Calculating the interdependency of the temperature and power production inside the Mo-99 research loop

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Abstract

Nuclear medicine procedures are fulfilling a key role in the modern day medical world. In 80% of all procedures $^{99m}$Tc is used. The “mother” of this isotope is Mo-99, which is the main subject of this thesis. At this moment, only 5 nuclear plants in the world are used to create Molybdenum and all of these are at least 43 years old. This thesis investigates the production of Mo-99 in the Hoger Onderwijs Reactor (HOR) at the TU Delft. This would be done by building a continuous loop near the core of the reactor, through which uranium salt flows slowly. Earlier research has been done about this subject by Elgin [1] and Huisman [2]. Elgin calculated the production of Mo-99 in the loop, considering how much fission reactions are started. The program used for this was Serpent, a Monte-Carlo based program. Based on this an advise for the concentration of uranium inside the salt was formed. This research was followed by Huisman, who studied the heat production caused by fission inside this loop. Huisman concluded the concentration found by Elgin would result in operating temperatures too high for the uranium salt. The next steps in this process would be to combine their results and construct a new advise for the operation conditions of the Mo-99 loop. Important to notice is Huisman had used a different program, MCNP, for producing his power input. This model was assumed to be correct and used as a benchmark for the Serpent model.

The main goal for this research is investigating the interdependency between the temperature and the power production inside the Mo-99 loop. As the cross sections change when the temperature rises, it is important to know this dependency so the stability of the system can be calculated. For this to be calculated, the models in Serpent and COMSOL have to be connected. First the results of the Serpent model were matched with the results for MCNP. Detectors were implemented in Serpent to recreate the power distribution found in MCNP. In the results the neutron flux in MCNP and Serpent were equal, however the produced power differed by a factor 1/2. It was discovered this could be because MCNP had used a different density for the uranyl salt, approximately differing by a factor of 2. The Serpent model was used for the further research. Here after the connection between Serpent and COMSOL was made. The output of each model were connected with Matlab models. The connected model was used for investigating the temperature dependency of the power production given by Serpent. The conclusion was that the research loop is stable, meaning that when the temperature rises the power production inside the loop becomes smaller. This means the temperature will be lower again. The influence of the temperature was found to be very small. This is what would be expected according to the theory. At last the new advised concentration of uranium in the salt was calculated for which the temperature does not exceed a 100 degrees Celsius. This was found to be 30 g/L, when using the power output of the Serpent model and calculating the temperature with COMSOL.
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Chapter 1

Introduction

1.1 Molybdenum-99

Nuclear medicine procedures are fulfilling a key role in the modern day medical world. Isotopes are linked to specific chemical compounds, after which physiological processes can be examined in for example the heart, liver or kidneys. The requirements for these isotopes are specific. They have to emit a gamma ray which is strong enough to pass through body. Moreover, their half-life must be long enough to prepare the equipment, yet short enough for it to decay during the procedure. The isotope which meets the requirements the best is Technetium-99m. This is used for 80% of all the procedures, which represents 30-40 million examinations worldwide every year. As Technetium has a half-life of only 6 hours, it is ineffective to be transported. That's why often its "mother", $^{99}$Molybdenum is considered. Technetium can be extracted from Mo-99 in special $^{99}$Mo/$^{99m}$Tc generators. This whole production process of $^{99m}$Tc is schematically shown in figure 1.1.[3]

![Figure 1.1: Mo-99 supply chain](image)

At this moment, there are only 5 nuclear plants in the world which are used to create Mo-99. As all of them are at least 43 years old, these reactors are more prone to get shutdown, either planned or unplanned. All of these reactors also run on Highly Enriched Uranium (HEU). After one of the reactors shut down in 2010 for about half a year, the vulnerability of this production was highlighted. This led to several researches by organisations such as the NEA (Nuclear Energy Agency) and OECD (Organisation for Economic Co-operation and Development), investigating the future demands [4] and other production methods. [5] One of the "short-term" solutions suggested in [5] is the production of 99-Mo by uranium fission in already existing research reactors, using Low Enriched Uranium (LEU).

1.2 OYSTER

The HOR is the Hoger Onderwijs Reactor in Delft. It is an open pool-type research reactor, which uses Low Enriched Uranium-235 (LEU) as a fuel. The reactor is not designed to generate energy. It mainly provides neutrons for various research facilities at the Reactor Instituut Delft (RID). Two large beam tubes are for example equipped with stacked-mirror neutron guides, to obtain clean beams of thermal neutrons with low contamination by fast neutrons and gamma rays. This set up provides a unique chance for scientists at the TU Delft to perform a variety of research on various systems and materials. The RID department always tries to make sure their latest researches stay up to date. A good example of this is the OYSTER project. (Optimized Yield -for
Science, Technology and Education - of Radiation) This is a programme to expand the potential of the research reactor by improving the reactor, the instruments and the facilities of the RID. Instruments which will be added include for example an Cold Neutron Source, a neutron powder diffractometer and a positron annihilation lifetime spectroscope. These instruments will be placed in a building next to the reactor as shown in figure 1.2. The reactor is also used for implementing new ideas on a small scale. Measurements can be done with these new installations which can verify the models created for this problem. When these models can predict the measurements correctly, it can be fairly safely said the physics which occur are understood. Which means the installation can be implemented on a bigger scale. This is where the Molybdenum-99 production loop comes into the picture.

1.3 Producing Molybdenum-99 inside the HOR

In this report will be elaborated on the idea of producing Molybdenum-99 in an already existing (research) reactor. The NEA and OECD decided these production plants should use Low Enriched Uranium (LEU) instead of HEU, because it has the following advantages.

- Better proliferation-resistance. When a nuclear system is proliferation-resistant, its deployment and use would not significantly increase the probability of proliferation of nuclear weapons. LEU can not be used for production of nuclear weapons, therefore it is more proliferation resistant.

- Easier availability of the target material.

- Easier access for target transportation and processing.

