



Master of Science Thesis

Development of a Niobium target for routine  
production of [ $^{18}\text{F}$ ]fluoride with a MC 16  
Scanditronix cyclotron

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## Abstract

The purpose with this work has been to develop and construct a Niobium target for the production of [ $^{18}\text{F}$ ]fluoride with a MC 16 Scanditronix cyclotron. The future demand larger amounts of [ $^{18}\text{F}$ ]fluoride and to satisfy the forthcoming needs an enhanced target needs to be installed on the cyclotron. The old target could have provided the forthcoming needs but to obtain good margins it is important with a better target that benefits the full potential of a MC 16 Scanditronix cyclotron. The old target could only stand approximately half the proton beam capacity of the cyclotron.

The work was carried out at the department of Medical Radiation Physics, Jubileum Institute and at the University hospital of Lund. Possible enhancements with the old target were identified and incorporated into the new target. Target parts were constructed in the hospital's own workshop meanwhile more advanced gadgets had to be ordered from specialized firms. The material of the target chamber was carefully chosen (Niobium) according to chemical inertness, mechanical strength and activation (radioactive isotopes generated when irradiated). Mathematical calculations were applied onto real situations to estimate pressure tolerances and heat transportations. Different foil materials were investigated. Irradiation experiments, of protons into the enriched  $\text{H}_2^{18}\text{O}$ -water, were performed with a MC 16 Scanditronix cyclotron.

The results are very good. For one hour irradiation at  $40\ \mu\text{A}$ , 93 GBq  $\text{F}^-$  was produced (extrapolated values) meanwhile for a two hour irradiation at  $40\ \mu\text{A}$ , 142 GBq  $\text{F}^-$  was produced (95 %  $^{18}\text{O}$ -water). The following synthesis resulted in 87 GBq which correspond to a yield of 62 %. The yield for the FDG synthesis is normal and the productions are large. No discoloration of water occurred. The chemical inertness of the Niobium material has been verified.

The new target has increased the production capacity of approximately 300 %, from 32 GBq for the old target, to 93 GBq for the new target (1 hour irradiation). The saturation value per micro amperes is approximately 7.30 GBq/ $\mu\text{A}$ . The biggest improvement originates from the new target's capacity to benefit the full proton beam intensity capacity of the MC 16 Scanditronix cyclotron. The Niobium target gives better margins when the demands will increase.

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# 1. Introduction

$^{18}\text{F}$  is a  $\beta^+$ -emitter (96.9 %) with a half life of 109.8 minutes. In the beginning,  $^{18}\text{F}$ , was produced by reactors in a two step process via the reactions:  $^6\text{Li}(n,\alpha)t$  and  $^{16}\text{O}(t,n)^{18}\text{F}$ . Nowadays cyclotron production is preferred because of, yield and physical and chemical state of the product.

There are many ways to produce  $^{18}\text{F}$  according to table 1.1. The two most favorable ones are the  $^{18}\text{O}(p,n)^{18}\text{F}$  and  $^{20}\text{Ne}(d,\alpha)^{18}\text{F}$  since these reactions only requires moderate particle energies and moderate beam intensities [1].

