

DELFT UNIVERSITY OF TECHNOLOGY

BACHELOR'S THESIS

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THE MPML:
⁹⁹Molybdenum Producing Mini Loop,
driven by natural circulation

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Abstract

Molybdenum-99 is one of the most common medical radioisotopes in the world because it is the nuclear parent of technetium-99m, a radioisotope that is used to diagnose more than 30 million patients each year. The production of molybdenum-99 is done in only five reactors worldwide, all of which are at least 40 years old. Shutdowns of those reactors can cause major shortages in the supply chain. To secure the supply of molybdenum-99, researchers at the Hoger Onderwijs Reactor in Delft are developing a new method to produce this valuable radioisotope: a U-shaped loop that circulates a liquid uranyl nitrate solution, irradiated by the neutron flux of the reactor core. The concentration of this uranyl nitrate solution determines the amount of molybdenum-99 that is produced, and is preferably as high as possible.

The current research loop has two practical drawbacks. The first one is that the flow inside the current research loop is controlled by a pump. If this pump would break down and the circulation would stop, temperatures inside the loop would rise above the boiling temperature of the uranyl nitrate solution. This is potentially very dangerous, since pressure building up inside the research loop could lead to severe damage. The second problem is the volume of the current research loop, which is 5 liters. If 5 liters of uranyl nitrate solution would have to be taken out of the loop in case of an emergency, it would be a too dangerous source of radiation.

The goal of this research is to design a small, molybdenum-99 producing loop with a maximum volume of 0.5 L, that is driven by natural circulation. Moreover, it aims to outperform the current research loop, in which the uranyl nitrate concentration is limited to 27.6 g/L. To investigate the feasibility of the new loop, a simplified mathematical model of the loop was made to calculate the temperatures in the loop for different uranyl nitrate concentrations and cooling conditions, and the maximum uranyl nitrate concentrations that can be reached for different cooling conditions. These calculations were done in MATLAB using the method of iteration.

The results show high potential for the new loop design that is proposed in this research. An uranyl nitrate concentration of up to 250 g/L could be reached, while keeping the temperatures in the loop below a safe 90 °C. This concentration would be nine times higher than that in the current research loop.

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1 | Introduction

1.1 Molybdenum-99

One of the most commonly used medical isotopes in radiology is technetium-99m. Well over 30 million patients worldwide are being diagnosed each year using this isotope [29]. One of the reasons technetium-99m is so popular, is its relatively short half-life of 6.0067 hours [21], allowing for rapid data collection while keeping the exposure for the patient low.

However, the short half-life of technetium-99m also makes it very difficult to transport it around the world. Since plenty of intercontinental transportation takes more than 6 hours, at least 50% of the technetium-99m would be lost on those journeys. To solve this problem, molybdenum-99 is used, the nuclear parent of technetium-99m. Molybdenum-99 has a half-life of around 66 hours [3], making it highly suitable for transportation. Since most transportation around the world can be done in under 24 hours, less than only 25% is lost in those journeys. Therefore, molybdenum-99 is also one of the most common medical radioisotopes in the world.

The production of molybdenum-99 is under high pressure. Due to the enormous demand, and the fact that there are only five reactors worldwide that produce molybdenum-99 on a large scale, shortages often occur [29]. To make things worse, all of those five reactors are at least 40 years old and will soon have to be replaced.

1.2 Current research loop

To find a solution for the inadequate supply chain of molybdenum-99, researchers at the Hoger Onderwijs Reactor (HOR) in Delft have been working on a new setup to produce molybdenum-99. This new setup uses a U-shaped loop that circulates a liquid uranyl nitrate solution, which is irradiated by the neutron flux from the reactor core to produce molybdenum-99. A cross section of this research loop can be found in figure 1.1. The design features, long term behaviour, heat transfer properties and cooling system requirements of this research loop have been explored extensively, and it has been proven feasible by Elgin [12], Huisman [16], Pothoven [24] and Pendse [23]. Collectively they showed that it could be capable of producing up to 2.37 mg of molybdenum-99 per week, which roughly is 2.4% of the world's total demand [23]. However, heat generation and a proper cooling system remain a bottleneck in this research loop.

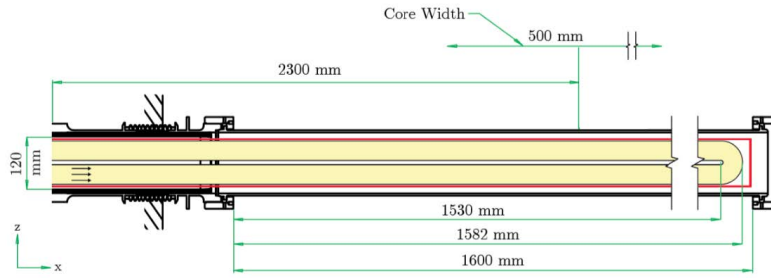


Figure 1.1: Cross section of the current research loop design [24].

1.3 Practical problems of the current loop

Though the current research loop does look promising, it comes with two practical problems. In a nuclear reactor, it is of top priority that the temperatures inside stay beneath $100\text{ }^{\circ}\text{C}$ (or boiling temperature to be exact) at all times, to prevent pressure from building up inside reactor components. If that would happen, it could lead to severe damage. The problem with the current research loop design, is that the flow inside the research loop is controlled by a pump. If this pump breaks down, the uranyl nitrate solution almost comes to a standstill (natural circulation will cause for some small but insufficient flow in the research loop). Since the fluid will still be exposed to the neutron flux from the reactor core, it will start to warm up more and more, soon to reach boiling temperatures.

The second problem arises when the uranyl nitrate solution would be taken out of the research loop in case of a pump breakdown or a different failure: the volume of the research loop. The current research loop has a volume of around 5 litres. If 5 litres of uranyl nitrate solution is taken out of the loop into a so called glove box, it would be a too dangerous source of radiation [20].

1.4 Goals

The goal of this research is to propose a new design for a molybdenum-99 producing loop that is based on the current design, but solves the two practical problems illustrated above. Key in this design is that the temperature inside the loop should always stays below $100\text{ }^{\circ}\text{C}$, while having a uranyl nitrate concentration that is as high as possible to facilitate the molybdenum-99 production.

The solutions to the problems mentioned in section 1.3 are pretty straight forward. First, the flow in the loop has to be driven by natural circulation due to the heat generation caused by the neutron and gamma irradiation. The second requirement is that the volume of the loop has to be significantly smaller than the volume of the current, U-shaped research loop. As a target for this research, a maximum volume of 0.5 L is set, ten times smaller than the current loop [20].

The cooling is also a significant factor in this research. It is important to investigate which uranyl nitrate concentrations can be achieved at different cooling conditions, to see which cooling conditions are needed for the loop to be feasible. This leads to the following three research questions for this thesis:

1. With the current research loop in mind, is it possible to design a smaller loop that is driven by natural circulation, with a maximum volume of 0.5 L?
2. If so, what is the maximum uranyl nitrate concentration in the loop before temperatures rise above 100 °C, and how does this maximum concentration vary for different cooling conditions?
3. Would the results of research questions 1 and 2 make the loop feasible for actual production purposes?

This research aims to answer these questions by designing a new loop and making a simplified mathematical model of this loop to calculate the temperatures inside the loop for different concentrations. As mentioned, the temperatures and concentrations inside the loop will be investigated for different cooling conditions.

1.5 Conceptual design

To give a general understanding of the concept for the new loop, a global sketch is given in figure 1.2. The loop will be constructed using cylindrical tubing, and will be placed vertically inside the reactor. The loop will be completely filled with a liquid uranyl nitrate solution. The idea is that the uranyl nitrate solution is heated up in the bottom part of the loop (the red part in figure 1.2), and cooled down in the top part of the loop (the blue part in figure 1.2).

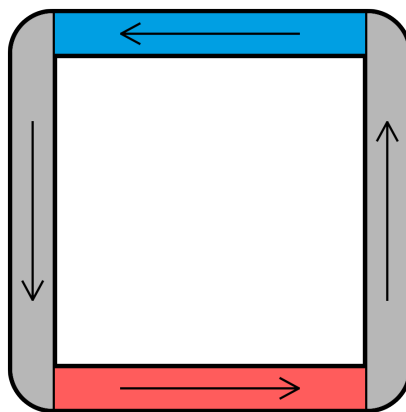


Figure 1.2: Conceptual sketch of the new loop. The red area represents the part of the loop that is heated due to neutron and gamma radiation, the blue area represents the part that is cooled, and the arrows indicate the flow inside the loop.

The heat generation will be the result of both fission in the uranyl nitrate solution, caused by the neutron flux coming from the reactor core, and deposition of heat in the construction material of the loop by the gamma flux. The cooling will be done by a concentric tube, countercurrent heat exchanger. The vertical parts and top part of the loop are shielded from all radiation, so that heat generation only occurs in the bottom part. The heating and cooling cause a difference in the density of the uranyl nitrate solution between the left and right part of the loop, and thus a difference in gravitational force, resulting in a natural circulation of the fluid inside the loop.

2 | Theory

2.1 Heat generation

Since this research is focused on the natural circulation of a uranyl nitrate solution and the temperatures at which this occurs, an important subject to understand is the heat generation inside the loop. As was introduced in section 1.5, heat generation will cause temperatures in the bottom part of the loop to rise, which in combination with cooling in the top part, will result in a flow of the fluid. There are two major sources of heat generation: the primary source is the energy released by the nuclear fission of uranium-235, and the secondary source is the heat deposited in the walls of the loop by the gamma radiation flux approaching from the HOR core.

2.1.1 Nuclear fission

The energy released from a single fission event E_f of a uranium-235 core by a neutron is on average 192 MeV [11]. Therefore, the total amount of energy generated in a reactor (or in this case the loop) can simply be calculated by multiplying with the total amount of fission events. To do so, the *reaction rate* (fission events per second per cubic meter) is needed [11]:

$$R''' = \sigma_{f,5} \epsilon_5 N \phi_n, \quad (2.1)$$

where $\sigma_{f,5}$ is the neutron cross-section of uranium-235 fission, ϵ_5 is the enrichment, N is the amount of uranium nuclei per volume and ϕ_n is the neutron flux. The values of the first three parameters in this equation are known or can easily be calculated. The cross section of uranium-235 fission for thermal neutrons is 583 barn [22], where a barn is equal to 10^{-28} m². The enrichment is 19.75%, which is low enriched uranium (LEU) [12]. Since uranyl nitrate [UO₂(NO₃)₂] only has one uranium nucleus, the amount of uranium nuclei per cubic meter can be calculated by dividing the concentration c of the uranyl nitrate solution by the molar mass M of uranyl nitrate, which is 394.04 g/mol, and multiplying by the constant of Avogadro N_A :

$$N = \frac{c \cdot N_A}{M} \quad (2.2)$$

The reaction rate, and thus the heat generation due to nuclear fission, is therefore directly dependent on the uranyl nitrate concentration c . The last parameter in equation 2.1, the neutron flux, is less straight forward. Conveniently, the value of the neutron flux around the HOR has previously been calculated. For the setup of the current research loop, the flux values are shown in figure 2.1. Heat production predominantly occurs due to fission by thermal neutrons, which have an energy