The only disadvantage LEU has compared to HEU is that the production yield will be lower, which means the total produced Mo-99 will be lower. However, the density of the LEU in the salt can be increased, causing an equal output. This will be discussed later in the report. For the uranium salt uranyl nitrate will be used. The reason for this can be read in [1]. As the HOR is a small reactor, the production yield in this reactor is not expected to be high. If the implementation of the Mo-99 is successful, the project can be upscaled and build
in the reactor in Petten, the Netherlands.

Something else which is also important to mention is the plans of the facility also includes the extraction of Mo-99 out of the uranium salt immediately as it leaves the reactor. Thus merging the first and second factories of figure 1.1. Because the Molybdenum has a half-life of 66 hours, the fact that less transportation of the salt is needed is projected to seriously increase of production. However research will have to be done regarding that subject.

This report will further build on two researches by Kenneth Elgin [1] and Jurriaan Huisman [2]. Because a good understanding of their reports is needed for the following of this report, their results will now shortly be discussed before outlining the goals of this report.

1.4 Results of the research done by Elgin

So far Elgin [1] has studied the feasibility of the loop by focussing on the total production of Molybdenum. The computer program which was used for this is called Serpent. Serpent is a Monte Carlo program which is well-known and often-used in the nuclear physics world. A deeper explanation of this program will be given in chapter 4. After studying the blueprint of the HOR, Elgin decided the production of Molybdenum could be realised inside an aluminium tube which already exists. This outside tube is called the DLDR tube. Inside this tube another U-shaped tube will be build, functioning as inlet and outlet of the uranium salt. In figure 1.3 the U-tube consists of the sections 1. As Elgin tried to maximize the Molybdenum production, it was concluded the concentration should be as high as possible and irradiation time as long as possible. The limiting factor would be the extraction plant which filters the Mo-99 out of the uranium salt. The recommended concentration was 310 g/L uranium in LEU. The irradiation time was calculated on 3 hours. Elgin however did not take heat production into consideration.

1.5 Results of the research done by Huisman

Huisman [2] followed up Elgin and determined the operating conditions which would result in a safe operating temperature of the research loop. A condition which was demanded was that the temperature of the uranium salt inside the loop could not exceed a 100 degrees Celsius as the salt would start to boil. When Huisman started his project however, the original design of the tube which Elgin used had already been changed. An additional safety tube was added. This makes sure in case the inner tube brakes, no radioactive salt or heavy water can enter the water surrounding the core. The safety tube creates section 2 in figure 1.3. For the calculation of the temperature, COMSOL was used. COMSOL is multi-physics program capable of doing various physics related calculations, using Finite Element Methods for solving them. The power input Huisman used was accumulated by August Winkelman in MCNP. This also is a Monte Carlo driven program. Huisman did not use Serpent for this because Elgin had focussed on the production of the Molybdenum, not the power generated inside the loop. Huisman concluded the 310 g/L concentration of uranium would cause a temperatures far above 100 degrees Celsius. While assuming a linear relation between the concentration and the power produced inside the loop, a concentration around 30 g/L was advised. Additionally it concluded that water should be added in the safety tube and DLDR tube, for a better heat transfer thus lowering the temperature. In the discussion of [2], Huisman states the next step in the development of the Molybdenum-99 loop would be an integral research combining both MCNP and COMSOL calculations.

1.6 Goals

The main scope of this thesis will to investigate the coupling between the increasing temperature and the power production inside the Mo-99 loop. To be able to calculate this, first a connection between Serpent and COMSOL will have to be created. As both models now function independently, a coupled system is desired. A system which will be able to predict the Mo-99 production in combination with the temperature profile when changing parameters.
Once this connected system is created, the influence of the increasing temperature in the loop on the power is calculated. As the rising temperature has an influence on several parameters, which will be discussed in this thesis.

The secondary aim will be to once again assess the feasibility of the Molybdenum-99 loop inside the reactor as Elgin did. However, in this thesis the heat production inside the loop will be taken into consideration. By combining the results of both models, the conditions for the uranium salt can be recalculated.

1.7 Outline

This thesis will have the following outline. First in order to understand the physics behind the production loop and each model, chapter 32 will be a theoretical background. This includes important formulas and physics behind Serpent and COMSOL models. Once this basic knowledge has been explained, the adjustments to each model will be discussed and the reason why these were made in chapter 3. After this the results from combining the models are given in chapter 4. Chapter 5 will provide the conclusion and discussion.
Chapter 2

Theoretical background

2.1 Nuclear physics

To be able to understand the methods Serpent uses and the output it generates, a basic understanding of nuclear reactor physics is needed. The following section will further expand on this subject. Starting with the basics and eventually touching on how the temperature has an influence on several variables in the reactor.

2.1.1 Uranium concentration

In this thesis will often be talked about the uranium concentration in the salt. It is important to notice that this is not the enrichment. The enrichment is the ratio between U-235 and U-238. The concentration of the uranium salt determines how much gram uranium the salt contains per liter. For the exact calculations of the atom densities of the elements in the salt read [1].

2.1.2 Cross sections

In a nuclear reactor heavy nuclei, for example uranium or plutonium, are split into two fragments by neutrons. With this reaction, a large amount of energy (circa 200 MeV) is released. The amount of reactions taking place is influenced by several variables. The output of Serpent is mostly influenced by the cross sections. When a neutron reacts with the nucleus of U-235, energy is released, but the fission reaction also releases a few neutrons which can then go on to induce more fission reactions. If every neutron would start another reaction, the chain reaction would quickly grow out of bounds. This doesn’t happen because the neutron doesn’t initiate fission every time it interacts with a nucleus. The reactions a neutron can have are summarized by 3 cases.

1. Nuclear fission.

\[ ^{1}n + ^{A_{1}}Z_{1}X \rightarrow ^{A_{2}}Z_{2}X + ^{A_{3}}Z_{3}X + \text{neutrons + 200MeV} \] (2.1)

In this $^{A_{1}}Z_{1}X$ is the heavy nucleus which captures a neutron. $^{A_{2}}Z_{2}X$ and $^{A_{3}}Z_{3}X$ are the two atoms which are created. This is the reaction we are most concerned with. The nucleus captures a neutron and will fission in two other, lighter atoms, releasing on average around 200 MeV of energy and on average 2.5 neutrons. The isotopes created are described by the so-called camel curve, which shows the probability each atom is created. Because there are a lot of atoms and reactions in a material, the laws of statistics tell us the atoms produced will pretty much accurately follow this curve, even though the y-axis indicates a certain probability. As Molybdenum has a mass number of 99, it can be seen that for 6% of the fission reactions Molybdenum is created. However the most Mo-99 is produced by the decay of other fission products. [7]
CHAPTER 2. THEORETICAL BACKGROUND

FIGURE 2.1: The thermal production yield of Plutonium-239.