Reaction	Target	E (MeV)	Thick target yield <sup>a</sup>	Main form	Reference
$^{18}\text{O}(p,n)^{18}\text{F}$	$^{18}\text{O}_2$	14-0	216 mCi/ $\mu\text{A}$ <sup>b,c</sup>		Ruth and Wolf (1979)
	$^{18}\text{O}_2$	10-0	150 mCi/ $\mu\text{A}$ <sup>c</sup>	$^{18}\text{F}]\text{F}_2$	Nickles <i>et al.</i> (1984)
	$\text{H}_2^{18}\text{O}$	16-0	110 mCi/ $\mu\text{A}$ <sup>c</sup>	$^{18}\text{F}]\text{F}^-$	Kilbourn <i>et al.</i> (1985)
$^{20}\text{Ne}(d,\alpha)^{18}\text{F}$	$^{20}\text{Ne}$	14	91.9 mCi/ $\mu\text{A}$ <sup>b,c</sup>		Casella <i>et al.</i> (1980)
	0.1% $\text{F}_2/\text{Ne}$	14-2	12.2 mCi/ $\mu\text{Ah}$ <sup>d</sup>	$^{18}\text{F}]\text{F}_2$	Casella <i>et al.</i> (1980)
	0.18% $\text{F}_2/\text{Ne}$	11.2-0	10 mCi/ $\mu\text{Ah}$	$^{18}\text{F}]\text{F}_2$	Blessing <i>et al.</i> (1986)
	15% $\text{H}_2/\text{Ne}$	11.2-0	10 mCi/ $\mu\text{Ah}$	$^{18}\text{F}]\text{HF}$	Blessing <i>et al.</i> (1986)
	6.7% $\text{H}_2/\text{Ne}$	11.2-0	8 mCi/ $\mu\text{Ah}$	$^{18}\text{F}]\text{F}^-$	Blessing <i>et al.</i> (1986)
$^{20}\text{Ne}(d,x)^{18}\text{Ne}^e$	10% $\text{H}_2/\text{Ne}$	6.3-0	11 mCi/ $\mu\text{A}$ <sup>c</sup>	$^{18}\text{F}]\text{HF}$	Dahl <i>et al.</i> (1980)
$^{16}\text{O}(\alpha,d)^{18}\text{F}$	$\text{H}_2\text{O}$	30	1.1 mCi/ $\mu\text{Ah}$	$^{18}\text{F}]\text{F}^-$	Clark and Silvester (1966)
		48	7.0 mCi/ $\mu\text{Ah}$	$^{18}\text{F}]\text{F}^-$	Lindner <i>et al.</i> (1973)
$^{16}\text{O}(\alpha,2n)^{18}\text{Ne}^e$	$\text{O}_2$	40	14 mCi/ $\mu\text{Ah}$	$^{18}\text{F}]\text{HF}$	Nozaki <i>et al.</i> (1968)
$^{16}\text{O}(^3\text{He},p)^{18}\text{F}$	$\text{H}_2\text{O}$	41-14	7 mCi/ $\mu\text{Ah}$	$^{18}\text{F}]\text{F}^-$	Fitschen <i>et al.</i> (1977)
$^{16}\text{O}(^3\text{He},n)^{18}\text{Ne}^d$	$\text{H}_2\text{O}$	36	7.6 mCi/ $\mu\text{Ah}$	$^{18}\text{F}]\text{F}^-$	Knust and Machulla (1983)
$^{20}\text{Ne}(^3\text{He},\alpha n)^{18}\text{Ne}^e$	2% $\text{H}_2/\text{Ne}$	27.5	5-7 mCi/ $\mu\text{Ah}$	$^{18}\text{F}]\text{HF}$	Crouzel and Comar (1978)
		n flux ( $\text{cm}^{-2}\text{s}^{-1}$ )	Yield		
$^{16}\text{O}(^3\text{H},n)^{18}\text{F}$	$\text{Li}_2\text{CO}_3$	$3 \cdot 10^{13}$	200 mCi/3h	$^{18}\text{F}]\text{F}^-$	Vera Ruiz (1988)
	$^6\text{LiOH} \cdot \text{H}_2\text{O}$	$3 \cdot 10^{13}$	250 mCi/3h	$^{18}\text{F}]\text{F}^-$	Vera Ruiz (1988)

<sup>a</sup>For 1 h experimental irradiation unless otherwise indicated by superscript.

<sup>b</sup>Theoretical yield

<sup>c</sup>Saturation yield

<sup>e</sup> $^{18}\text{Ne}$  decays to fluorine-18 with a half-life of 1.67 s

Table 1.1: Literature data for the production of  $^{18}\text{F}$  for medical use from different nuclear reactions on thick targets

In the fifties the rectilinear scanner was discovered by Ben Cassen. For many years it served as a standard imaging device where  $^{18}\text{F}$ fluoride was one of the useful radioisotopes. In the sixties  $^{18}\text{F}$ fluoride was used as a bone scanning agent. Due to the high gamma energy ejected (511 keV) no suitable camera with appropriate collimator was available. For a 511 keV photon, the walls of the collimator holes had to be very thick to avoid penetration that otherwise resulted in false events. The thick walls resulted in dotted pictures and very heavy collimators. When  $^{99\text{m}}\text{Tc}$ -labeled phosphonate derivatives became available they quickly won the competition in the bone imaging area. The gamma energy from  $^{99\text{m}}\text{Tc}$  (140 keV) is more favorable for a gamma camera.

When the PET-technique (Positron Emission Tomography) entered the market,  $^{18}\text{F}$  found new applications in the nuclear medicine because of its  $\beta^+$ -decay. The annihilation photons can be detected in coincidence defining a line. From a large number of such lines the activity distribution can be calculated with mathematical algorithms.