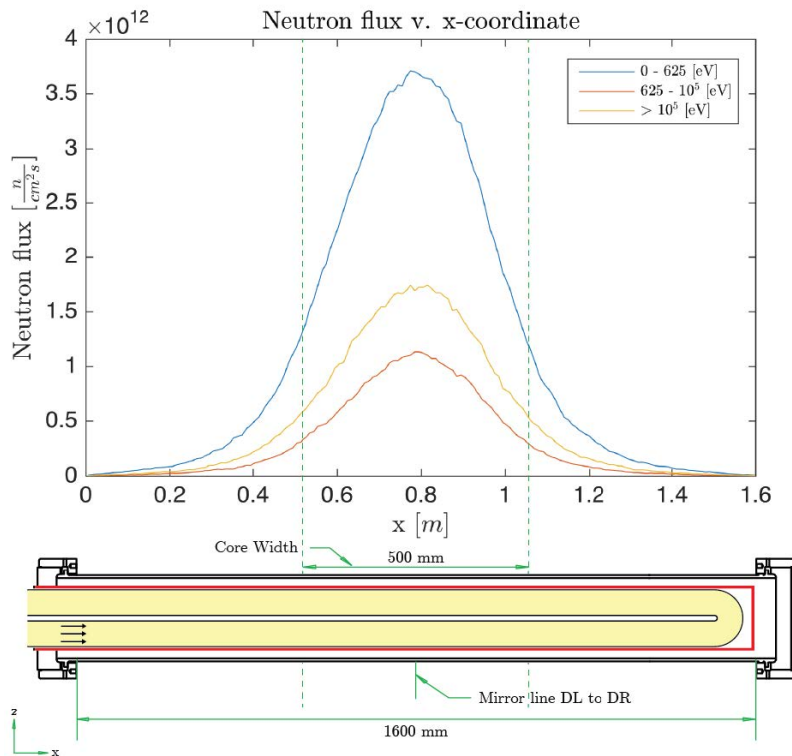


Figure 2.1: Neutron flux density inside the current research loop along the x-direction and a schematic cross section of the setup [16].

between 5 and 100 meV [27]. Therefore, the neutron energy range of 0 - 625 eV (represented by the blue curve in figure 2.1) is of interest to determine the neutron flux in the setup of the new loop. Since a high reaction rate is desired in the production of molybdenum-99 [12], it follows from equation 2.1 that the neutron flux should be as high as possible. Also, the neutron flux preferably has an approximately constant value over the entire irradiated part of the loop. As can be seen in figure 2.1, both requirements are approximately met when the loop is placed directly in front of the reactor core with a maximum length of around 20 cm. In that case the neutron flux can be assumed to be $3.5 \cdot 10^{12} \text{ cm}^{-2}\text{s}^{-1}$.

The total heat production per second in the irradiated part of the loop can now be calculated as follows:

$$Q = VE_f R''' , \quad (2.3)$$

where V is the total volume of the irradiated part of the loop.

2.1.2 Gamma heating

Another important source of heat in the loop is the interaction of gamma radiation with the construction material of the loop (zirconium, see chapter 3). The absorption of gamma radiation results in heat deposition into the construction material, which will increase its temperature. As a result, there will be a heat flux from the wall to the uranyl nitrate solution. The HOR Development section uses a rule of thumb of approximately 0.3 W/g for gamma heating of all construction materials close to the reactor core [10].

An estimation of the total amount of energy deposited by gamma radiation in the irradiated part of the loop can thus be made by multiplying that value with the total mass of the construction material in this section:

$$P_\gamma = u \cdot \rho \cdot V_{\text{tube}}, \quad (2.4)$$

where u is the energy density used by the HOR department as mentioned above, ρ is the density of the construction material and V_{tube} is the volume of the construction material in the irradiated part of the loop. For a straight, cylindrical tube, this volume can be calculated as follows:

$$V_{\text{tube}} = \pi L((r + d)^2 - r^2) = \pi L(d^2 + 2dr), \quad (2.5)$$

where L is the length of the tube, r is its radius and d is the thickness of the tube wall.

The heat production inside the construction material results in a flux of heat from the wall of the tube into the uranyl nitrate solution. For this research, a worst case scenario is assumed where all the energy is transferred to the uranyl nitrate solution and not to the surroundings of the loop. The energy flux is then:

$$\phi''_{q,\gamma} = h(T_w - T_{\text{bulk}}) = \frac{P_\gamma}{A}, \quad (2.6)$$

where h is the *heat transfer coefficient* between the tube and the uranyl nitrate solution, and A is the contact area between the tube and the uranyl nitrate solution. When the energy flux is multiplied with A , it results in *Newton's law of cooling*, see equation 2.25.

2.2 Transport phenomena

To answer the research questions, it is necessary to know what the heat production terms mentioned above attribute to the temperatures inside the system and how they affect the flow of the uranyl nitrate solution. The field of transport phenomena describes the transport of momentum, energy and mass in the form of mathematical relations. It uses laws of conservation combined with laws that describe the fluxes of those conserved quantities. Specifically, it uses the *principle of balancing* to describe systems. This principle states that for a certain control volume (on either *macro* or *microscopic* level), the change in a certain quantity can be calculated as the sum of the inflow, the outflow and the production and/or loss of that quantity [1]:

$$\frac{d}{dt} = \phi_{\text{in}} - \phi_{\text{out}} + \text{production/loss} \quad (2.7)$$

In this research, and often in transport phenomena in general, the situations and systems observed are in *steady state*, meaning that the change in time $\frac{d}{dt}$ in equation 2.7 is zero. In such cases, the outgoing flow thus balances the incoming flow and production/loss of the quantity. To describe the temperatures and flow inside the loop that this research aims to design, the balances of two specific quantities are needed: the internal energy balance and impulse balance.

2.2.1 Dimensionless numbers

Before elaborating on the calculations of these balance equations, a few *dimensionless numbers* have to be introduced. Dimensionless numbers are widely used in the field of transport phenomena to describe processes or situations that include multiple types of transport (for example convective and molecular transport). They are based on ratios and they are *dimensionless*, meaning that they do not have any assigned physical dimension. The ones used in this research are the following:

- The Reynolds number is defined as the ratio between inertia forces and viscous forces within a fluid, and is used to describe the state of a flow (laminar or turbulent). The critical value above which a flow becomes turbulent differs for different geometries: for a cylindrical tube it is in the range of 2000 - 2500 [1].

$$\text{Re} = \frac{\text{inertia forces}}{\text{viscous forces}} = \frac{\rho \langle v \rangle D_h}{\mu}, \quad (2.8)$$

where ρ is the density, $\langle v \rangle$ is the average velocity (also known as the bulk velocity), D_h is the hydraulic diameter and μ is the dynamic viscosity. In this research, where a cylindrical, completely filled tube is used, the hydraulic diameter D_h is equal to the regular diameter D of the tube.

- The Prandtl number is the ratio between momentum diffusivity and thermal diffusivity [9]. It is used to describe heat transfer to flowing media, as it typically determines the *thermal boundary layer thickness* and indicates the dominance of either convective or conductive heat transfer [6].

$$\text{Pr} = \frac{\text{momentum diffusivity}}{\text{thermal diffusivity}} = \frac{\nu}{a} = \frac{c_p \cdot \mu}{\lambda}, \quad (2.9)$$

where ν is the kinematic viscosity, a is the thermal diffusivity, c_p is the specific heat capacity and λ is thermal conductivity. Note that the Prandtl number is a fluid property, in contrast to the Reynolds number, and thus it is independent of flow geometry.

- The Graetz number is the ratio between conductive heat transfer and convective heat transfer, and is also used to describe heat transfer to flowing media [17].

$$\text{Gz} = \frac{\text{conductive heat transfer}}{\text{convective heat transfer}} = \frac{a \cdot L}{D^2 \langle v \rangle}, \quad (2.10)$$

where L is the length of the tube.

- The Nusselt number is the ratio between the total heat transfer and the conductive heat transfer, and is used to calculate the heat transfer coefficient h [1]. The Nusselt number can be expressed as a function of the dimensionless numbers above, as can be seen in equation 2.26.

$$\text{Nu} = \frac{\text{total heat transfer}}{\text{conductive heat transfer}} = \frac{hD}{\lambda} \quad (2.11)$$

2.2.2 Internal energy balance

Now that the necessary dimensionless numbers are introduced, further transport phenomena can be explained. As mentioned, most important are the internal energy balance and momentum balance. The internal energy balance over a control volume is as follows [1]:

$$\frac{dU}{dt} = \frac{d}{dt}(\rho V u) = \phi_{m,\text{in}} \cdot u_{\text{in}} - \phi_{m,\text{out}} \cdot u_{\text{out}} + \phi_q + P_u, \quad (2.12)$$

where ϕ_m is the flow of mass, ϕ_q is the net flow of heat and P_u is the total production of internal energy. Note that capitol U denotes the total internal energy in the control volume, whereas small u denotes the *specific internal energy*, given in J/kg. For constant volume, $u = c_V dT$, where c_V is the *specific heat capacity* at constant volume. Because c_V is constant to the first order for small temperature differences, u can be expressed as $c_V T$. For fluids, because of their small *thermal expansion coefficient*, it is a valid approximation that $c_V = c_p$, and thus $u = c_p T$ [1]. Here, c_p is the specific heat capacity at constant pressure.

In steady state, where $\frac{dU}{dt} = 0$ and $\phi_{m,\text{in}} = \phi_{m,\text{out}} = \phi_m$, and at constant pressure and density, equation 2.12 becomes:

$$0 = \phi_m c_p (T_{\text{in}} - T_{\text{out}}) + \phi_q + Q + \phi_m e_{\text{fr}}, \quad (2.13)$$

where T_{in} and T_{out} are the temperatures of the fluid at the in- and outlet of the control volume, Q is the heat production in the control volume, and $\phi_m e_{\text{fr}}$ is a dissipation term caused by the friction.

2.2.3 Momentum balance

The other important balance is the momentum balance. Momentum is a vector, thus there are three momentum balances: one for each of the x, y and z components. All three look like this [1]:

$$\frac{d}{dt}(M v_i) = \phi_{m,\text{in}} \cdot v_{i,\text{in}} - \phi_{m,\text{out}} \cdot v_{i,\text{out}} + \sum F_i, \quad (2.14)$$

where M is the total mass in the control volume, v_i is the velocity in the i-direction and $\sum F_i$ is the sum of all forces in the i-direction. Again, in steady state and for equal in- and outflow of momentum, equation 2.14 reduces to a *balance of forces*:

$$0 = \sum F_i \quad (2.15)$$

In- and outflow of momentum are equal in this research, since the control volume for momentum is the entire loop. In that case the in- and outlet are in one and the same location, and thus the inflow of the control volume is the same as the outflow.

2.2.4 Equation of states

The momentum balance, or the force balance, will determine the velocity of the flow in the loop. As shall be further explained in chapter 3, one of the key terms in this force balance will be the gravity working on the fluid in the loop. As was introduced in section 1.5, the idea behind this system is namely that the fluid in the loop is warmer and thus less dense on one side of the loop than the other. That means that the gravitational force on the fluid is larger on one side of the loop than the other, causing the fluid to flow. In a system where the temperature change impacts the force balance and eventually impacts the flow, the Boussinesq approximation can be used to simulate this phenomenon [23]. The main criteria for the Boussinesq approximation to be valid is [28]:

$$\beta\Delta T \ll 1, \quad (2.16)$$

where β is the thermal expansion coefficient of the uranyl nitrate solution, and ΔT is the maximum temperature difference in the system. In this research, this maximum difference is approximately 90 °C and $\beta = 0.000523 \text{ K}^{-1}$ [23], so therefore $\beta\Delta T \approx 0.047 \ll 1$, and thus the Boussinesq approximation is valid.

To describe how the density of the uranyl nitrate solution is dependent on temperature, the Boussinesq approximation uses the following *equation of states* [28]:

$$\rho(T) = \rho_0 - \rho_0\beta(T - T_0), \quad (2.17)$$

where 0 denotes an arbitrary reference value for the temperature and density at that temperature. In this research, the reference temperature is set at 333.15 K, or 60 °C.

