem with float numbers rounding wrong when subtracted from larger number.</td>
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</table>

Table A1: Extra packages used. All packages were found through the ubuntu 16.04 pip repository.

10.4  **Volatility group inventories**

10.4.1  **$^{141}$Xe model data**

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Table A2: Number of $^{141}$Xe atoms found in the different volumes for different iterations. All are extracted from the last data point after a 28 day run which is turned on. Volumes are discs, target vacuum, extraction station, mass separator, transport tube, collimator, casserole and decay tank.
### 10.4.2 $^{94}$Rb model data

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Table A3: Number of $^{94}$Rb atoms found in the different volumes for different iterations. All are extracted from the last data point after a 28 day run which is turned on. Volumes are discs, target vacuum, extraction station, mass separator, transport tube, collimator, casserole and decay tank.

### 10.4.3 $^{114}$In model data

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Table A4: Number of $^{114}$In atoms found in the different volumes for different iterations. All are extracted from the last data point after a 28 day run which is turned on. Volumes are discs, target vacuum, extraction station, mass separator, transport tube, collimator, casserole and decay tank.
10.4.4 $^{225}$Ac model data

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</tbody>
</table>

Table A5: Number of $^{225}$Ac atoms found in the different volumes for different iterations. All are extracted from the last data point after a 28 day run which is turned on. Volumes are discs, target vacuum, extraction station, mass separator, transport tube, collimator, casserole and decay tank.
10.5 Different targets

Figure A2: The production of isotopes in the pure thorium target per proton. The production is visualised using a base 10 logarithm and smaller white dots are used to visualise the stability line.
Figure A3: The production of isotopes in the pure uranium target per proton. The production is visualised using a base 10 logarithm and smaller white dots are used to visualise the stability line.
Figure A4: The production of isotopes in the uranium carbide target per proton. The production is visualised using a base 10 logarithm and smaller white dots are used to visualise the stability line.

As shown in figure A2, A3, A4 and 18 the production becomes more concentrated around the centre of the fission products in cases of no carbides in the target. This is most likely caused by less atoms being present in the target which means the proton energy will be more discrete and thus a more select group of isotopes will be formed. While there are differences between the uranium and thorium targets these are most noticed around the higher Z-numbered isotopes.