In the same period of time, the glucose analogue 2- $^{18}\text{F}$ fluoro-2deoxy-D-glucose (FDG) was introduced. The molecule carries one radioactive  $^{18}\text{F}$  in position 2. The FDG uses the same transportation into the cell as glucose. Inside the cell, FDG is metabolized, only one step, to 2- $^{18}\text{F}$ FDG-6- $\text{PO}_4$  that remains trapped inside the cell. The concentration of radioactive metabolite grows with time in proportion to the cells metabolism of glucose. This results in a greater accumulation of  $^{18}\text{F}$ -activity in tissues where the glucose metabolism is higher than for normal tissues (ex cancer tumors). This activity distribution can be detected and visualized by a PET-camera.

Before 1986, FDG could only be synthesized from fluorine,  $^{18}\text{F}$  $\text{F}_2$ , with the available syntheses [2,3,4]. When the Hamacher [5] synthesis entered the market, FDG could be manufactured from fluoride,  $^{18}\text{F}^-$ . With a much better yield (~50 %) it resulted in an expansion of water targets. The advantages of water targets is the easiness in handling of water and the fact that no carrier is needed to extract the  $^{18}\text{F}$ . One negative aspect is the cost of enriched water. Normal water, mainly consist of  $^{16}\text{O}$  and only 0.2 %  $^{18}\text{O}$ . The reaction requires enriched water and therefore expensive water. This results in small targets of some milliliter volumes.

FDG is the most used radiopharmaceutical in the PET technique

The reasons why the  $^{18}\text{F}$  radionuclide is a suitable positron emitter in PET facilities are many:

1. 96.9 %  $\beta^+$ -decay
2. The half life is 109.8 min and is therefore manageable and results in acceptable absorbed doses for the patient
3. It is possible to label different compounds that trace physiological and biochemical processes with  $^{18}\text{F}$ .  $^{18}\text{F}$  is useful as a hydrogen substitute  
The use of isotopes of the biologically ubiquitous elements makes it possible to label radiopharmaceuticals that trace biochemical processes precisely
4. The positron energy is low and does not influence the spatial resolution

Important radio nuclides are presented in table 1.2.

<b>Radionuclide</b>	<b><math>T_{1/2}</math> (min)</b>	<b><math>\beta^+</math>-decay (%)</b>	<b>Max. positron energy (MeV)</b>
$^{18}\text{F}$	109.8	96.9	0.64
$^{11}\text{C}$	20.4	99.8	0.96
$^{15}\text{O}$	2.04	99.9	1.72
$^{13}\text{N}$	9.96	100	1.19

Table 1.2: Most used  $\beta^+$  radioisotopes

## **1.1 Reasons and backgrounds for a new target**

The purpose with this work has been to develop a target for enriched  $^{18}\text{O}$ -water in production of radioactive  $^{18}\text{F}$ fluoride with a MC16 Scanditronix cyclotron. The old target [6] made of silver has provided the facility in Lund with  $^{18}\text{F}$ fluoride for some years. Now there is an interest in a new target mainly because of two new combined PET-CT cameras that will be installed at the University Hospital of Lund and Malmö Hospital. The forthcoming need will result in two daily productions. Today's weekly schedule consists of a production every other Monday for Gothenburg, a production for Växjö and Lund every Thursday and a production every Friday for Lund. The old target could have provided the forthcoming needs but to obtain good margins it is important to develop an enhanced target that benefits the full potential of a MC 16 Scanditronix cyclotron.

The drawbacks with the old target are:

1. The material (silver) is not optimal according to chemical inertness
2. Open system which leads to refills with enriched water during longer production runs
3. Support bars that "steals" intensity from the beam
4. It only benefits half of the cyclotron's proton beam current capacity due to the thin water layer
5. High energy degradation due to thick target foil

An improved design with a better material (more chemical resistant), deeper target and a closed type will result in a better target that can stand higher beam intensities. As a result of higher beam intensities and improved design the production can be increased and therefore provide the facility with larger amounts of  $^{18}\text{F}$ fluoride.

A closed target gives a higher pressure inside the chamber than an open target does. This must be taken under consideration while constructing the chamber parts, especially the foils which are a critical part of the construction.

Other reasons why one would like to construct a new target can be explained through the nature of a true physicist: curiosity and the will to improve. When constructing the target new ideas and perspectives can be borne and viewed which in the future can be shown to be valuable.