2.2.5 Friction

Another term that will play an important role in the force balance, and also in the internal energy balance, is friction. In the force balance, it is present as a frictional force that works against the direction of the flow. This force can be calculated as follows [1]:

$$F_{\text{fr}} = \tau_{w \rightarrow f} \cdot SL \quad (2.18)$$

In equation 2.18, S is the wetted perimeter, the part of the circumference of the tube that is wetted by the fluid. In this research, where the tube is circular and completely filled, this is equal to πD , where D is the diameter of the tube. L is the total length of the tube. $\tau_{w \rightarrow f}$ is the *shear stress* that the wall of the tube exerts on the flowing fluid, which can be calculated as follows [1]:

$$\tau_{w \rightarrow f} = -f \cdot \frac{1}{2} \rho \langle v \rangle^2, \quad (2.19)$$

where ρ is the density of the fluid, $\langle v \rangle$ is the average flow velocity, and f is the Darcy-Weisbach friction factor. Note that, as mentioned before, the minus sign in front of f in equation 2.19 indicates that the frictional force works against the direction of the flow. There are more than a dozen equations that can be used to calculate the Darcy-Weisbach friction factor, for all different types of flow regimes. Most of them are only applicable for either laminar flow or turbulent flow. In laminar flow,

the Hagen-Poiseuille equation is valid [7]. In turbulent flow, the Haaland equation (which is an approximation of the implicit Colebrook-White equation [8]) is one of the mostly used equations for explicit calculations [14].

However, it is uncertain beforehand in which regime the flow in the new loop will be. Therefore, one of the few equations for the Darcy-Weisbach friction factor that cover all flow regimes is preferred, even though they are significantly less simple than the Haaland equation. In this research, a recent equation by Bellos et al. (2018) is used [4]:

$$f = \left(\frac{64}{\text{Re}}\right)^a \left[0.75 \ln\left(\frac{\text{Re}}{5.37}\right)\right]^{2(a-1)b} \left[0.88 \ln\left(6.82 \frac{D}{\epsilon}\right)\right]^{2(a-1)(1-b)}, \quad (2.20)$$

where

$$a = \frac{1}{1 + \left(\frac{\text{Re}}{2712}\right)^{8.4}} \quad (2.21)$$

and

$$b = \frac{1}{1 + \left(\frac{\text{Re}}{150 \frac{D}{\epsilon}}\right)^{1.8}} \quad (2.22)$$

In equation 2.20, ϵ/D is the *relative roughness* of the pipe, and ϵ is the *effective roughness*.

Apart from in the force balance, friction also plays an important role in the internal energy balance. Friction between the wall of the tube and the fluid will cause the system to warm up. Mechanical energy from the fluid is *dissipated* into internal energy. The energy per mass unit that is dissipated due to friction in a circular, completely filled tube with length L and diameter D , is as follows [1]:

$$e_{\text{fr}} = f \cdot \frac{L}{D} \cdot \frac{1}{2} \langle v \rangle^2 \quad (2.23)$$

2.2.6 Flow of mass

Both the internal energy balance and the momentum balance include the flow of mass ϕ_m , that is expressed in kg/s . However, in most equations for the dimensionless numbers and friction, the bulk velocity $\langle v \rangle$ is used. The following equation can be used in a cylindrical tube to calculate ϕ_m when $\langle v \rangle$ is known, or vice versa [1]:

$$\phi_m = \rho \pi r^2 \langle v \rangle, \quad (2.24)$$

where ρ is the density of the fluid and r is the radius of the tube.

2.2.7 Flow of heat

As was already mentioned in section 2.1.2, the flow of heat through an interface is described by *Newton's law of cooling (and heating)*:

$$\phi_q = hA\Delta T, \quad (2.25)$$

where A is the surface area through which the heat flows, ΔT is the temperature difference between the two sides of the interface, and h is the *heat transfer coefficient*.

The heat transfer coefficient can be calculated using the Nusselt number, as follows from equation 2.11 [17]:

$$h = \frac{\langle \text{Nu} \rangle \lambda}{D} = \begin{cases} \frac{0.027 \cdot \text{Re}^{0.8} \cdot \text{Pr}^{0.33} \cdot \lambda}{D}, & (\text{for } \text{Re} > 10^4, \text{ Pr} \geq 0.7) \\ \frac{1.62 \cdot \text{Gz}^{-\frac{1}{3}} \cdot \lambda}{D}, & (\text{for } \text{Gz} < 0.05) \\ \frac{3.66 \cdot \lambda}{D}, & (\text{for } \text{Gz} > 0.1) \end{cases} \quad (2.26)$$

In this equation, $\langle \text{Nu} \rangle$ is the average Nusselt number in the fluid.

Equation 2.25 could be used in describing the flow of heat from the wall of the tube to the fluid, caused by gamma heating. However, as is further explained in chapter 3, the cooling of the fluid in the loop is done using a countercurrent heat exchanger. For such a heat exchanger, the equation for the flow of heat is slightly different. In essence, it looks the same as Newton's law of cooling [26]:

$$\phi_q = U \pi D l \Delta T_{LM} \quad (2.27)$$

In this equation, $\pi D l$ still resembles the area A through which the heat flows, but the other two factors are different. First, ΔT_{LM} is the *logarithmic mean temperature difference*, defined as:

$$\frac{(T_{\text{in,loop}} - T_{\text{out,C}}) - (T_{\text{out,loop}} - T_{\text{in,C}})}{\ln \left(\frac{T_{\text{in,loop}} - T_{\text{out,C}}}{T_{\text{out,loop}} - T_{\text{in,C}}} \right)}, \quad (2.28)$$

where C denotes the cooler. Second, the h in equation 2.25 is replaced by the *total heat transfer coefficient* U [1]:

$$\frac{1}{U} = \frac{1}{h_{\text{loop}}} + \frac{d}{\lambda_w} + \frac{1}{h_C}, \quad (2.29)$$

where d is the thickness of the wall between the loop and the cooler, λ_w is the thermal conductivity of the wall material, and h_{loop} and h_C are the heat transfer coefficients between the wall and both of the fluids involved. Note that in a steady state situation, the flow of heat in the cooler should be equal to the total production of heat in the irradiated part of the loop $\phi_q = Q + P_\gamma$.

3 | Design

As mentioned before, the goal of this research is to design a loop that naturally circulates a uranyl nitrate solution in order to produce molybdenum-99. In this chapter, the design and properties of this loop will be discussed. Both the loop itself, e.g. the construction material and the dimensions, as well as the fluids that are being used are specified. Also, the theories discussed in chapter 2 are further elaborated for the specific situation of this loop.

3.1 The loop

3.1.1 Construction material

For the construction material of the new loop, the same material is chosen that is used in the current research loop: zircaloy (zirconium alloy). This material is commonly used for nuclear purposes, because of its excellent corrosion resistance, good mechanical properties, and very low thermal neutron cross section [2]. The high corrosion resistance is needed due to the high acidity of the uranyl nitrate solution [12]. The thermal neutron cross section of only 0.18 barn is essential to ensure that the material practically doesn't influence the neutron flux [16].

Zircaloy has a density of $6.55 \cdot 10^3 \text{ kg m}^{-3}$ [2]. Since the wall of the current research loop is 2 mm thick [16], this thickness is also used for the design of the new loop. The effective roughness of a zircaloy tube has not been found, therefore the general value for the effective roughness of drawn tubes (e.g. aluminum) is used: $\epsilon = 1.5 \cdot 10^{-6} \text{ m}$ [19].

3.1.2 Loop design

The new loop will be square shaped and constructed using cylindrical tubing. A 2D sketch of the parallel cross section can be found in figure 3.1. As was mentioned in section 2.1.1, the sides of this loop can be no longer than 20 centimeters, in order to have a high and constant neutron flux. Therefore the length l of the sides of the loop is set at 20 centimeters. For this research, there are three important sections in the loop: section A, the part of the loop that is irradiated by the neutron flux, section B, the part of the loop that is being cooled, and section C, the cooler around section B of the loop. Note that the horizontal sides of the loop are tilted at a 2° angle to ensure that the fluid inside the loop always flows in the same direction, and that both the vertical parts of the loop (shaded in figure 3.1) and the cooling part are assumed to be completely shielded from neutrons and gamma radiation.

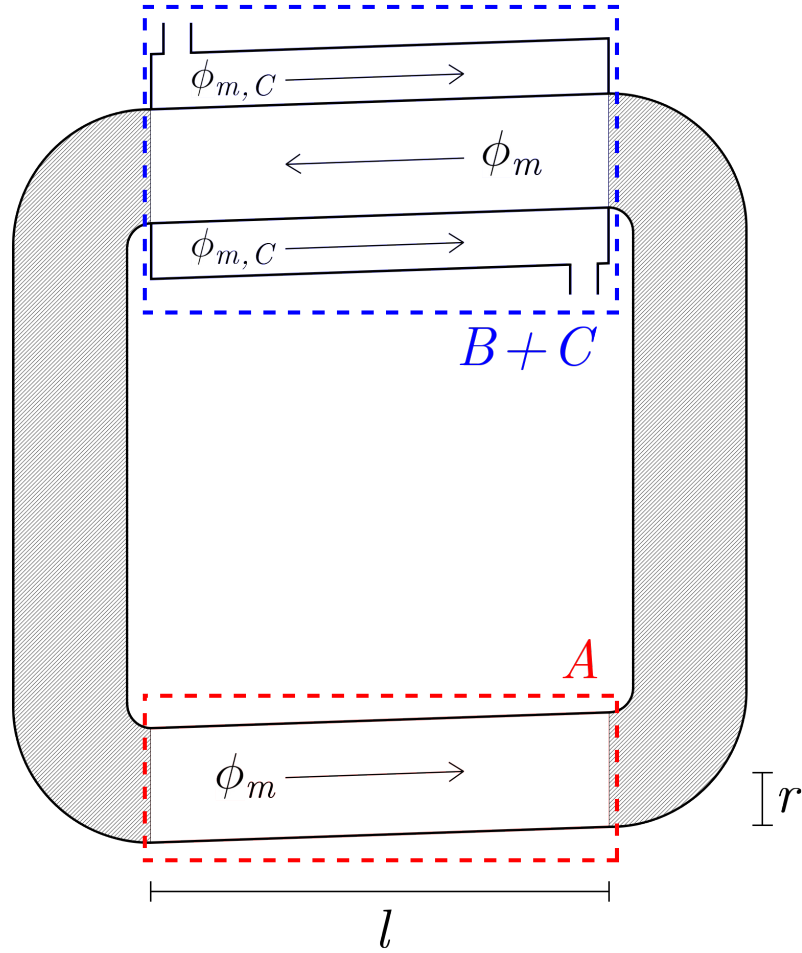


Figure 3.1: A sketched, 2D cross section design of the loop. Indicated are the mass flows ϕ_m and $\phi_{m,C}$, the length of the sides l and the radius of the tube r . Also, the important sections in the loop are marked.

3.1.3 Radius of the tube

A large diameter of the tube is beneficial for the flow in the loop, since the friction terms gets smaller as the diameter is larger (see equations 2.20 and 2.23). On the other hand, a large diameter also means that cooling will be less efficient, since the relative contact area of the fluid with the wall is smaller. This can be seen in equation 2.26, where the heat transfer coefficient gets smaller for a larger diameter. The optimal radius r of the tube will thus have to be calculated.

As stated in the introduction of this research, the total volume of the loop can be no more than 0.5 liter. To make an estimation of the volume, the volumes of the four sides are added, plus the volume of a so called *horn torus*, of which the distance from the center of the torus to the center of the tube is equal to the radius of the tube, to account for the corners of the loop [31]. This results in the following equation:

$$V = 4l\pi r^2 + 2\pi^2 r^3 = (4l + 2\pi r)\pi r^2 \leq 0.0005 \text{ m}^3 \quad (3.1)$$

As mentioned in section 3.1.2, l is set at 20 cm to ensure a high and approximately constant neutron flux. Therefore the maximum value for r can be calculated using the following equation:

$$0.0005 = 4 \cdot 0.20 \cdot \pi r^2 + 2\pi^2 r^3 \quad (3.2)$$

This equation has three possible solutions of which only one is positive, $r \approx 0.0134157$ m. Thus, $r_{\max} = 13.4$ mm is used as the maximum radius in this research.

3.1.4 The cooler

As mentioned in section 2.2.7, section B of the loop is cooled using a countercurrent, concentric tube heat exchanger. This means the coolant flows around the tube in the opposite direction of the uranyl nitrate solution inside the loop. A countercurrent heat exchanger is used because countercurrent flow is more efficient in cooling than co-current flow, since the temperature gradient remains higher throughout the cooler [25]. This results in a larger logarithmic mean temperature difference ΔT_{LM} in equation 2.27.