There is symmetry in the graph because the sum of the mass numbers has to stay equal as there is a conservation of the number of nucleons involved in the reaction and a conservation of total charge.

2. Radiative capture.

\[ ^1_0 n + ^A_Z X \rightarrow ^{A+1}_Z X + \gamma \] (2.2)

In radiative capture, the nucleus will capture the neutron and may eventually emit a high energy photon, a gamma ray. The energy of these gamma rays are not negligible and will also be taken in account in the calculation as will follow later.

3. Scattering.

The third reaction of importance is scattering. In this reactions the neutron simply scatters of the nucleus. If the reaction is elastic, both the neutron and the nucleus stay intact. When the reaction is inelastic, they will form a compound nucleus for a short time before the neutron is re-emitted along with some gamma ray's. The emitted gamma ray's are the lost kinetic energy from the inelastic collision.

It is important to know the probability's for every one of these interactions happening. These probabilities are expressed in terms of a parameter known as the nuclear microscopic cross section. This is very much compatible with a classic cross section, as it also has a unit of \([1/cm^2]\). Generally, the microscopic cross section is expressed in units of \(10^{-24} cm^2\), which from now on will be called 1 barn. The value of the microscopic cross section can differ between a few barn or even 10,000 barns. Every kind of reaction before mentioned has a different cross section. The total cross section for one atom is the summation as stated in the following equation.

\[ \sigma_t = \sigma_f + \sigma_c + \sigma_s \] (2.3)

In which the f stands for fission, the c for capture and the s for scattering.

The macroscopic cross section is now defined. The macroscopic cross section is the property of a material instead of one atom. It is defined as the total microscopic cross section of every atom multiplied by the atom density of every atom in the material. So for example for water the macroscopic cross section is calculated as follows:

\[ \Sigma_w = N_H \sigma_H + N_O \sigma_O \] (2.4)

Taking in considering the material does not have to be uniform, as well as the energy dependence of the cross section which will be discussed in section 2.1.4, the general equation for the macroscopic cross section is:

\[ \Sigma(r, E, t) = \sum_i N_i(r, t) \sigma_i(E) \] (2.5)

In equation 2.5, the summation i is over the amount of different isotopes in the material. As the microscopic cross sections are available in large cross section libraries, created by experiments, and the atom density is most of the time known in the material, the macroscopic cross section can often be calculated.
### 2.1.3 Flux through a material

The cross sections, together with the neutron flux determine the amount of reactions taking place inside a material. The number of reactions per volume per second can be written as [8]:

\[ R'''' = \sigma I N_A \]  

(2.6)

Here \( \sigma \) represents the cross section of the material, \( I \) the neutron flux of the beam and \( N_A \) the atom density. [8] The microscopic cross section can be considered as the effective ‘target area’ the nuclear represents to the neutron.

![Figure 2.2: Schematic view of a neutron flux colliding with a material with the thickness of 1 atom. The neutron flux is here given the letter I.](image)

Figure 2.2 shows the calculation of the number of reactions for a slab with a thickness of one atom. This is expanded to a thicker layer with more atoms, while assuming no fission or scattering occurs. The intensity of the beam \( I \) becomes a function of \( x \). The nuclear density inside a layer of \( dx \) is equal to \( N_A = N dx \). According to the definition of the microscopic cross section, the decrease of the beam intensity is equal to the reaction rate,

\[ -dI = N\sigma I(x) dx \]  

(2.7)

Integrating this gives the flux as a function of the position in the material:

\[ I(x) = I_0 e^{-N\sigma x} = I_0 e^{-\Sigma x} \]  

(2.8)

This means the neutron flux decays exponentially through a material.

### 2.1.4 Energy dependence

The classical comparison between the cross section is broken when taking in consideration that neutrons can have different energies. And as was visible in equation 2.5, the cross sections are dependent on the energy. The relation between the cross section for capture and the neutron energy is shown in figure 2.3.
In figure 2.3 the difference between fast and thermal neutrons can clearly be seen. The neutrons released with fission are in the blue area. Keep in mind the axis are logarithmic, so the cross sections for the thermal neutrons are on average 100x bigger. The moderator, often water, around a nuclear reactor slows the fast neutrons down to thermal neutrons, thus creating more fission with U-235 and Pu-239.

The peaks in the cross sections are called the resonance peaks. A peak in figure 2.3 represents an energy for which the kinetic energy of the neutrons is equal to the energy of the nuclei. The different peaks refer to the different quantum mechanic energy levels of the nuclei. Breit and Wigner have derived the following expression for the cross section $\sigma_{na}$ for which a neutron is captured and a particle is emitted. [8]

$$\sigma_{na}(E) = \frac{\lambda^2}{4\pi} \frac{\Gamma_n \Gamma_p}{(E - E_r)^2 + (\Gamma/2)^2}$$ (2.9)

- $\sigma_{na}$ is equal to the microscopic cross section for which an neutron is included in the nucleus and a particle (possibly the same neutron) is emitted.
- $\lambda = \frac{h}{mv}$ is equal to the de Broglie wavelength.
- $\Gamma_n$, $\Gamma_p$ and $\Gamma$ are the partial level width between energies for respectively the neutron, the particle and the total level width.
- $E$ is the kinetic energy
- $E_r$ is the resonance energy

The reason this equation is introduced is because an important quality can be noticed from this equation. When the kinetic energy of the neutron is almost equal to the resonance energy, the cross section will have a sharp peak. This is because $(E - E_r)^2$ will become very small. Equation 2.9 is plotted in figure 2.4 for Plutonium-238. This graph will be used when considering the temperature dependence in the next subsection.
2.1.5 Temperature dependence

A schematic view of the temperature dependence is shown in figure 2.5. [8].