## 2. Material and methods

### 2.1 The optimization of a target

The available accelerator for the production of [ $^{18}\text{F}$ ]fluoride in Lund is a MC 16 Scanditronix cyclotron. It accelerates protons to the energy of 17 MeV. It is an  $\text{H}^+$  cyclotron with a deflection system to extract the beam. The beam has no focusing part i.e. a beam line. The principle of a water target is as in figure 2.1. Since the old Helium-cooling part will be reused, all efforts and focus will be on the target part lined red in the picture.

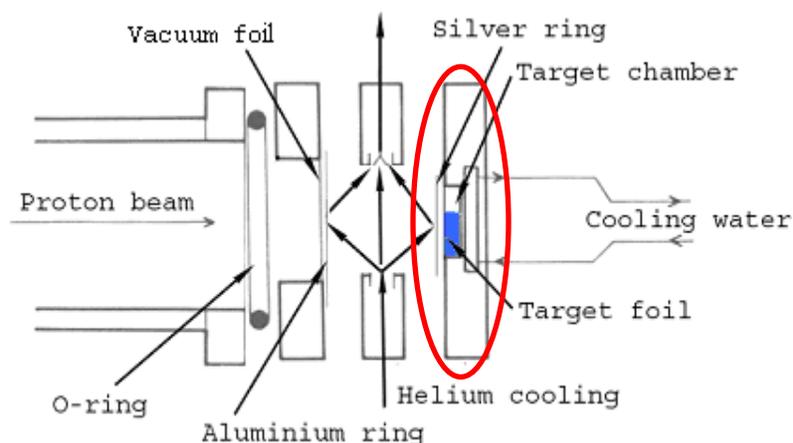


Figure 2.1: Principle of a water target

The proton beam profile is visualized in figure 2.2. The beam profile fits into a rectangle 4 times 1 centimeter. The target dimensioning has to be guided by this beam profile.



Figure 2.2: Beam profile of the MC 16 Scanditronix cyclotron in the target foil caused by burn mark from the beam. The beam profile encloses the light brown grey area in the middle. The swelling dot to right is a hot spot

In a first approach it is important to verify those factors and parameters that are involved in the improvement of a target. A good start is to study the formulas for production of activity (A), the activations formulas derived in appendix A1.1a and A1.1b.

The more proton beam intensity ( $\mu\text{A}$ ) put into the reaction with water the more activity is produced. The MC 16 Scanditronix machine has the capacity to deliver beam currents up to 40-50  $\mu\text{A}$ . At this date the old target only can benefit beam intensities up to 22  $\mu\text{A}$  before the yield starts to drop and the water gets discolored [7]. A risk with putting in much intensity in a small and thin water volume is that the beam misses the water target

because of boiling bubbles and cavitations [8]. To avoid this, the target must be constructed, geometrically, in an optimized way. The depth has to be increased because the yield for the old target drops at higher beam intensities. A thicker water volume will compensate for bubbles and cavitations. The new target design incorporates calculations of heat transfers, target depth and dimensioning that takes gas formations and pressure builds up under consideration. Another disadvantage with the old silver target is the formation of colloids during the irradiation process. The colloids give a lower yield of [ $^{18}\text{F}$ ]fluoride and obstructions in valves and tubing [9]. A new material that is more chemical resistant and that can stand higher intensities must be found.

Another drawback with the old target is that the system is open. When irradiating the enriched  $^{18}\text{O}$ -water, the production of oxygen and hydrogen gas will start due to radiolysis [8]. When the chamber is of an open type i.e. it is open under irradiation, the gases will exit through ventilation which results in a water level that successively decreases. A closed target would terminate these problems.

A refill is necessary to prevent that:

- A part of the beam will miss the water which will result in a lower yield
- The cooling effect of the foil will decrease. The water in the chamber has a cooling effect of the foil even when the temperature is one hundred degrees

As a result of the open system bars are used to counteract the He-cooling pressure against the water chamber. A closed target will have an outwards bending due to the higher pressure inside and therefore the bars are unnecessary.

Another way to improve the target is to make sure that the impinging energy into the water is in the range that the target is optimized for. For this target it means as high energy as is available ( $E_{\text{max}}=17\text{ MeV}$ ). An increase from 13 MeV to 16 MeV results in an increase in yield of about 12% according to fig 2.3. The foils should be kept as thin as possible to minimize the energy degradation. The threshold value for the vacuum foil thickness is decided from the strain of the He-cooling pressure against the cyclotron vacuum. The threshold value for the target foil will be decided from the pressure inside the chamber. Since the new target will be of a closed type it will result in high pressures inside the chamber. This will result in higher stress on the target foil than for an open system.