3.1.5 Safety

As mentioned in chapter 1, the main restriction in this research is that the temperatures inside the loop have to stay below 100 °C. To keep an ample safety margin in the loop design, the maximum temperature in the loop is set at 90 °C. Note that the maximum temperature in the loop will occur at the walls of the loop, due to the gamma heating. Therefore, the average wall temperature of the irradiated part of the loop will be used as the maximum temperature in the loop, which can be calculated using equation 2.6.

3.2 Fluids

3.2.1 Coolant

Regular water is used as the coolant in the countercurrent heat exchanger. In table 3.1, the properties of water at 10°C are given. The dynamic viscosity μ of water is very dependent of temperature, which has to be taken into account if the initial temperature of the cooling water is varied. The following formula can be used to calculate μ for temperatures in the range of 10-70°C [18], but when matched with the data from the Data Companion [17], it shows to be very precise in the entire range of 0-100°C as well:

$$\log \frac{\mu_T}{\mu_{20}} = \frac{A(20 - T) - B(T - 20)^2}{T + C}, \quad (3.3)$$

where μ_T is the viscosity at a certain temperature, μ_{20} is the viscosity at 20°C (1.0020 mPa s), $A = 1.1709$, $B = 0.001827$, and $C = 89.93$.

Table 3.1: Properties of water at 10°C and 10⁵ Pa. [17]

Density ρ_w	999.63 kg m ⁻³
Specific heat capacity $c_{p,C}$	4185 J kg ⁻¹ K ⁻¹
Thermal conductivity λ_w	0.607 W m ⁻¹ K ⁻¹
Viscosity μ	0.001307 kg m ⁻¹ s ⁻¹
Thermal expansion coefficient β	0.000088 K ⁻¹
Thermal diffusivity a	0.138 · 10 ⁻⁶ m ² s ⁻¹

3.2.2 Uranyl nitrate

The properties of the uranyl nitrate solution can be found in table 3.2. The uranium-235 enrichment ϵ_5 that is used in equation 2.1 is 19.75%, low enriched uranium (LEU) [23]. Since the solvent for the uranyl nitrate is water, the viscosity of the solution is dependent of temperature in the same way water is. Moreover, the viscosity is also dependent of the concentration of the uranyl nitrate, which can be calculated using a Jones-Talley type equation [13]:

$$\frac{\mu_c}{\mu_0} = 1 + A \cdot \sqrt{c} + B \cdot c, \quad (3.4)$$

where μ_c denotes the viscosity at a concentration c , μ_0 is the viscosity of the solvent, and A and B are constants. The mean values of the slightly temperature dependent constants A and B that Grant et al. [13] found are used in this research: $A = -0.1687$ and $B = 0.7904$.

Table 3.2: Properties of an aqueous uranyl nitrate solution with concentration 310 g L⁻¹ [23]. *: Property of water at 60 °C and 10⁵ Pa used [17].

Density ρ_0	1330.6 kg m ⁻³
Specific heat capacity c_p	2905.5 J kg ⁻¹ K ⁻¹
Thermal conductivity λ^*	0.665 W m ⁻¹ K ⁻¹
Thermal expansion coefficient β^*	0.000523 K ⁻¹
Thermal diffusivity a	$\frac{\lambda}{\rho c_p}$

3.3 Balances

The balances given in equations 2.12 and 2.14 have to be further elaborated for the specific situation of this loop. Note that the loop is assumed to be in steady state, meaning that all $\frac{d}{dt}$ terms vanish and equations 2.13 and 2.15 can be used.

3.3.1 Internal energy balance section A

Following the order of chapter 2, the internal energy balances are discussed first. In figure 3.2, a schematic presentation of the control volumes and heat flows in the system is given to support this discussion. Starting from equation 2.13 for the internal energy balance of section A, it follows that the net flow of heat coming from the walls of the tube ϕ_q , the heat production Q and the frictional dissipation term

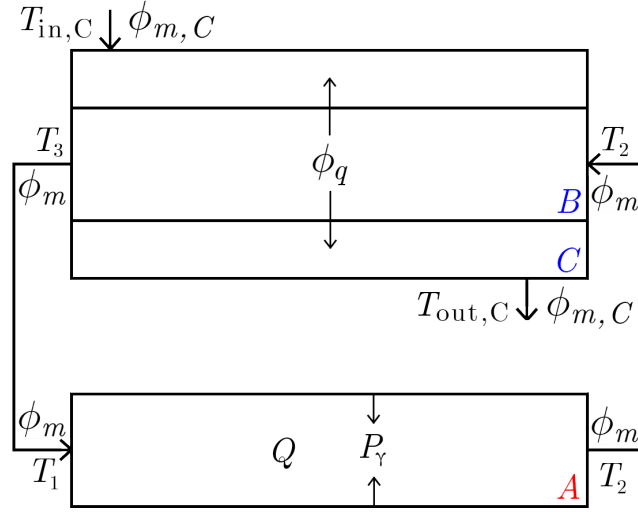


Figure 3.2: Schematic representation of the control volumes and heat flows in the system. Note that the frictional dissipation terms are not displayed in this figure.

$\phi_m e_{\text{fr}}$ are needed to complete the balance equation. Since a worst case scenario is assumed, where the loop is not cooled by energy loss to the surrounding, ϕ_q is given by $\phi_{q,\gamma}'' \cdot A = P_\gamma$. Q is given in equation 2.3. The friction term can be calculated using equation 2.23. The temperatures T_{in} and T_{out} will be labeled T_1 and T_2 respectively (see figure 3.2). Putting all this together leads to the following equation:

$$0 = \phi_m c_p \cdot (T_1 - T_2) + Q + P_\gamma + \phi_m e_{\text{fr}} \quad (3.5)$$

3.3.2 Internal energy balance section B

For section B, the same procedure applies as for section A. Only now, since it is assumed that every part of the loop but section A is shielded from neutrons and gamma rays, Q is zero and ϕ_q is the flow of heat *extracted* from the uranyl nitrate by the cooler. The magnitude of this flow is given by equation 2.27 and it has a negative sign in front, as heat is leaving the control volume of section B. For computational purposes (further explained in chapter 4), T_{in} is assumed to be T_2 , neglecting the small temperature gradient due to friction in the vertical parts of the loop, and T_{out} is labeled T_3 . The friction term is not neglected in section B itself, giving the following balance equation:

$$0 = \phi_m c_p \cdot (T_2 - T_3) - \phi_q + \phi_m e_{\text{fr}} \quad (3.6)$$

3.3.3 Internal energy balance section C

Just as for sections A and B, the internal energy balance for section C is formulated. Again, Q is zero and ϕ_q is the flow of heat in the cooler. However, ϕ_q is now accompanied by a plus sign, since the heat is entering section C, and the friction term in section C is neglected:

$$0 = \phi_{m,C} \cdot c_{p,C} \cdot (T_{\text{in},C} - T_{\text{out},C}) + \phi_q \quad (3.7)$$

Note that the index C denotes the cooler or the cooling water.

3.3.4 Momentum balance loop

As was explained in section 2.2.3, the momentum balance reduces to a force balance when the system is in steady state, with equal in- and outflow of momentum. Also, to simplify the calculations, this force balance is composed in only one direction: the direction of the flow in the loop. There are two forces working on the fluid in the loop, the frictional force and the gravity. The frictional force is given by equation 2.18, where L is approximated by $4l$ (the total length of the four sides of the loop), and it works against the direction of the flow. The gravitational force is given by mg , where m is the mass and g is the gravitational acceleration (9.81 m/s² in the Netherlands). It works in the direction of the flow in the two vertical sides of the loop: in the opposite direction in the warm side of the loop (the right side in figure 3.1), and in the same direction in the cold side. Note that the gravitational force due to the small incline in the horizontal sides is neglected. All these forces considered, the balance equation is as follows:

$$0 = \pi r^2 l \rho_B \cdot g - \pi r^2 l \rho_A \cdot g - F_{\text{fr}}, \quad (3.8)$$

which can be further specified to be:

$$0 = g \pi r^2 l (\rho_B - \rho_A) - 2\pi D l f \rho_0 \cdot \langle v \rangle^2 \quad (3.9)$$

In equation 3.9, ρ_A is the density after section A, based on T_2 , and ρ_B is the density after section B, based on T_3 . These densities are calculated using equation 2.17.

As will be further explained in chapter 4, the momentum balance is used to calculate the average velocity in the loop $\langle v \rangle$, which is needed in other calculations, e.g. for the mass flow ϕ_m in the loop. From equation 3.9, the following equation for $\langle v \rangle$ follows:

$$\langle v \rangle = \sqrt{\frac{g r^2 (\rho_B - \rho_A)}{4 D f \rho_0}} \quad (3.10)$$

3.4 Production of molybdenum-99

The last aspect that needs to be clarified is the production of molybdenum-99. The reaction rate, the number of fission events per second per cubic meter, is given in equation 2.1. However, not every fission event results in a molybdenum-99 nucleus. Pothoven showed how the *fission yield* γ , the fraction of a fission product produced per fission event, of molybdenum-99 is 0.0613 [24]. This means the *production rate*, the number of molybdenum-99 molecules produced per second per cubic meter, can be expressed as:

$$P'''_{\text{Mo}} = \gamma_{\text{Mo}} \cdot R''' \quad (3.11)$$

When multiplied by the volume of the irradiated part of the loop, this results in the actual molybdenum-99 production of the loop. Since the volume is kept small in this research, the focus is to maximize the production rate by maximizing the reaction rate. The reaction rate is increased by increasing the concentration. Elgin showed that for the production of molybdenum-99, the desired concentration of uranium in the uranyl nitrate solution is as high as possible [12]. In the current research loop, this is limited to 310 g/L by the capabilities of the molybdenum-99 extraction

facilities. However, with the current geometry and circumstances, this concentration could not be achieved without having hot spots of more than 100 °C due to heat production [16]. Therefore a concentration of only 27.6 g/L could be used until now. The goal in this research is to outperform the current research loop and thus to reach a uranyl nitrate concentration in the new loop that is higher than 30 g/L.

4 | Numerical method

4.1 Iteration

As mentioned in chapters 2 and 3, the temperatures in the loop will be calculated by evaluating balance equations 3.5, 3.6, 3.7 and 3.9 simultaneously. However, it is not possible to calculate the temperatures directly from these equations, since they all have multiple unknown variables. Hence the method of *iteration* is used to find the steady state values of the temperatures. Iteration is a repetitive process that approximates the explicit solution of a problem, in which the outcome of each repetition is the input of the next one. It uses an initial guess of the solution to do the first calculation. When an iterative method is *convergent*, the approximation of the solution improves with each iteration, eventually converging to the actual solution of the problem [15]. When the outcome of the n -th iteration is the same as the outcome of the $n - 1$ -th iteration, the iterative method is completed and the solution to the problem has been found.

However, in the computation of an iteration process, it is almost impossible to get the *exact* same result twice. There will practically always be a slight offset and hence the iteration will run forever. Therefore an *error* ϵ is introduced:

$$\epsilon = \frac{F_{n+1} - F_n}{F_n}, \quad (4.1)$$

where F is the outcome of a certain quantity (temperature, velocity, etc.) and n denotes the n -th iteration. When this error is smaller than a predetermined threshold, the iteration is considered done and the outcome is the approximation of the explicit solution. The smaller the threshold is, the more precise the result of the iterative method will be. However, a very small threshold can be very demanding, causing very long computation, and it is often unnecessary to achieve a sufficient approximation. The optimal value can easily be checked by running the iteration for different error thresholds. Note that in the further calculations in this research, ϵ is used to denote the threshold.

4.2 Parameters and unknown variables

In the loop, and thus in the iteration process, there are three parameters that can be controlled. These are the temperature of the cooling water $T_{in,C}$, the mass flow inside the cooler $\phi_{m,C}$, and the uranyl nitrate concentration c . The first two determine the cooling power of the countercurrent heat exchanger. The larger the mass flow, or the lower the cooling temperature, the the more effective the cooler is.