In order to describe the influence of all these factors, a temperature coefficient $\alpha$ is defined.

$$\alpha = \frac{d\rho}{dT}$$  \hspace{1cm} (2.10)

For a finite change, this can be written as follows.

$$\Delta \rho = \alpha \Delta T$$  \hspace{1cm} (2.11)

In which case $\rho$ is equal to the reactivity, defined as

$$\rho = \frac{k - 1}{k}$$  \hspace{1cm} (2.12)

This means if the temperature coefficient for a reactor is positive, the system is unstable. If the temperature coefficient is negative, an increase in temperature will be counteracted by a power decrease. For the Molybdenum-99 tube inside the reactor it is thus important to investigate whether $\alpha$ is negative or positive.

Because this $\alpha$ is dependent on so many different variables, it is hardly ever possible to calculate it analytically for a system. By defining it like this however, some basic principles can be deducted. As the temperature inside a nuclear reactor is not uniform, $\alpha$ can be divided into a spatial dependency regarding the fuel the moderator. This turns equation 2.11 into:
\[ \Delta \rho = \alpha_f \Delta T_f + \alpha_m \Delta T_m \]  

Now focusing on the DLDR-tube, the uranium salt can be seen as a fuel and the water present inside the aluminium and safety tube be seen as a moderator. The temperature coefficient of the uranium salt is determined by the Doppler effect. This is called "the theory of Doppler broadening of neutron absorption resonances". The Doppler effect, discovered by Christian Doppler in as early as 1842, is well known when dealing with sound and light waves. In the 20th century was discovered this effect is also applicable on neutron cross sections. The mathematical calculations behind this are beyond the scope of this project, but can be read in [9]. Instead, only the consequences from this Doppler broadening will be discussed. If the temperature of the uranium salt goes up, the peak in the cross section spectrum will change. This is clearly visible in figure 2.6. The peak will become a smoother function, spreading over the energy, while the surface beneath it remains the same. In 2.6 the absorption cross section is plotted while applying the Doppler broadening when the temperature rises.

![Figure 2.6: Absorption cross section with the Doppler broadening.](image)

The spreading of the peaks will cause that the probability of absorption in the flanks of the peaks will overlap and increase. When the absorption in the resonance region in $^{238}U$ dominates the fission in $^{235}U$, so for not too high a degree of enrichment, $\alpha_f$ will be negative and the reactivity will decrease with increasing temperature of the nuclear fuel. [8]

The temperature coefficient of the moderator will mostly be influenced by the change of density inside the water. [8], chapter 6.6, states the temperature coefficient of the moderator can be both positive and negative, depending on the amount of moderators in the core. A core has to be under-moderated for the temperature coefficient to be negative. It is not yet known if this will be the case. In this thesis the influence of the rising temperature of the water (or moderator) inside domain 2 and 3 (figure 1.3) will not be calculated. This will be for further research. With the numerical method only the $\alpha$ of the fuel of equation 2.10 will be investigated.

Because the uranium salt in the tube will be LEU, the expectations are the $\alpha$-coefficient will be negative and the power will slightly decrease.

### 2.2 Bateman equations

In the next section the Bateman equations will be discussed. These equation describe the composition of the salt after some time has passed and reactions have taken place. Considering an important output of Serpent is the amount of Molybdenum-99 produced, it is important to have a basic understanding of the way Serpent calculates this.
2.2.1 Burnup calculations

While the reactor is running and the salt is irradiated with neutrons, the composition of the salt will change. This is called the burnup of the uranium salt. The general equation for the decay of an isotope in the salt can be constructed as the following equations:

\[
\frac{dN_i}{dt} = -\lambda_i N_i 
\]  
(2.14)

\[
\frac{dN_i}{dt} = \lambda_{i-1} N_{i-1} - \lambda_i N_i 
\]  
(2.15)

Where \( \lambda_i \) is the decay constant of the \( i \)th nuclide. Equation 2.14 and 2.15 show the amount of atoms present of a certain isotope are equal to the amount produced by decay of other, higher isotopes minus the decay of the isotope itself. These equations are for a simple decay of one kind of atom.

When considering the uranium salt in the tube, the Bateman equations will have the following form. [1]

\[
\frac{dN_j}{dt} = \sum_{i \neq j} \left[ \gamma_{i \rightarrow j} \sigma_{f,i} \Phi + \lambda_{i \rightarrow j} + \sigma_{i \rightarrow j} \Phi \right] N_i - \left( \lambda_j + \sigma_j \Phi \right) N_j 
\]  
(2.16)

- \( N_j \) is the atomic density of isotope \( j \).
- \( \gamma_{i \rightarrow j} \) is the fraction of the fission product yield of \( j \) by the fission of isotope \( i \). This can for example be read out of figure of the camel curve. 2.1.
- \( \sigma_{f,i} \) is the microscopic cross section for fission of isotope \( i \).
- \( \Phi \) is the neutron flux in the material
- \( \lambda_{i \rightarrow j} \) is the decay constant of isotope \( i \) to \( j \) decay.
- \( \sigma_{i \rightarrow j} \) is the microscopic transmutation cross section of the isotope \( i \) to \( j \).
- \( N_i \) is the atomic density of isotope \( i \) item \( \sigma_j \) is the microscopic transmutation cross section of isotope \( j \)

In this equation the depletion term is also taken in account. This term is summarized over \( i \) isotopes which can create the isotope \( j \). The first term within the summation of equation 2.16 represents the number of isotopes from \( j \) produced by fission, according the the camel curve shown in figure 2.1. The second term represents amount of isotopes produced by the decay from an isotope \( i \) to \( j \). The last term is the transmutation cross section, which sums up all the transmutations from \( i \) for which \( j \) is formed. The part after the minus in equation 2.16 is composed of the decay of \( j \) and the cross section for \( j \) starting a reaction with a neutron. For complicated materials and long irradiation time, these equations can form huge matrices for each isotope, all dependent on each other. Serpent is able to solve these matrices.