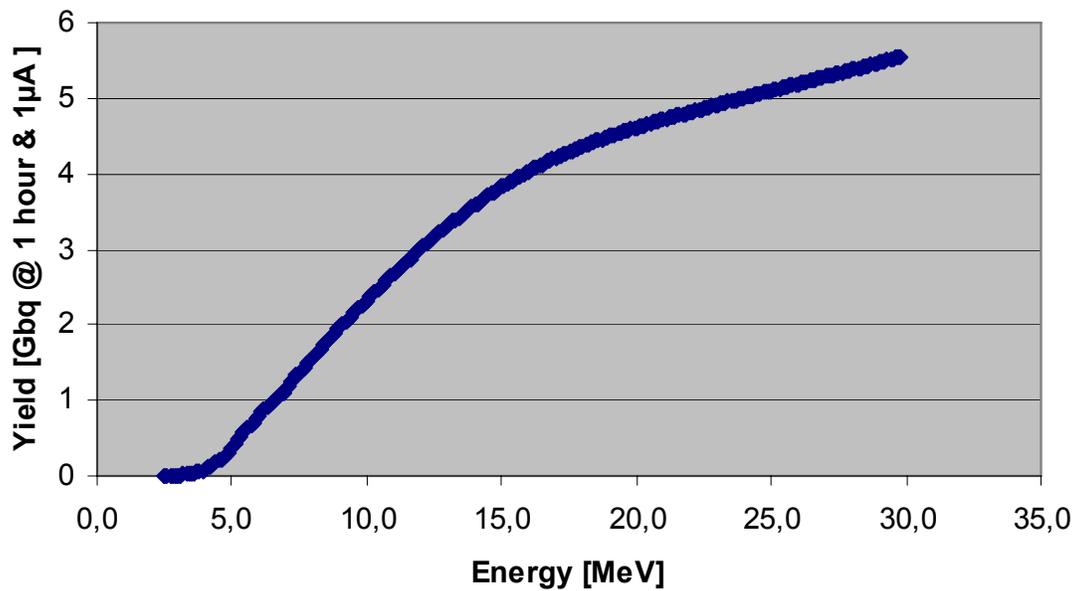


Fig 2.3: Yield<sup>1</sup> as a function of beam energy [10]

The possible improvements for the new target can be summarized as five:

1. New thicker target design that benefits the full beam intensity capacity of a MC 16 Scanditronix cyclotron. The old target can only benefit 22 µA.
2. New target material; more chemical resistance and more beam intensity resistance. The old target produced colloids. High proton beam intensities gave lower yield and discoloration of water
3. Use of thinner foils that give smaller energy degradation. The old foils degraded the beam to 13 MeV
4. Closing the system (no refills)
5. No support bars that steels intensity (approximately 10 %)

All these changes must be carried out with the practical problems under consideration, mainly the construction difficulties. The construction is a give and take problem which must be optimized.

<sup>1</sup> The curve corresponds to a yield of protons on pure <sup>18</sup>O<sub>2</sub> not on H<sub>2</sub><sup>18</sup>O. The yield for water is however almost proportional to the shown curve.

## 2.2 Material Choice

The first problem, and one of the most important to solve, is to find a suitable material for the new target. Meanwhile looking for a new material it is favorable that the new target remains most of the advantages of silver.

There are several of parameters to consider when choosing the material. Material properties to be taken into account are [11, 12]:

- *Thermal conductivity (cooling):* The power input to the target under irradiation is approximately 700 W for a beam intensity of 40  $\mu\text{A}$  (17 MeV  $\cdot$  40  $\mu\text{A}$ ). This power must be dissipated away from the target. A material with high heat conductivity can more easily transport the heat from the chamber through the targets wall out to surrounding cooling. The surrounding cooling is provided letting water streaming around the target body's back.
- *Induction of radioisotopes in the target chamber:* After an irradiation the target can be activated more or less depending of how much the beam will miss the water and hit the target material. It is often necessary to perform adjustments and maintenance on or near the target especially when developing a new target. To avoid unnecessary radiation it is of high priority to choose a material that is not easily activated or where the induced activity decreases fast
- *Chemical resistance and contaminants in the [ $^{18}\text{F}$ ]fluoride produced:* A material with chemically inertness is preferred otherwise the target will add ions to the water that disturb the Synthesis
- *Reactivity of [ $^{18}\text{F}$ ]fluoride produced:* For the synthesis of the FDG it is important that the produced [ $^{18}\text{F}$ ]fluoride is highly reactive i.e. it is in a state to form the 2- [ $^{18}\text{F}$ ]fluoro-2-deoxy-glucose molecule

In addition to these demands the mechanical strength, the ease of work up of the material and the cost/availability has to be consider. It is also an advantage if the material is easily welded.