The third parameter determines how much heat is generated due to fission, and at the same time how much molybdenum-99 is produced. Since the goal of this research is to get c as high as possible, it is likely that the heat production will be large and thus that a strong cooler is needed. Therefore, $T_{\text{in,C}} = 10 \text{ }^\circ\text{C}$ and $\phi_{m,C} = 0.70 \text{ kg/s}$ are used as the standard cooling conditions for most calculations. As mentioned in section 3.1.3, r needs to be varied as well to determine the optimal radius, so in some of the calculations, r is also a parameter.

The unknown variables that are approximated using the iteration are the temperature T_2 at the end of the irradiated part (section A), the average wall temperature T_w of section A, the temperature T_3 after the cooler (section B), and the average flow velocity $\langle v \rangle$. These four values, in combination with the concentration and the cooling conditions, determine whether or not this loop is feasible: i.e. the maximum temperature T_{max} determines if the loop is safe to operate and the concentration and required cooling conditions at that temperature determine if the production of molybdenum-99 is sufficient. Note again that the maximum temperature T_{max} in the loop is the wall temperature T_w in section A, which is calculated using equation 2.6, where the average value of T_1 and T_2 is used as T_{bulk} .

To be exact, $T_{\text{out,C}}$ is also an unknown variable that is approximated. Its value is however irrelevant to answer the research questions, since it is not important for the production of molybdenum-99 at what temperature the cooling water leaves the cooler, nor does it change the safety threshold for the operating conditions (since it is assumed that there is no heat generation inside the cooler, $T_{\text{out,C}}$ can never be larger than T_2).

The results of the iteration process should be plotted for different values of the parameters to make conclusions regarding the research questions. Plots of T_{max} vs. c and $\langle v \rangle$ vs. c are needed for different values of $T_{\text{in,C}}$ and $\phi_{m,C}$. Also, to study the feasibility of the loop, a plot is needed of c_{max} vs. the cooling conditions $T_{\text{in,C}}$ and $\phi_{m,C}$, where the maximum concentration c_{max} is the concentration for which the maximum temperature in the loop reaches the threshold of $90 \text{ }^\circ\text{C}$. Since the current research loop works with a uranyl nitrate concentration of approximately 30 g/L and the optimal concentration for the current research loop is 310 g/L , the uranyl nitrate concentration c is varied between 30 and 310 g/L . The cooling water temperature $T_{\text{in,C}}$ is varied between 1 and $30 \text{ }^\circ\text{C}$, and the mass flow in the cooler $\phi_{m,C}$ between 0.01 and 0.70 kg/s .

4.3 Initial estimations

As mentioned in section 4.1, initial estimations of the solutions have to be made for the first step of the iteration process. First, the temperature at which the uranyl nitrate solution enters the system, T_1 , and the bulk velocity $\langle v \rangle$ have to be estimated. The initial guess for T_1 in this research is $40 \text{ }^\circ\text{C}$, the maximum temperature of the cooling water in the HOR. The initial value for $\langle v \rangle$ is set at 0.02 m/s , or 2 cm/s . Based on this T_1 and $\langle v \rangle$, guesses for T_2 and T_3 are made. Since the system is assumed to be in steady state, T_3 should be equal to T_1 (see figure 3.2), so also $40 \text{ }^\circ\text{C}$. The initial value for T_2 is estimated by multiplying the sum of the increase

in temperature due to nuclear fission and due to gamma heating by the time spent in section A of the loop, and adding that to T_1 :

$$T_2 = T_1 + \frac{Q + P_\gamma}{c_p \rho_0 \pi r^2 l} \cdot \frac{l}{\langle v \rangle} \quad (4.2)$$

The last guess that has to be made is the wall temperature in the irradiated part of the loop, T_w . This temperature is set to 40 °C as well.

4.4 Flowchart

The global structure of one iteration step, giving only the main calculations, is as follows. First the densities of the uranyl nitrate solution in both vertical parts of the loop are calculated using the equation of states (equation 2.17). With these densities, the average flow velocity (and thus the mass flow) in the loop can be calculated using equation 3.10. Next, T_2 is calculated using the internal energy balance in section A (equation 3.5). Also, $T_{\text{out,C}}$ is calculated using the internal energy balance in section C (equation 3.7), assuming the system is in steady state ($\phi_q = Q + P_\gamma$). T_2 and $T_{\text{out,C}}$ are then used to calculate the flow of heat in the cooler ϕ_q using equation 2.27. Finally, T_2 and ϕ_q are put in the balance equation of section B (equation 3.6) to calculate the final temperature T_3 . After these calculations, T_1 is equalled to T_3 and then compared to the previous T_1 using equation 4.1. If the error threshold condition is met, the final solution is found, otherwise the whole process is repeated in a new iteration step. The complete flowchart of the iteration process is given in figure 4.1 and the MATLAB script used to do the main calculations can be found in appendix A.

4.5 Code validation

It is important to validate whether the model used in this research is giving physical results. A few tests can be done to establish this, by putting in values of the parameters for which the results are known. Note that these tests are done for $T_{\text{in,C}} = 10$ °C and $\phi_{m,C} = 0.7$ kg/s. Also note that, as is further discussed in section 5.4, it is often impossible to put very small values into the calculations.

Firstly, the iteration can be run for a situation where there is no (or very little) heat production. This should cause the temperatures in the loop to drop to (around) the temperature of the cooling water. As can be seen in figure 4.2, this is indeed what happens when the gamma heating P_γ is set at 2 J/s and the heat production due to fission Q is varied between 0.50 and 50 J/s. As Q gets closer to zero, the temperature after the cooler T_3 drops to almost 10 °C, the inlet temperature of the cooling water.

The second test that can be done, is turning off the cooling. In that case the temperatures should become very high, or even go to infinity. This can be done in two ways: by setting the total heat transfer coefficient U to (near) zero, or by setting $\phi_{m,C}$ to (near) zero. Again, both cases hold: when U is set to 10^{-5} W/m²K, the temperatures (virtually) go to $2.4075 \cdot 10^6$, and for $\phi_{m,C} = 10^{-5}$ the system crashes into complex numbers.

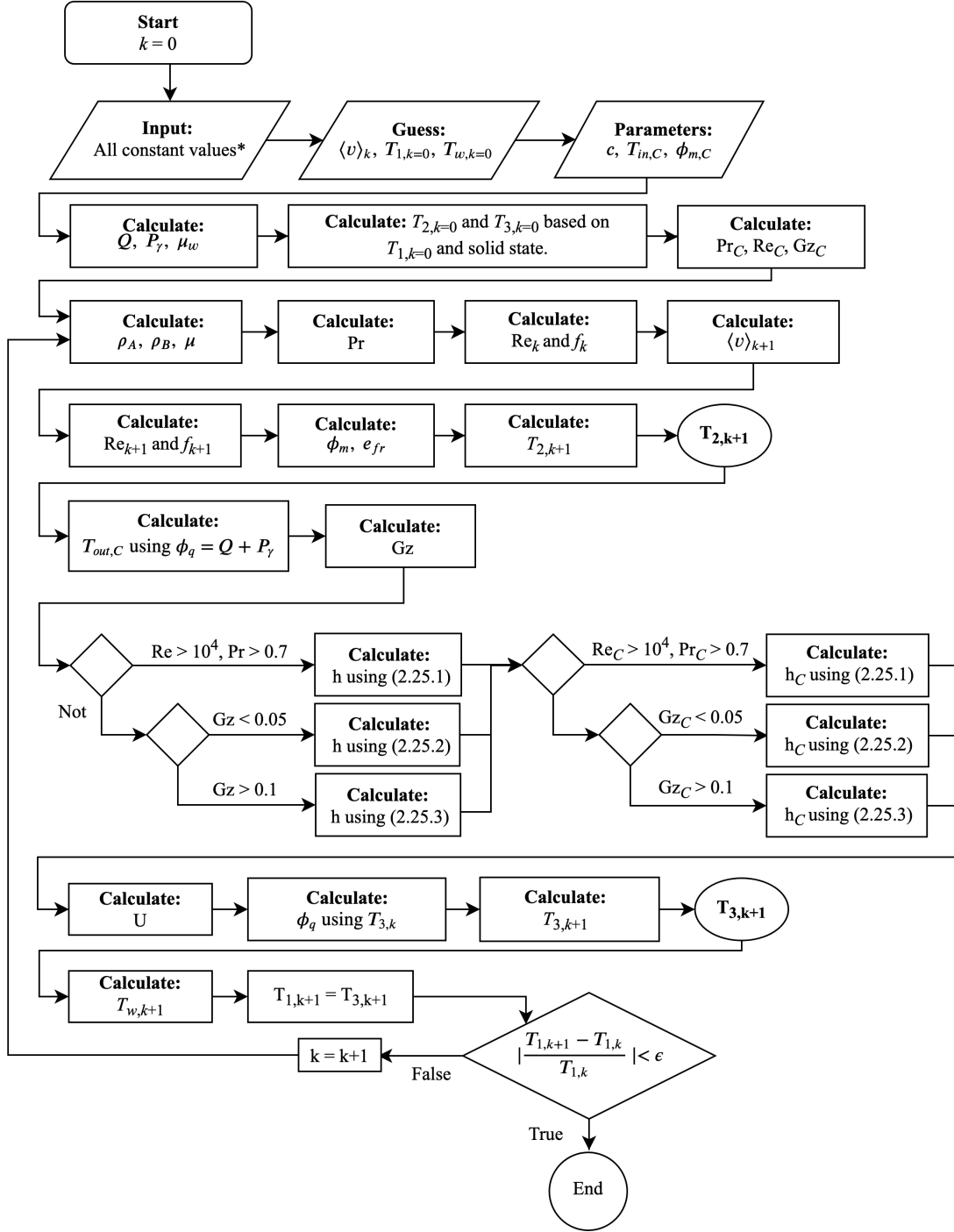


Figure 4.1: Flowchart of the iteration process. *These are: $r, D, \epsilon, l, r_{max}, \epsilon, T_0, g, d, E_f, \sigma_f, \phi_n, N_A, u, \rho_{zirc}, \lambda, c_p, \rho_0, a$, the enrichment, $\lambda_w, c_{p,w}, \rho_w, D_C, a_C$

The last test would be to make the cooler extremely powerful, in which case the temperatures in the loop should again drop to near the temperature of the cooling water. The cooler can be made more powerful by increasing U . As can be seen in figure 4.3, the temperature after the cooler T_3 indeed decreases towards the temperature of the cooling water (10 °C) when U is increased.

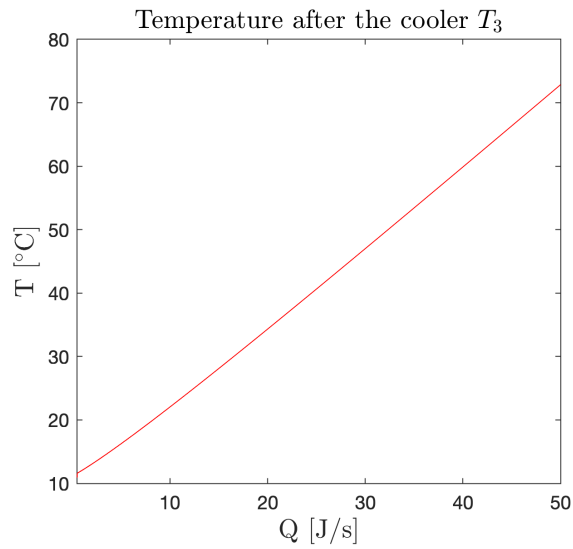


Figure 4.2: The temperature after the cooler T_3 vs. the heat production Q , for gamma heating set at $P_\gamma = 2$ J/s, radius $r = 4$ mm, and standard cooling conditions.