2.3 Heat production inside the tube

In this section the physics behind the general heat production inside the tube will shortly be discussed. As Huisman did an extended analysis, only the calculation method by COMSOL will be briefly discussed. However for the extended version please read [2] chapter 3 as well as [10], chapter 3.

Akker [10] derived the general equations for a small volume cubic volume as in figure 2.7.

Figure 2.7: Small cubic volume
The equation for the change of temperature inside such a volume was equal to equation
\[
\frac{\partial \rho c_p T}{\partial t} + \rho v_x \frac{\partial c_p T}{\partial x} + \rho v_y \frac{\partial c_p T}{\partial y} + \rho v_z \frac{\partial c_p T}{\partial z} = \lambda \left( \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} \right) + q
\] (2.17)

q is in this case the power generated inside this cube. The derivation of this formula can also be done for the transfer of mass and the transfer of impulse. The transfer of mass, transfer of impulse in combination with equation 2.17 form 3 which equations HAVE to apply for every volume, however most of the time they are either too complicated to solve or not enough boundary values are known.

These are also the equations COMSOL solves. In COMSOL the geometry is divided into small volumes using a mesh. COMSOL then solves equation 2.17 for every volume. COMSOL starts with the volumes for which boundary conditions are specified. The output is then used as the boundary input for the neighbouring volume. Equation 2.17 can also be derived for more complex shapes such as a tetrahedral or a triangle, however this will not be shown here.

2.3.1 Fission

The power within the volume of figure 2.7 is generated by the fission reactions with in that volume. Serpent is capable of calculating the energy released for every reactions which took place. This distribution will be shown in chapter 4.

2.3.2 Gamma heating

Another big contributor to the heat production inside the loop is gamma heating. This phenomenon is caused by the interaction between gamma particles with the construction materials in the setup. These reactions increase the material temperature. Within the HOR development the thumb rule of approximately 0.3 W/g is used. Huisman determined the total power distributed within the material as 325 W. This is also taken in account in the COMSOL model.
Chapter 3

Numerical methods

Both Elgin as Huisman were doing their own independent researches, with different goals in mind. Elgin focussed on calculating the Molybdenum-99 produced in the loop. Huisman after this used the input from another computer program, MCNP, as power production for the calculation of the temperature in the loop. For these models to be combined, changes have to be implemented in each model. This next chapter will first elaborate on the methods these models use. After this the changes made will be discussed. In chapter 4, the results of changes will be validated.

3.1 Serpent

3.1.1 Introducing Serpent

Serpent uses the mathematical Monte-Carlo method. A Monte Carlo simulation is often used when the results in a model are dependent of stochastic processes. It is a brute force calculation method, which solves problems by simulating a large amount of different solutions. In this case, the Monte Carlo method generates neutrons with a random speed (or energy) and direction. In Serpent the geometry of the HOR is created. Serpent then uses the cross sections of every material to calculate with a certain probability what happens with every neutron which is generated. When taking more neutrons, the predictions will get more accurate. However it will also take more time to compute the result. Because computing the actual amount of neutrons produced in the reactor would take too long, Serpent is able to normalise its results to a certain power. The power of the HOR is set to 2 MW. [11] Serpent uses huge libraries with cross sections, fission product yield, thermal scattering data and more for every atom at different temperatures, called the ENDF libraries. (Evaluated Nuclear Data File).

3.1.2 HOR in Serpent

Serpent is a code which cannot simulate flows. Because of this, a flow of the salt was mimicked by creating 12 sections in the tube. These sections are only present in the code, not in the real tube. The time after which Serpent gives the output is set to 15 minutes. This means Serpent gives the composition of the salt when the reactor has irradiated it for 15 minutes. After the first calculation the composition in each section is transferred to the next section with a script written in Matlab. This means after 12 calculations by both Serpent and Matlab, the inventory in section 12 contains the composition of the salt "flowing" out of the tube. This salt has then been in the reactor for 15x12 = 300 minutes, or 3 hours. For further details, see [1]. Some important constants for the tube are:

- The length from B to B in figure 3.2 is 1.6 meter.
- \( r = 2.2 \) for domain 1, the inner tube containing the uranium salt. The thickness of the tube is 0.2 cm, for a total of 2.4 cm.
- \( r = 5.4 \), with a thickness of 0.3 cm for domain 2, the safety tube.
- \( r = 7 \) with a thickness of 0.5 cm for domain 3, the outer tube.

As every calculation runs about 30 minutes in Serpent, Elgin selected 12 sections for a good mixture between reasonable results and computational time. His code was fixed on calculating
Figure 3.2: Cross section of the DLDR tube in the YZ-plane. The 12 sections inside the tube are clearly visible. The uranium salt 'flows' through the green parts. The arrows indicate the movement of the salt.

how much Molybdenum was produced and he examined several geometry's of the tube and different concentrations of uranium in the salt.

Figures 3.3 and 3.4 show two views of the geometry created in Serpent.

Figure 3.3: Top view of the geometry. The raster on the left is the reactor core. Each color stands for a different amount of uranium.

Figure 3.4: Side view of the geometry. The fuel rods and the Mo-99 both clearly visible. The safety tube has already been build in.
3.1.3 Adjustments in Serpent

The first thing which had to be done was reproduce the results in Serpent which Huisman had used as input. These were accumulated in MCNP by August Winkelman. As some changes were made in the design of the tube since Elgin had done his research, some changes had to be made in the Serpent code. The following adjustments were made to the model created by Elgin [1].

1. The main thing was the addition of the safety tube, which is already visible in figure 3.3. Because of this tube, the radius of the pipe through which the uranium salt flows changes from 3.3 to 2.2 centimeter [1]. The new tube is already visible in figure 3.3. As a consequence the mass flow through the pipe reduces, which means less Molybdenum is produced.

2. In the compartments around the flow water was added. This was a result from Huisman [2] as this was necessary for sufficient heat transfer out of the inner tube. This should have an influence on the neutron flux through the uranium salt, as water has a bigger cross section than air, thus moderating more neutrons. However because the compartments are small, the influence is expected to be minimal.

3. The inner tube was constructed of a zirconium alloy. Zirconium alloys are common when construction a nuclear reactor, as it has a high tolerance for heat as well as relatively small cross section for the neutrons. As a result neutrons pass through easily. This however does lower the neutron flux a little bit.