In a first approach to find a material, focus was on the elements, not alloys, because if the material is composed of different elements it can be hard to estimate all the possible activation. With just one material it is easier to only consider the isotopes of that element in the activation process

The reading of literature resulted in an extra interest of Titanium and especially Niobium as possible target materials. There are articles which describe Titanium and Niobium targets [9,12,13] and its advantages. One benefit with Titanium and Niobium is the good chemical resistance. The draw back of Titanium and Niobium is the poor thermal conductivity and the high activation in comparison with the old target in silver (especially Titanium). The time course of the dose rate was investigated by a group at IBA [12] and is here shown in figure 2.4 and figure 2.5.

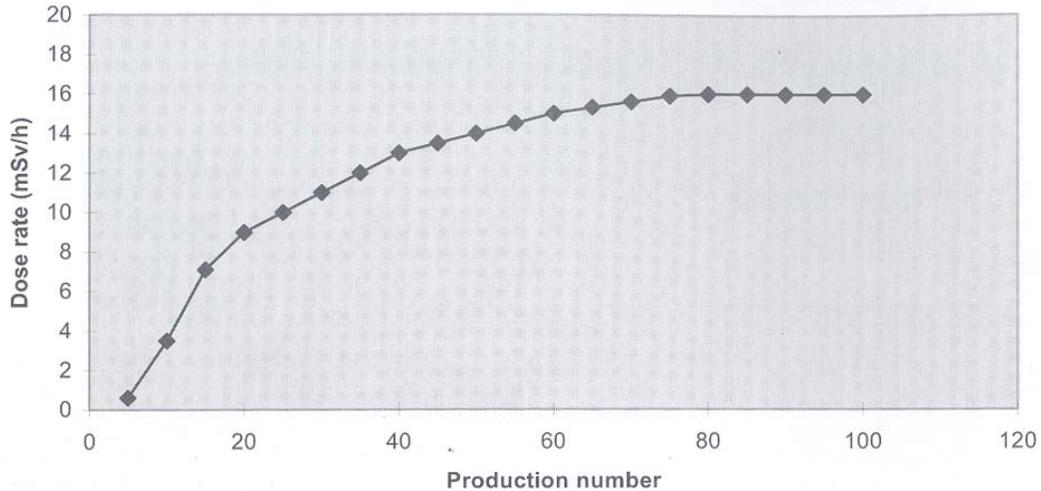


Fig 2.4: Dose rate evolution, 30 cm from Ti target, Cyclone (18 MeV)