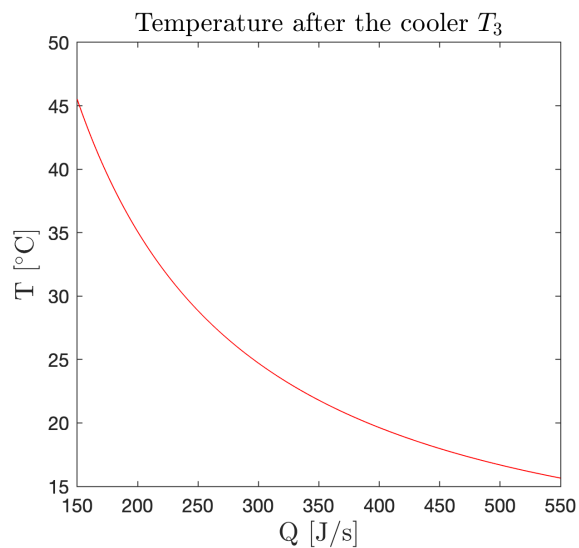


Figure 4.3: The temperature after the cooler T_3 vs. the total heat transfer coefficient U , for concentration $c = 30$ g/L, radius $r = 4.0$ mm and standard cooling conditions.

5 | Results

In this chapter, the results of the calculations done for this research will be shown. First, the optimal iteration error ϵ is determined. After that, the optimal radius of the tube is established. Then, for that radius, the results of the temperature and concentration calculations are given. As mentioned in section 4.2, all of the following results are made using the following cooling conditions, unless indicated otherwise:

Table 5.1: Standard cooling conditions.

Temperature	$T_{in,C} = 10 \text{ }^\circ\text{C}$
Flow	$\phi_{m,C} = 0.70 \text{ kg/s}$

$T_{in,C}$ is also referred to in this chapter as simply T_C . Please note that in all the plotted results, values above $100 \text{ }^\circ\text{C}$ do not have any physical meaning, since the uranyl nitrate solution will turn to vapor at these temperatures, making the laws and formulas used inapplicable.

5.1 Iteration error ϵ

Before calculating other results in this research, the optimal value of ϵ had to be established. This is the value of ϵ for which the iteration returns sufficient approximations, without ϵ being too small. This optimal value was found by calculating the maximum bulk temperature in the loop for different values for ϵ , varying from 10^{-1} to 10^{-10} . The values of the other variables can be chosen arbitrarily in this case, since the relation for the error remains the same. The results of these calculations are shown in figure 5.1. It shows that for an error of 10^{-5} or smaller, the iteration gives the same result for T_{max} , so $\epsilon = 10^{-5}$ is initially used as the error in all further calculations. However, for some situations this error was too demanding, causing the iteration to run endlessly. In those cases the error was raised to a value for which the iteration did give a final answer. As can be seen in figure 5.1, the iteration still gives acceptable results up to $\epsilon \approx 10^{-2}$, but for larger values of ϵ the approximations become relatively inaccurate.

5.2 Radius

Next, the optimal value for the radius r of the tube had to be determined. This is the radius for which the maximum temperature in the loop is the lowest (for a constant uranyl nitrate concentration). A low temperature namely means that the uranyl nitrate concentration could be increased, considering that the temperature

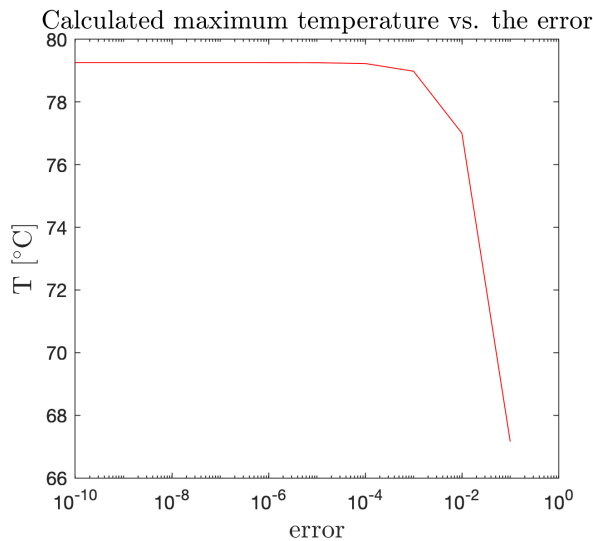


Figure 5.1: Calculated values for the maximum temperature in the loop vs. the error that was used in the iteration. This plot was made with radius $r = 3.5$ mm, concentration $c = 30$ g/L and standard cooling conditions (table 5.1).

is dependent on the concentration. Thus, the radius corresponding to the lowest maximum temperature in the loop allows for the highest possible uranyl nitrate concentration. In order to determine this optimal radius, the maximum temperature in the loop was calculated for different radii, followed by finding the minimum value of this temperature and the corresponding radius. The results of this calculation can be found in figure 5.2. For a concentration of $c = 30$ g/L, they show that the maximum temperature in the loop gets higher as the radius gets larger, and thus that the smaller the radius, the better it is for the maximum uranyl nitrate concentration in the loop. The optimal radius found is $r = 2.5$ mm, the smallest value used in the variation of r .

In the same calculation, a graphic of the average flow velocity in the loop for different radii was made, shown in figure 5.3. This is not relevant for further conclusions about the radius of the loop, but it is interesting to see how the bulk velocity is dependent on r , and it shows that natural circulation actually occurs.

The previous calculation for the maximum temperature in the loop has to be repeated for different concentrations, to determine whether the uranyl nitrate concentration has an influence on the optimal radius. The results of these calculations showed that also in the range of $c = 30$ g/L to $c = 100$ g/L, the optimal value of the radius was always 2.5 mm, the minimal value for the radius in these calculations.

A different way to determine the optimal radius, is by looking at the maximum concentration for each radius. This maximum concentration is found by varying the uranyl nitrate concentration in the loop and finding the concentration at which the maximum temperature in the loop hits the safety threshold of 90 °C. The results of these calculations can be seen in figure 5.4. They confirm the results of figure 5.2: the smaller the radius, the higher the uranyl nitrate concentration in the loop can be before the temperatures in the loop exceed 90 °C.

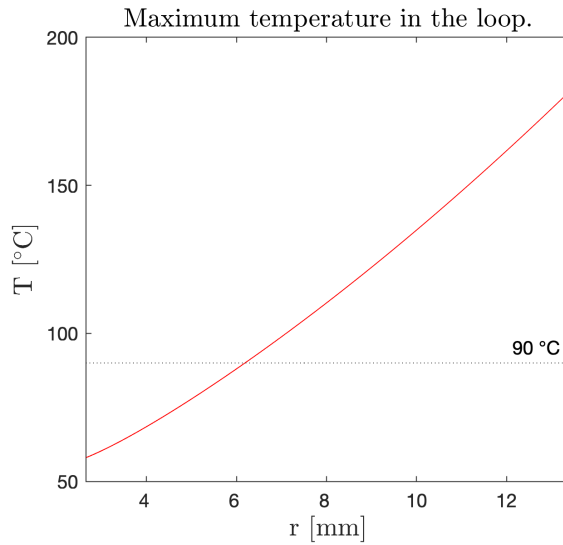


Figure 5.2: The maximum temperature in the loop (the maximum wall temperature) vs. the radius of the loop. The concentration used in this calculation was $c = 30$ g/L. Made with standard cooling conditions (table 5.1).

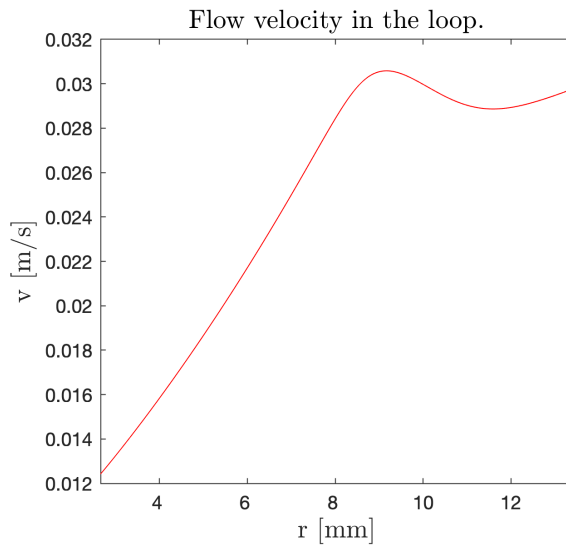


Figure 5.3: The average flow velocity of the uranyl nitrate solution in the loop vs. the radius of the loop. The concentration used in this calculation was $c = 30$ g/L. Made with standard cooling conditions (table 5.1).

The results in this section show that a small radius is beneficial for the temperatures in the loop, and thus for the uranyl nitrate concentration. Therefore, the radius of the tube for further the calculations is set at $r = 3$ mm. There are two reasons why it is not set smaller. The first reason, as will be further discussed in section 5.4, is that the MATLAB script has trouble with calculations for very small radii, and these problem start to occur when $r < 3$ mm. The second motivation is the production of molybdenum-99. While the production rate is linearly dependent on the uranyl nitrate concentration (as seen in section 3.4) and the goal of this research is to get that concentration as high as possible, the actual molybdenum-99 production is also

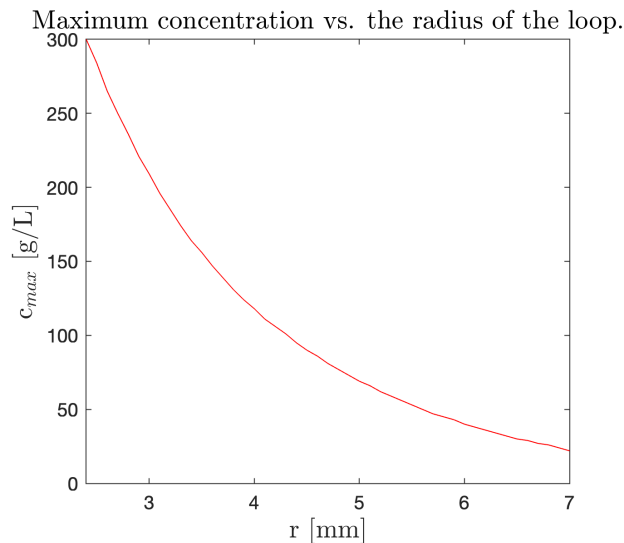


Figure 5.4: Maximum uranyl nitrate concentration vs. the radius of the loop. The error used in this calculation was $\epsilon = 0.005$. Made with standard cooling conditions (table 5.1).

linearly dependent on the volume of the irradiated part of the loop ($V = \pi r^2 l$), and thus on r^2 . Therefore, a larger radius is beneficial for the actual molybdenum-99 production of the loop. The volume of the loop for $r = 3$ mm is $2.3152 \cdot 10^{-05}$ m³, or approximately 0.0232 L.

5.3 Main results

The main calculations in this research are focused on the temperatures in the loop and the concentration of the uranyl nitrate solution, since these values determine whether the loop is safe to operate and feasible for the production of molybdenum-99 (research questions 2 and 3). Also, they focus on the average flow velocity in loop to see if natural circulation indeed occurs and if it drives the uranyl nitrate solution in the loop (research question 1). The first results of these calculations are the maximum bulk temperature, maximum wall temperature, and the average flow velocity in the loop, all for different concentrations and cooling conditions (see table 5.2). Some of these results can be seen in figures 5.5, 5.6 and 5.7. All these calculations were made using an error of $\epsilon = 0.005$. Note that the irregularities in the graphs are most likely due to the alternating calculations of the heat transfer coefficient h , since the three different calculations in equation 2.26 give different values for h .

Table 5.2: Variables for main calculations.