The impact on the Molybdenum production by these changes will be shown in the results.

3.1.4 Adding detectors

To be able to recreate the output of MCNP, Serpent also has to generate a power output. For this, detectors are used. Detectors are non-physical elements which can be plotted in Serpent and they do not influence the results. They are solely used for calculations. These detectors are used for the calculation of the power generated inside the loop. Detectors in Serpent work along the following formula.

\[
R'' = \frac{1}{V} \int \int \int f(r, E) \phi(r, E) d^3r dE
\]  

(3.1)

In this equation \( f(r, E) \) is the input value that can be set in the script. When calculating the flux in a certain volume, \( f(r, E) \) will be specified by Serpent as 1. Important to notice is that every assigned function \( f(r, E) \) is multiplied by the flux through the volume. The option selected in Serpent is calculating the total macroscopic fission cross section. This would mean \( f(r, E) = \sum_i N_i(r, t) \sigma_i(E) \) as in equation 2.5. With \( \sigma_i \) as the total fission cross section, as only the fission reactions have to be calculated. Because this is automatically multiplied by the flux and the volume is not specified, according to equation (3.1) the output will be total fission reactions per volume according to equation 2.6.

As the input which is recreated is a power along the x-axis, see figure 4.3, a grid of detectors is implemented along the x-axis. These detectors are constructed such that they only register the neutrons going through the uranium salt. The grid consists of 200 volumes. The measurements from each individual grid combined will create the total power function. The power produced is then calculated by multiplying this output with the volume of such a grid volume. As well as multiplying it by 200 MeV, the average power produced in a fission reaction, and converting to Joules.

3.1.5 Doppler broadening

In the ENDF cross section libraries Serpent uses, the cross sections for each isotope are given for temperatures of 300, 600, 900, 1200 and 1500 Kelvin. Serpent chose these temperatures as a good fit between accurate calculations and storage space, as these libraries contain a lot of data. The only way Serpent is able to take temperature in account between each of these temperatures is by using Doppler broadening. Serpent uses the physical relations and math described in [12], chapter 4. This theory will not be elaborated on. The actual cross sections for a given temperature are approached using the Doppler broadening. The cross section is always interpolated to the lower known cross section because this is the way the formulas for the Doppler broadening in [12] have been derived. So for all the temperatures under 600 K, the Doppler broadening will interppolate to 300 Kelvin. Because our temperature is relatively close to 300 Kelvin the results will be trustworthy.
3.2 COMSOL

COMSOL is a multi-physics programme capable of performing various calculations with complicated physics problems. For the model of the tube, Huisman used the heat transfer interface. As input for COMSOL Huisman needed a function of the power production inside the tube. This power production is present because of the energy released with every fission reaction in the tube. This power input was accumulated for Huisman by August Winkelman, using a programme called MCNP (Monte Carlo N-Particle) Transport code. This program also uses the Monte Carlo method for calculations, so the results should match between the programs.

3.2.1 Changes made to create the connection

The adjustments made to the COMSOL model created by Huisman were necessary for the connection between Serpent and COMSOL. Because the model took approximately 8 hours to run, a fresh look was taken at the mesh used. This will be specified in section 3.2.2. The following adjustments were made:

- The temperature dependence of Serpent can only be implemented by assigning a temperature to each section of the tube. This means the output from COMSOL should give 12 average temperatures for each section. However COMSOL commands work with lines, surfaces and domains. For the program to be able to calculate the desired temperatures, the tube in COMSOL also had to be divided into 12 sections, while preserving the results.
- Once this was done, COMSOL was assigned to calculate the average temperature over each separate section and import this to Matlab. Another Matlab script is used to import these temperatures and add them to the Serpent script.
- The third and biggest implementation added to the COMSOL model was a different mesh. The next section will further explain what has been done.

3.2.2 Applying a different mesh

The model constructed by Huisman used an automatically created physics-controlled mesh. As the program had to run for almost 8 hours because of this mesh, a different, user created mesh has been build. This new mesh follows the blueprint as explained in these articles written by COMSOL. [13] [14] The changes applied are now further discussed. For the new mesh, something which is called ‘partitioning of the geometry’ is used, as well as a swept mesh. When the geometry is partitioned, the complex parts of a large domain are separated from the domain. Because of the complexity of these parts, these new domains can be assessed to a finer mesh while the other domains use a more coarse mesh. In the geometry of the tube, the U-turn has the most difficult shape. This part of the geometry is assigned the tetrahedral mesh, a fine mesh. Recognizing that the flow field in the long straight sections will vary quite gradually along the axis, for this part the swept mesh is set. In these parts, a relatively coarse mesh can be used. In the results will follow wether or not these big meshes are justifiable, by comparing the new results with the results from Huisman.

![Figure 3.5: Physics controlled grid, automatically created by COMSOL.](image1)

![Figure 3.6: New grid constructed](image2)

In figure 3.5 and 3.6 the difference in the meshes between the model created by Huisman and the new model are visible for a small part of the geometry. These changes greatly reduce the number of equations COMSOL has to solve, hereby speeding up the calculation time. The results of the programme with the new mesh will be compared with the old results to verify whether the new mesh qualifies.
3.3 Forming the connection

For the connection to be made, the power output from the Serpent model will have to be linked to COMSOL, as well as the temperature output from COMSOL to Serpent. Serpent uses text files as input and generates Matlab and text files as output.

In COMSOL the power distribution has to be imported into the interface. COMSOL is able to export the calculated average temperatures of each section into a text file.

This output from COMSOL has to be added to the input text file for Serpent. To create this link Matlab is used as Matlab is suited for both importing the data en creating the new text files. A schematic view of the process is shown in figure 3.7. In Chapter 4.4 will be discussed how this connection was made.