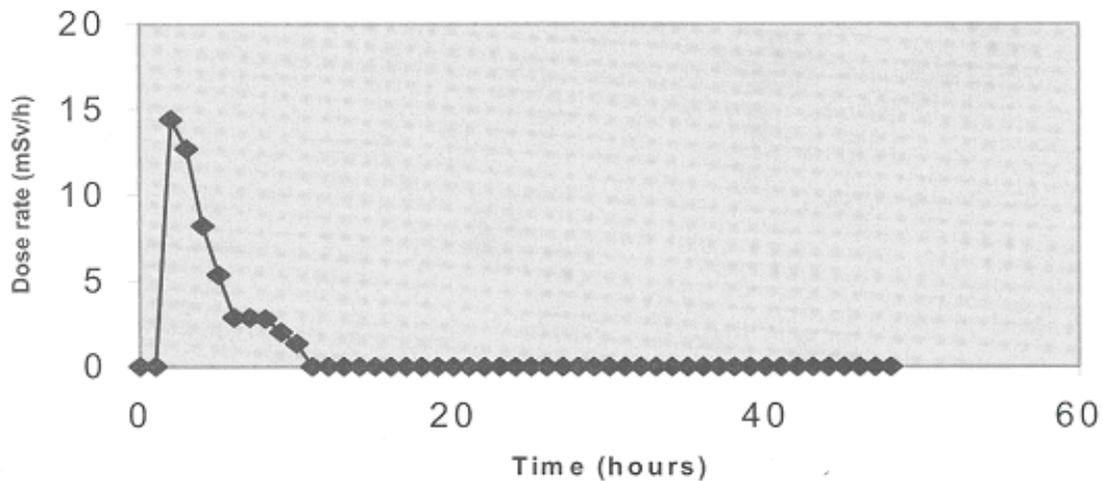
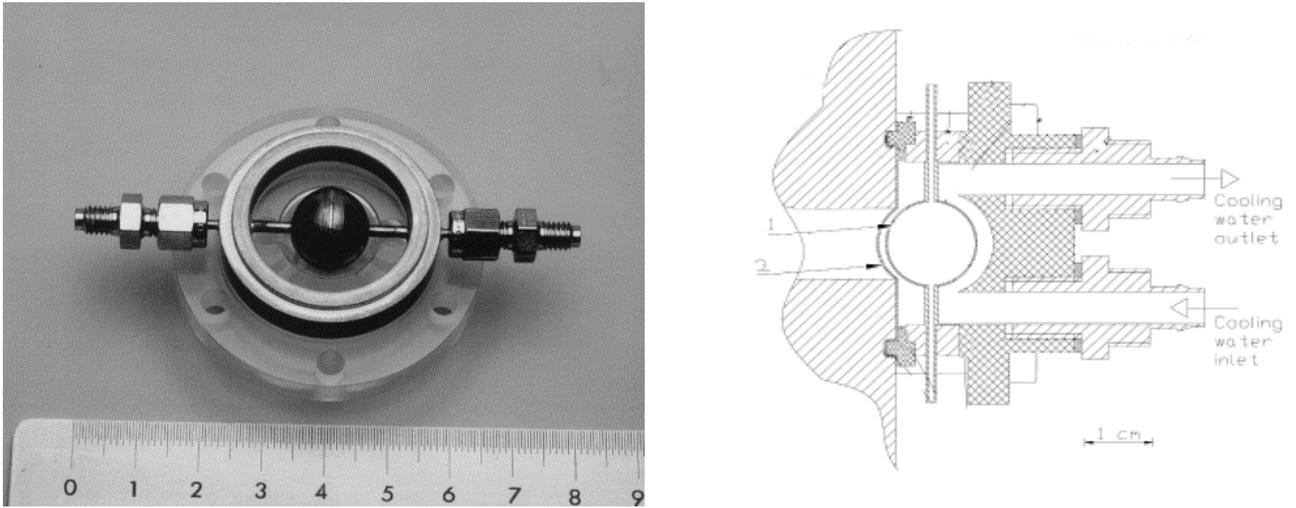


Fig 2.5: Dose rate evolution, 30 cm from Niobium target, Cyclone (18 MeV)

The dose rate is approximately zero 14 hours after an irradiation at the Niobium target. The activity originates from the produced isotope  $^{93m}\text{Mo}$  (6.9 hours). This will decay to a lower energy state of Molybdenum to  $^{93}\text{Mo}$  with a half life of 4000 years. The dose rate evolution curve shows that maintenance at the Niobium target can be tolerable one night after irradiation stop. The induced activity in the Titanium material consists mostly of Vanadin-48 with a half life of approximately 16 days. From a radiation safety point of view the Niobium material is better than the Titanium material.

It has been reported earlier in literature [9,12,13] that Niobium has a great potential as a target material. Niobium targets for irradiation of enriched  $^{18}\text{O}$ -water have already been constructed at other facilities. One of the first was made in Heidelberg, Germany [9]. The construction consisted of a spherical target which was water cooled via water streaming around it. The target routinely produce batches of 56 GBq (1.5 Ci at 30  $\mu\text{A}$ , 1h,  $n=45$ ). This construction is however only functional for those facilities equipped with higher proton energies because of the great energy degradation (i.e. 8 MeV) in the cooling water and the target body's relatively thick wall. The beam penetrates the target's wall (1 in

figure, 0.25 mm) and the water streaming around it (2 in figure, 1.1 mm layer at entrance). The target is shown in figure 2.6.



*Fig 2.6: Spherical target in Niobium made in Heidelberg 2000*

Another target was constructed by a group from IBA in 2002 [12]. The target was fitted into an IBA 10/5 and an IBA 18/9 (10, 18 MeV). The activity produced with the 18 MeV-machine was 160 GBq (4.4 Ci at 35  $\mu$ A, 120 min) and 130 GBq for the 10 MeV-machine (3.5 Ci at 65  $\mu$ A, 120 min).

Since the beam profiles for these cyclotrons are circular the targets are not functional for a MC 16 Scanditronix with a broad beam.

A design more interesting for a MC 16 Scanditronix cyclotron is the one developed for a MC-17 Scanditronix cyclotron (no beam line) [13] at University and University Hospitals of Cleveland 2001. The target produce mostly batches of 78 Gbq (2.1 Ci at 30  $\mu$ A, 75 min). Up to 90 GBq (2.5 Ci) has been produced up today.