Cooling temperature	$1 \leq T_{in,C} \leq 30$ °C
Cooling flow	$0.01 \leq \phi_{m,C} \leq 0.70$ kg/s
Uranyl nitrate concentration	$30 \leq c \leq 300$ g/L

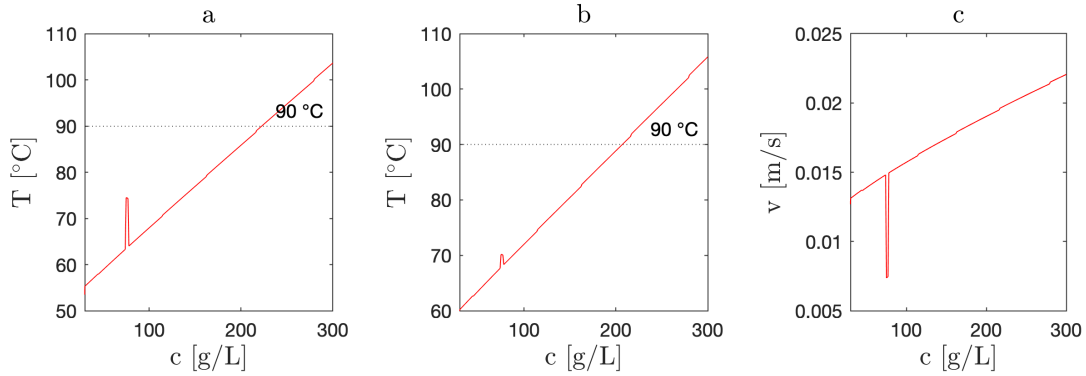


Figure 5.5: Maximum bulk temperature (a), maximum wall temperature (b), and average flow velocity in the loop (c). The temperature of the cooling water in this case was $T_C = 10$ °C and the cooling flow was $\phi_{m,C} = 0.50$ kg/s.

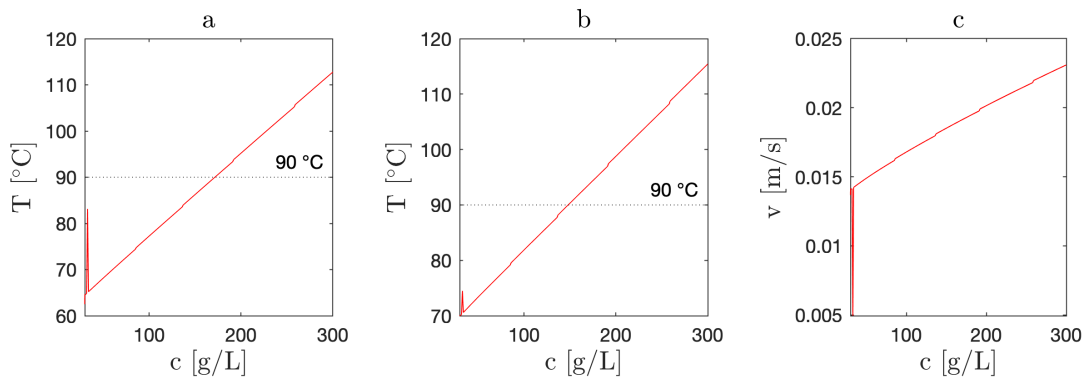


Figure 5.6: Maximum bulk temperature (a), maximum wall temperature (b), and average flow velocity in the loop (c). The temperature of the cooling water in this case was $T_C = 20$ °C and the cooling flow was $\phi_{m,C} = 0.50$ kg/s.

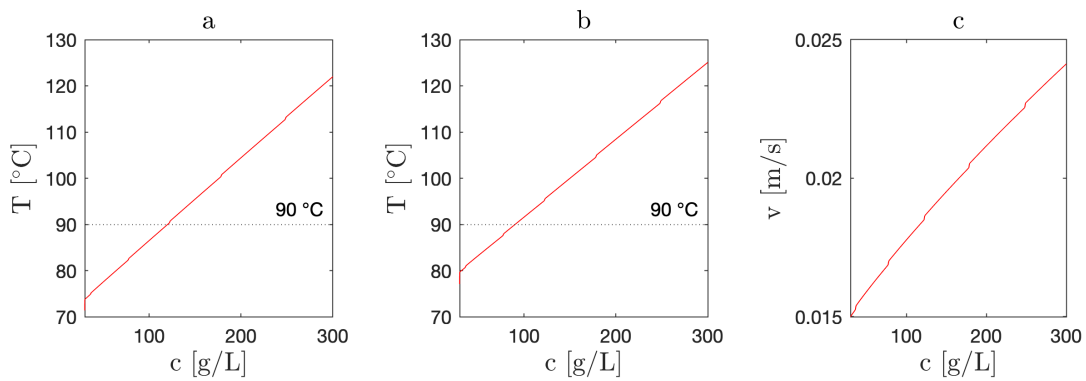


Figure 5.7: Maximum bulk temperature (a), maximum wall temperature (b), and average flow velocity in the loop (c). The temperature of the cooling water in this case was $T_C = 30$ °C and the cooling flow was $\phi_{m,C} = 0.50$ kg/s.

More important for the feasibility of the loop, is the maximum uranyl nitrate concentration for different cooling conditions. The results of these calculations can be seen in figure 5.8, where $\phi_{m,C}$ is varied on the horizontal axis and multiple plots for different values of T_C are shown.

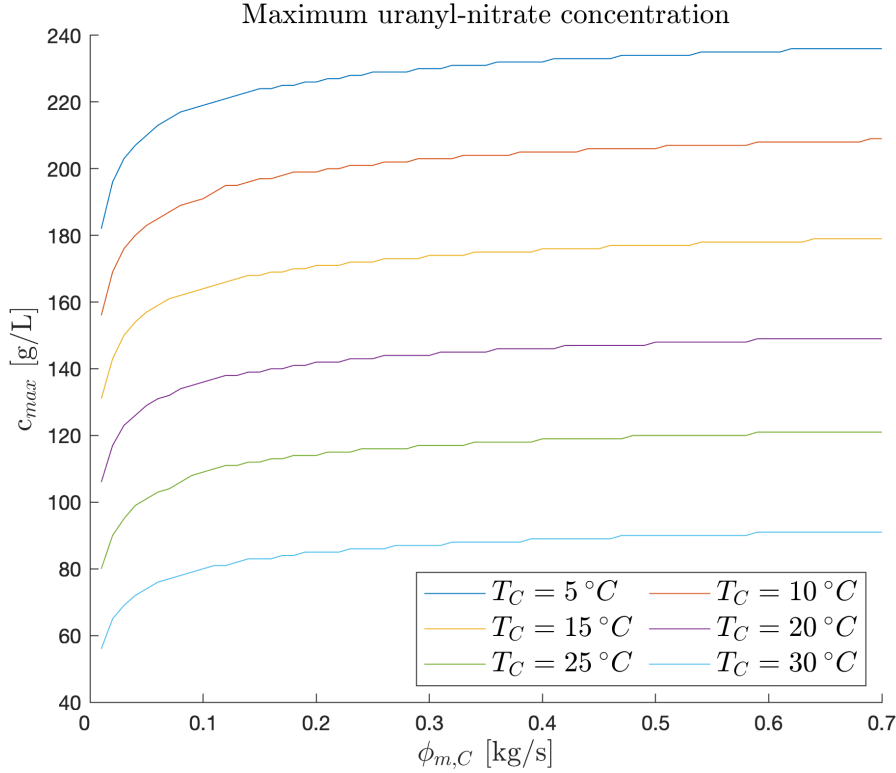


Figure 5.8: The maximum uranyl nitrate concentration in the loop for different values of the mass flow in the cooler $\phi_{m,C}$ and the initial temperature of the cooling water T_C .

5.4 Discussion

There are a few shortcomings that have to be mentioned and notes that need to be made about these results. First of all, the MATLAB code that was used, does not work for very small values of either the radius r or the uranyl nitrate concentration c . This is because the code contains a few square roots and logarithms (e.g. equations 2.20, 2.26 and 2.28) that give a complex output if what is inside them is either negative or zero, which can happen for very small values of r and c . Moreover, MATLAB works with only four decimals. If r and c are decreased, the difference between ρ_A and ρ_B becomes smaller. If the difference is so small that it is in the fifth decimal, it is too small for MATLAB to recognize, stopping the loop from continuing. This is not so much of a problem for the uranyl nitrate concentration, since the goal of this research is to have a high concentration, but for the radius it is. It was shown in section 5.2 that a small radius is beneficial for the temperatures and uranyl nitrate concentration in the loop, but a smaller radius than $r = 3$ mm could not be used due to this shortcoming.

On a more general note, this is the very first look into this loop design, and therefore the model used has been oversimplified a lot. Of course there is a lot more happening in the loop than just two sections that are either heated or cooled. The vertical parts of the loop are left out of the model in this research, but in reality it will be impossible to completely shield them from neutrons and gamma rays, so the fluid will be affected in those parts as well. Also, in a real world situation, the loop as a whole will be surrounded by water or some other cooling fluid, resulting in a heat flux from the loop to the surrounding fluid. In this research, a worst case scenario was assumed where there was no extra cooling at all.

Another note is that the cooling used in this research reaches very high flow velocities in this setup. Perhaps too high for the loop to be feasible, since they are higher than most companies suggest as a maximum for their piping [5]. This problem of very high flow velocities could be fixed by using a concentric heat exchanger with a bigger diameter, or a different type of cooler. On the other hand, even with low values for the mass flow (and thus the flow velocity) in the cooler that was used in these calculations, the maximum concentrations are still high and thus relevant in the conclusions of this research. All in all, this research offers a simplistic overview of the new design. The results are valid, but should be interpreted with some caution.

6 | Conclusions

In this research, the goal was to investigate whether a natural circulation driven loop with a maximum volume of 0.5 L would be possible and feasible for the production of molybdenum-99. The maximum temperature in the loop was not allowed to exceed 90 °C and the maximum uranyl nitrate concentration was calculated for different cooling conditions. This was done by making a simplified model of the loop, with an irradiated section and a cooled section, and calculating the transport of heat and momentum in this model. The results of these calculations were shown in chapter 5.

6.1 Conclusion

The goals of this research have been achieved, and the results look promising. First of all, they demonstrate that natural circulation indeed occurs in the loop that was designed in this research and thus that it is possible to design a loop that is driven by natural circulation. Figures 5.5, 5.6 and 5.7 show that the uranyl nitrate solution will flow with an average velocity of roughly 0.015-0.020 m/s, or 1.5-2.0 cm/s. The velocity slightly increases as the concentration gets higher, and slightly decreases as the cooler gets more powerful.

More importantly, the results indicate that the new loop could achieve much higher uranyl nitrate concentrations than the current research loop (as described by Pendse [23], Elgin [12], Huisman [16] et al.), while keeping the temperatures in the loop below a safe 90 °C. When using a relatively strong cooler, with an inlet temperature of 5 °C and a mass flow of 0.70 kg/s, the maximum uranyl nitrate concentration can be up to 236 g/L. This concentration is almost nine times higher than the one in the current loop, and close to the ideal concentration of 310 g/L. But also for less powerful cooling conditions (with a lower and more realistic flow velocity, as mentioned in section 5.4), the maximum concentration is still very high. For instance, if the inlet temperature is 15 °C (the average temperature of tap water in the Netherlands [30]) and the mass flow is 0.20 kg/s (which is equivalent to approximately 2.4 m/s in this setup), the maximum concentration is still 171 g/L. Even for a moderate cooler of 30 °C and 0.10 kg/s, the maximum concentration is still almost three times higher than in the current loop: 80 g/L. So the results show high potential for the new loop design.

6.2 Recommendations

The first recommendations for further research follow directly from the discussion of the shortcomings of this research. First and foremost, a more detailed and extensive model of the loop should be made, in order to calculate the temperatures and maximum concentrations in the loop more accurately. This could be done in place, by dividing the loop in more than just two sections and making a three dimensional model, as well as in time, by not assuming steady state. Also, the surroundings of the loop should be taken into account in a more accurate and realistic model. Secondly, a solution could be found to do calculations for smaller radii than 3 mm, to see if that would give even better results. And lastly, the cooler of the setup requires more attention. In this research, a concentric tube, countercurrent heat exchanger was used, but there might be a more efficient way of cooling available, e.g. a shell and tube heat exchanger or a plate heat exchanger (especially when combined with the suggestion about the actual use of this loop in the production of molybdenum-99, made in section 6.2.2).