![Schematic view of the coupling between the models.](image)

The Matlab files have to be created. This can be an iterative process. If the temperature coefficient $\alpha$ from equation 2.11 is negative, the iterative process should result in a stationary solution where the temperature and power don't change anymore over time. If $\alpha$ is positive, the temperature will get higher and higher. It is clear a negative $\alpha$ is desired for the Molybdenum-99 loop.
Chapter 4

Results

In this chapter the results from the adjustments to Serpent and COMSOL will be shown. First needs to be confirmed whether the new geometry implemented in Serpent can match the power distribution gathered in MCNP by August Winkelman. If this power distribution is created in Serpent, the COMSOL model has to be adjusted for the coupling to be created. If both of these things are done, the temperature dependence of the power distribution will be calculated. As well as new running conditions for the Mo-99 research loop.

4.1 Changing the geometry

First the changes in production of Mo-99 will be considered, as this was the main output by Elgin. [5] When Elgin did his research the safety tube (domain 2 in figure 1.3) was not implemented yet. When only applying this change, in combination with the new materials for the tubes in the geometry, figure 4.1 shows the difference in the results.

Figure 4.1: Molybdenum production per volume of the new geometry compared to the old geometry.

Figure 4.1 shows the new geometry slightly lowers the Molybdenum production. This seems plausible, as the only things which have been changed for these new calculations are the water in the tube and the zirconium alloy. The smaller tube has its influence on the total production, as this graph shows the Molybdenum produced per volume. When considering the difference in volume of the cylinders, the total becomes

\[
\frac{A_{\text{new}}}{A_{\text{old}}} = \frac{2.2^3}{3.3^3} = 40\% \text{ less.}
\]
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More importantly, figure 4.1 confirms that the new geometry in Serpent still renders an expected output. The new geometry will be used from now on.

4.2 Recreating the power of MCNP

Because of the connection it is desired for Serpent to give the input for COMSOL. That’s why detectors were implemented in Serpent. The results from these calculations are now compared.

The flux and power given by MCNP inside the loop is shown in figures 4.2 and 4.3. As the calculations in MCNP were done for a uranium concentration of $310 \, \text{g/L}$, so were the calculations in Serpent.

These graphs form the benchmark for our Serpent model. At first the thermal flux inside the tube was calculated with Serpent. Thermal neutrons are neutrons with an energy lower than 625 MeV. This would compare to the blue line in figure 4.2.

The flux produced when normalising the power of the reactor in Serpent at 2MW is shown in figure 4.2. As the flux is a stochastic process in Serpent, the collective output of the detectors does not form a smooth line. To create a smooth flux, 2 methods were used. The first method exploits the fact that the detectors measure the flux for every time step. The flux should be almost constant over time on a specified position. Serpent transfers the composition of each section to the next 15 times. Because there are 12 sections, only after 12 time steps the salt in the last section will have passed through the whole tube. Elgin took 15 time steps to investigate whether the output would remain the same at that point. Meaning the output from the last 3 steps should render the same Mo-99 production and power production. This can clearly be seen in figure 4.1. By taking the average of each of the 15 time steps, a smoother line can be created. The second method fits a Poisson function in Matlab against the flux from 1 time step. In figure 4.2 these 3 lines are compared.
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Figure 4.4: Measurements of the flux by Serpent, transformed to a smooth line.

The fitted flux will be the function used from now on. Now the flux and power calculated by Serpent are compared to MCNP.

Figure 4.5: Comparison of the neutron flux density inside loop along the x-direction for Serpent and MCNP.
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In figure 4.5 one can conclude that the results obtained in Serpent compare to the ones generated by MCNP. The small displacement to the right can be explained by the simplification of the core by Elgin in two ways. First the reactor core computed by Elgin is simplified to a raster of 42 fuel rods of uranium. This can be seen in figure 3.4. In the actual reactor a control rod is always surrounded by water as a moderator. In MCNP this is modelled correctly.

Secondly, as Serpent needs a critical reactor for its calculations, Elgin also had to make a further simplification to the core. This also can be seen in figure 3.3. The cells in the core have different colors. Elgin added extra uranium to certain rods to create a stable core. This explains the displacement of the flux.

When comparing the power density in figure 4.6, however, the power differs with a factor 1/2. The calculation by Serpent and MCNP use the same cross section library which means they should gather the same results if the fluxes are equal. This means something has to be different in the composition of the salt.

It was found that the MCNP model used a density of the uranium salt in domain 1 of $1.33 \, \frac{g}{cm^3}$ while for the calculation of Serpent a density of $2.81 \, \frac{g}{cm^3}$ was used. This does seem to be approximately the factor 2 which the calculated powers differ. However an increasing density of the uranium salt, is expected to increase the power. This because there simply is more uranium per liter present in the inner tube. This can not yet be explained. The density of 2.81 was found in the literature by Elgin. This website also confirms the density of $2.81 \, \frac{g}{cm^3}$ [15]. Since the flux does match the results from MCNP and a possible reason for the difference in power has been found, it is concluded the new Serpent model is correct and can be used in the future. The next calculation will all be done with the Serpent model and a density of the uranyl nitrate of $2.81 \, \frac{g}{cm^3}$.

4.3 Adjusting the COMSOL model

In this section the adjustments to the COMSOL model will be discussed. In the model the 12 sections were added and the new mesh was implemented. The original result will be compared to the new results.
These figures show the temperature profiles are very similar. The most important value, i.e. the maximum temperature only differs 2 degrees. This means that the adjusted COMSOL model can safely be used for further research and the temperature output of the 12 sections, needed for the coupling with Serpent, has successfully been created.

### 4.4 Creating the link

The connection of figure 1.4 will be discussed now.
From Serpent to COMSOL
The loop starts by running Serpent without the added temperatures inside the loop. For the link from Serpent to COMSOL Matlab is used. The data from the detectors are fitted to a Gaussian function like in figure 4.2. The y-values of 200 points for this Gaussian function are then exported to a Microsoft Excel file. This file has to be manually imported in COMSOL. COMSOL is then able to create the power distribution function according to this data.

From COMSOL to Serpent
For this connection one must first manually command COMSOL to export the 12 calculated average temperatures in each section to a separate file. The composition of the salt in domain 1 (figure 1.3) is imported via a separate text file in Serpent. A Matlab file is created to extract the temperatures out of the exported COMSOL data and add them to the salt in each section of the text file containing the composition. Serpent then uses the Doppler broadening in each of these sections for a different temperature obtained out of the COMSOL model. After this the connection is complete.

At first the goal was to automatically run the models successively on for example a cluster and calculate several iterations at once. This was not possible, because COMSOL has to be manually instructed to import the power function and export the temperature results to a Matlab model. COMSOL is not able to do this automatically, that’s why one step always has to be done by hand.

Now that a link between the models is created, two more subjects will be investigated before the new advise about the running conditions of the Molybdenum-99 loop is given.

1. The effects of the rising temperature in the DLDR loop on the power input by Serpent.
2. The maximum uranium concentration for which the temperature inside the loop does not exceed a 100 degrees Celsius.

4.4.1 Temperature dependence
First the power function is generated with Serpent, for a uranium concentration of 30 g/L. After this, the temperatures are added in Serpent, using the volume averages calculated in COMSOL. For the temperature calculations the model from Huisman was used which generated a maximum temperature of 90 degrees Celsius. With the temperature dependence implemented, a new power production was calculated. These are compared in figure 4.9.
Figure 4.9: Difference in power when taking the temperature feedback in account.

Figure 4.9 shows the peak in the power production becomes lower. The result is that an rising temperature slightly spreads the power function. The temperature coefficient $\alpha$ is negative. This means the uranium inside the loop is stable. Because the uranium salt contains of LEU, this is what was expected from the literature discussed in section 2.1.5.

### 4.4.2 Uranium concentration

Now that the two models are complete, the maximum concentration of uranium inside the salt can be calculated. Huisman did not have access to a computer program which could calculate the power produced in the salt. When COMSOL showed a concentration of 310 g U/L was too high, a linear relation between the power produced and the uranium concentration in the salt was assumed by Huisman. The maximum concentration of the uranium salt with that assumption was $c = 27.6$ g U/L.

Having a model which can calculate the exact power produced with this concentration and the temperatures reached inside the loop, this can be verified. The maximum concentration of uranium in the salt for which the temperature does not exceed the limit of 100 degrees Celsius is indeed 30 g U/L. For this calculation 1 iteration was done. Note however this is while using the power production as calculated in Serpent.
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Figure 4.10: Plot of the power created. Comparing 310 g U/L with 30 g U/L.

Figure 4.11: Temperature profile for a concentration of 30 g U/L.

Figure 4.10 shows the big difference in the power created for 30 g U/L. However with this concentration, the temperature stays just under 100 degrees Celsius, at 96 degrees. This means 30 g U/L is the absolute maximum concentration for the uranium salt to contain in the set up. With the assumption the density of the uranyl nitrate is $2.8 \text{ g cm}^{-3}$.

4.4.3 Gamma heating

When taking the volume average of the power function in COMSOL it was calculated the total power produced by fission should have a maximum of 287 Watt. The total heat production by gamma heating out of the tubes is 325 Watt [2]. Assuming the concentration used will be taken lower to be on the safe side, this means the gamma heating will be the biggest contributor to the heat production.
Chapter 5

Conclusions and discussion

The main goal of this thesis was to investigate the coupling between the increasing temperature and the power production inside the Mo-99 loop. For this a connection between the models created by Elgin [1] and Huisman [2] was created. The second goal was to revise the feasibility of the Molybdenum-99 research loop. This because in the time passed between these two researches, changes had been made to the design of the loop and the research from Elgin did not take the heat production in account. This research began with implying the same geometry and boundary conditions for each model.

5.1 Changes to the models

Comparing Serpent to MCNP
The model created in Serpent was adapted to match the MCNP results. Detectors were added to measure the flux and power produced inside the loop. The flux differed a little bit because in Serpent the core had been simplified. The power generated by Serpent however was 2 times as small. It was found out MCNP used a different density of the uranyl nitrate than Serpent. This might be the reason for the factor 2. According to literature the Serpent model uses the right density, this means Serpent can be used for the calculation of the power production.

COMSOL
The mesh from the COMSOL model was changed to shorten the computational time. The model was also adapted in such a way the output could function as input for Serpent. The results generated by the new model was compared to the results from Huisman. It was concluded the results did not change and the new model can be used.

Connection between the models
A connection between the models was created. However the connection does require some manual work. It was tried to avoid this, but this was not possible. The reason for this is COMSOL cannot automatically import or export its data, this has to be done manually. Once this connection was made the temperature dependence of the Serpent output was investigated. The results from this was a small spread in the power production while the peak lowers. This means the $\alpha$ coefficient of the reactor is negative for the fuel which is a requirement for the research loop. This is also what was to be expected for the fuel according to the theoretical research.

5.2 New condition for the uranium loop

New conditions for the uranium loop have been investigated while using the requirement that the temperature inside cannot rise above a 100 degrees. The maximum concentration of uranium which can reside in the loop, calculated with the Serpent and COMSOL models would be 30 g/L. With this concentration the maximum temperature is 96 degrees Celsius, which is just below the limit. The recommendation for the research loop would be to create a buffer and use a lower concentration.
5.3 Further research

Although this report gives a fairly good approximation, some other things have not been taken in consideration. Further research for the implementation of the Mo-99 research loop could include one of the following subjects.

First of all, the temperature dependence of the fuel has been calculated. As was suggested in subsection 2.1.5, the heating of the water around the uranium salt will also have an impact on the total temperature coefficient $\alpha$ of the system, because this water acts as a moderator. It seems possible to calculate this in Serpent.

Secondly, for the Molybdenum-99 loop further research would be about the Mo-99 processing facility which will be placed in the building. (See figure 1.1) At this moment this processing facility is the reason why the salt has to stay in the reactor for 3 hours as Elgin found out. If the salt would be able to flow faster through the reactor the concentration and thus the Mo-99 production could become higher.

At last it might be interesting to investigate the implementation of a turbulent flow of the water inside the safety tube. A turbulent flow greatly improves the heat transfer from the uranium salt to the water, practically cooling the salt. If that is possible the concentration of uranium can be increased for a higher Mo-99 production. Important to mention is when this is investigated, the new created mesh might be too big on the edges of the tube, visible in figure 3.6, as the heat transfer to the turbulent flow will be much more difficult to calculate.
Bibliography