6.2.1 Gammaheating

Another factor in the design of the loop that stood out during the calculations, was the gamma heating. The heat production due to gamma radiation was 19.75 J/s in this design, which is roughly in the same order of magnitude as the heat production by nuclear fission. (For example, for $c = 85$ g/L, $\phi_{m,C} = 0.7$ kg/s and $T_{in,C} = 30$ °C, $Q = 32.14$ J/s.) Therefore, it would be considerable to investigate whether the gamma heating could be reduced. This could be done by reducing the thickness of the tube wall. Lowering the thickness from 2 mm to 1 mm, for example, would result in a gamma heating of just 8.64 J/s. When gamma heating is accounted for in all parts of the loop, not just the irradiated part of the loop as was done in this study, the difference would be even more relevant. However, reducing the thickness of the wall may have consequences for the durability of the loop, since a thinner tube will be more likely to be damaged by corrosion. Reducing the thickness of the tube wall could thus be beneficial for the amount of gamma heating, but also dangerous for the safety of the loop. Hence, this possibility should be further investigated.

6.2.2 Molybdenum-99 production

The last aspect of the loop that this study has not explored is the actual production and extraction of molybdenum-99. It has to be investigated how the molybdenum-99 should be extracted from the loop and if this would affect the natural circulation in the loop. Drawing off uranyl nitrate solution from the loop for molybdenum-99 extraction could for instance alter the flow in the loop. Also, looking a little further into the future, it would be interesting to look at the implementation of this loop. It is clear that, despite the high production rate, the production of the new loop on its own will not even come close to the production of the current loop. Since the volume of the new loop is deliberately kept small, with the irradiated part of the new loop (roughly 1/4 of the total volume) being a little more than 860 times smaller than

the current loop (that is irradiated as a whole), the uranyl nitrate concentration in the new loop would have to be about $24 \cdot 10^3$ g/L to achieve the same level of production, being an impossible situation.

A solution to this problem could be to increase the volume by using multiple loops side-by-side. Say the uranyl nitrate concentration that could be achieved in the new loop is 150 g/L, which is five times more than in the current loop, then the combined volume of the irradiated parts of the loops could be just 1 L. That would make the total volume just 4 L instead of 5 L. Also, these loops could be emptied or replaced separately, reducing the amount of radioactive material in case of any malfunctions. However, such a system would require a total of about 170 loops, which would introduce a whole variety of new challenges. For instance, the neutron flux will not have the same, high value in every loop, and the heat transport from one loop to another will play a significant role. However, the cooling will probably be much more efficient than that of the research loop that was being designed before this research, because of the much larger contact area. The high production rate in combination with a large volume, could thus mean that a setup with multiple mini loops is a very efficient way to produce molybdenum.

All in all, this first global study shows promising results and demonstrates a lot of positive aspects to this new design for a molybdenum-99 producing mini loop, the MPML, as well as the suggested implementation of multiple MPMLs. It is however a limited study, and it also predicts some possible drawbacks. Therefore it is surely recommended to do further, more extensive research into this new loop design, with high expectations for the final result.

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Appendices

A | MATLAB code

```
%% Initialize

clear all
close all
clc

% Input
% system
r = 0.003;
D = 2*r;
eps = 1.5e-6; % effective roughness
l = 0.20;
r_max = 0.0134;
if r>r_max
    disp('r is too big for this setup.')
    return
end
V = 4*l*pi*r^2+2*pi^2*r^3;
disp('Volume ='), disp(V);
error = 0.005;
T_0 = 60; % celsius
g = 9.81;
d = 0.002;
enr = 0.1975; % enrichment

% fission
E_f = 192*1.60217662e-13; % MeV to Joules
sigma_f = 583e-28;
phi_n = 3.5e16;
N_A = 6.022e23;

% gamma
u = 300; % W/kg
rho_z = 6.55e3; % density zircaloy kg/m3
V_tube = 1/4*pi*l*(4*d^2+8*d*r);

% uranyl nitrate solution
lambda = 0.665;
c_p = 2905.5;
rho_0 = 1330.6;
a = lambda/(rho_0*c_p);
beta = 0.00052;

% cooler
lambda_w = 0.607;
```

```

rho_w = 999.63;
c_pw = 4185;
D_C = D;
a_C = 0.158e-6;

% Guess
v(1) = 0.01;
T1(1) = 40; %celcius
T3(1) = T1(1);
T_w(1) = 40;

%Calculations
P_gamma = u*rho_z*V_tube;

% Text strings
s1 = ['90 ' char(176) 'C'];
s2 = ['T [^\circ]C'];

%% Iteration
T_max = zeros(100,70,30);
T_wmax = zeros(100,70,30);
v_loop = zeros(100,70,30);
phi_m_loop = zeros(100,70,30);
c_max = zeros(70,30);

for o = 1:30 %variation of cooling temperature
Tin_C = o;
mu_w = 10^(-3)*1.002*10^((1.1709*(20-Tin_C)-0.001827*(Tin_C-20)^2)/(...
↪ Tin_C+89.93));

for j = 1:70 %variation of cooling flow
phi_mC = 0.01*j;
v_C = phi_mC/(3*pi*r^2*rho_w);

Pr_C = c_pw*mu_w/lambda_w;
Re_C = rho_w*v_C*D_C;
Gz_C = a_C*l/(D_C^2*v_C);

for i = 30:300 %variation of concentration
c = i;
c_mol = c/394.04; %concentration mol/L
N = c*N_A/0.39404;
Q = pi*r^2*l*E_f*sigma_f*enr*N*phi_n;

T2(1) = T1(1)+(1/v(1)*Q)/(c_p*rho_0*pi*r^2*l);

k = 1;
diff = 1;

while abs(diff) > error %iteration

rho_A = rho_0/(1+(T2(k)-T_0)*beta);
rho_B = rho_0/(1+(T3(k)-T_0)*beta);

T_mean = (T1(k)+T2(k))/2;
mu_ref = 10^(-3)*1.002*10^((1.1709*(20-T_mean)-0.001827*(T_mean...
↪ -20)^2)/(T_mean+89.93));
mu = mu_ref*(1-0.1687*sqrt(c_mol)+0.7904*c_mol);

```

```

Pr = c_p*mu/lambda;

Re(k) = rho_0*v(k)*D/mu;
a = 1/(1+(Re(k)/2712)^8.4);
b = 1/(1+(Re(k)/(150*D/eps))^1.8);
f(k) = (64/Re(k))^a*(0.75*log(Re(k)/5.37))^(2*(a-1)*b)*(0.88*log...
    ↪ (6.82*D/eps))^(2*(a-1)*(1-b));

v(k+1) = sqrt(g*r^2*(rho_B-rho_A)/(2*D*f(k)*rho_0));

Re(k+1) = rho_0*v(k+1)*D/mu;
a = 1/(1+(Re(k+1)/2712)^8.4);
b = 1/(1+(Re(k+1)/(150*D/eps))^1.8);
f(k+1) = (64/Re(k+1))^a*(0.75*log(Re(k+1)/5.37))^(2*(a-1)*b)*(0...
    ↪ .88*log(6.82*D/eps))^(2*(a-1)*(1-b));

phi_m(k+1) = rho_0*pi*r^2*v(k+1);
e_fr = f(k+1)*l/D*(1/2)*(v(k+1))^2;

T2(k+1) = T1(k)+(Q+P_gamma+phi_m(k+1)*e_fr)/(phi_m(k+1)*c_p);

Tout_C = Tin_C+(Q+P_gamma)/(phi_mC*c_pw);

Gz = a*l/(D^2*v(k+1));

if Re(k+1) > 10^4 & Pr ≥ 0.7
    h = 0.027*Re(k+1)^0.8*Pr^0.33*lambda/D;
elseif Gz < 0.05
    h = 1.62*Gz^(-1/3)*lambda/D;
else
    h = 3.66*lambda/D;
end

if Re_C > 10^4 & Pr_C ≥ 0.7
    h_C = 0.027*Re_C^0.8*Pr_C^0.33*lambda_w/D_C;
elseif Gz_C < 0.05
    h_C = 1.62*Gz_C^(-1/3)*lambda_w/D_C;
else
    h_C = 3.66*lambda_w/D_C;
end

U = (1/h+1/h_C+d/lambda_w)^(-1);
phi_q = U*pi*D*l*((T2(k+1)-Tout_C)-(T3(k)-Tin_C))/(log(T2(k+1)-...
    ↪ Tout_C)-log(T3(k)-Tin_C));

T3(k+1) = T2(k+1)+(phi_m(k+1)*e_fr-phi_q)/(phi_m(k+1)*c_p);
T1(k+1) = T3(k+1);
T_meank = (T1(k)+T2(k+1))/2;
T_w(k+1) = P_gamma/(pi*D*l*h)+T_meank;

diff = (T1(k+1)-T1(k))/T1(k);

k = k+1;
end

T_max(i,j,o) = T2(k);
T_wmax(i,j,o) = T_w(k);

```

```

v_loop(i,j,o) = v(k);
phi_m_loop(i,j,o) = phi_m(k);
end

cmax = find(T_wmax(:,j,o)>90,1)-1;
if isempty(cmax) == 1
    cmax = 300;
end

c_max(j,o) = cmax;
end

if (o/5) == fix(o/5)
s3 = ['T_C = ', num2str(o), char(176), 'C'];
figure('Position',[50 50 600 200],'Resize','off','Name',s3);
i = 1:300;
subplot(1,3,1,'OuterPosition',[0 0 0.3 1],'Box','on'), hold on;
plot(i,T_max(i,50,o),'r'), yline(90,'k:',s1);
xlim([30 300]);
title('a','Interpreter','latex','fontsize',14), xlabel('c [g/L]','...
    ↪ Interpreter','latex','fontsize',14), ylabel(s2,'Interpreter','...
    ↪ latex','fontsize',14);
subplot(1,3,2,'OuterPosition',[0.33 0 0.3 1],'Box','on'), hold on;
plot(i,T_wmax(i,50,o),'r'), yline(90,'k:',s1);
title('b','Interpreter','latex','fontsize',14), xlabel('c [g/L]','...
    ↪ Interpreter','latex','fontsize',14), ylabel(s2,'Interpreter','...
    ↪ latex','fontsize',14);
xlim([30 300]);
subplot(1,3,3,'OuterPosition',[0.66 0 0.3 1],'Box','on'), hold on;
plot(i,v_loop(i,50,o),'r');
title('c','Interpreter','latex','fontsize',14), xlabel('c [g/L]','...
    ↪ Interpreter','latex','fontsize',14), ylabel('v [m/s]','...
    ↪ Interpreter','latex','fontsize',14);
xlim([30 300]);
print(['temperatures_T_C_', num2str(o)], '-dpng', '-r300')
end
end

%% plot phi_mC vs. c_max
figure('Position',[50 50 500 400])
j = 1:70;
hold on;
plot(j,c_max(j,5)), plot(j,c_max(j,10)), plot(j,c_max(j,15)), plot(j...
    ↪ ,c_max(j,20));
plot(j,c_max(j,25)), plot(j,c_max(j,30));
label(1) = legend('$T_C = 5\$: ^{\circ}C$', '$T_C = 10\$: ^{\circ}C$', '...
    ↪ $T_C = 15\$: ^{\circ}C$', '$T_C = 20\$: ^{\circ}C$', '$T_C = 25\$: ...
    ↪ ^{\circ}C$', '$T_C = 30\$: ^{\circ}C$', 'Location','southeast','...
    ↪ Orientation','horizontal','NumColumns',2);
label(2) = title('Maximum uranyl-nitrate concentration');
label(3) = ylabel('c_{max} [g/L]');
label(4) = xlabel('$\phi_{m,C}$ [kg/s]');
xtk = get(gca, 'XTick');
xtklbl = xtk*0.01;
set(gca, 'XTick', xtk, 'XTickLabel', xtklbl);
set(label, 'Interpreter', 'latex', 'fontsize', 14);
print('main_results', '-dpng', '-r300')

```