These results can be compared with the existing silver target which can produce approximately 32 GBq (0.86 Ci for 1 hour at 22  $\mu$ A beam intensity).

With the reports of Niobium as a good target it was investigated even more. Information of the characteristics [14] of Niobium in comparison with Silver and Titanium is presented in table 2.1.

Properties	Melting point [°C]	Thermal Conductivity at 0-100 °C [wm <sup>-1</sup> k <sup>-1</sup> ]	Density [g/cm <sup>3</sup> ]	Atomic number
Niobium	2468	53.7	8.57	41
Silver	961.9	429	10.5	47
Titanium	1660	21.9	4.5	22
Mechanical properties	Tensile Strength [MPa]	Yield Strength [MPa]	Tensile Modulus [GPa]	
Niobium	585, 330*	550, 240*	104.9	
Silver	330	-	82.7	
Titanium	230-460	140-250	120.2	

Table 2.1 properties from Goodfellow [14], \* Hard Niobium and Soft Niobium

As one can see from table 2.1, the tensile strength and the yield strength of Niobium (585, 550 MPa respectively) is higher than those for Silver and Titanium. The “yield strength” gives the value of the tension (MPa) where the metal can not relax to its initial shape. This region is called the plastic region. The “tensile strength” gives the value when the metal brakes down totally i.e. there is a fracture.

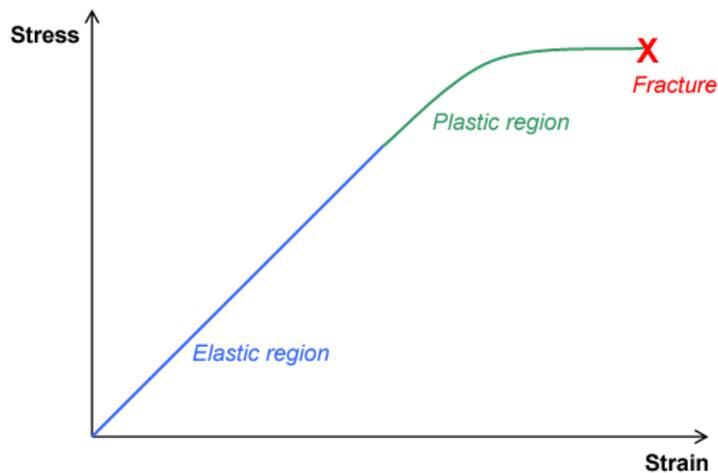


Fig 2.7: Stress strain curve

One drawback with Niobium is its poor heat conductivity which is about eight times smaller than that for silver according to table 2.1. Another disadvantage is that niobium is not easily welded.

Niobium has a very good chemical resistance and therefore finds many applications in the chemical industry. Many tons of niobium has been used in advanced constructions for project in outer space. The capture cross sections for thermal neutrons are low and therefore Niobium can find functions in the nuclear industry. Niobium often appears in compositions with Tantalum and can be found in ores. The name is adopted from the Greece mythology. Niobe was Tantalus daughter.

## 2.3 Dimensioning of the target body

The dimensions of the new target are mostly the same as the old one but with some modifications. The new material, Niobium, does not possess the same good heat conductivity as silver. To compensate for that it is necessary to have a thin wall against the cooling water. The new thickness of the wall will only be 0.5 mm. To roughly estimate the equilibrium temperature inside the chamber a simple two dimensional equation can be used [13]:

$$\Delta t = HT / (Ak) \quad (2.2)$$

where:

$\Delta t$  = the temperature rise in the target chamber over cooling water temperature

H = is the heat load (700 W = 40  $\mu$ A · 17 MeV)

T = thickness of metal wall

A = area of metal in contact with target water

k = thermal conductivity

The water cooling surface corresponds to the machined area (9 cm<sup>2</sup>) and not the beam entrance window (4 cm<sup>2</sup>) since the deposit energy is believed to be well mixed in the entire target water volume due to cavitations and boiling.

With H = 700 W, T = 0.005 m, A = 0.0009 m<sup>2</sup> and K = 53.7 Wm<sup>-1</sup>k<sup>-1</sup> equation 2.2 becomes:

$$\Delta t \approx 73^\circ$$

The result shows that the temperature raise will not boil the target water. The temperature raise of 73° in addition to 20°C (cooling water has room temperature to avoid condensation) is approximately 93°C. But since equation 2.2 does not consider the poor heat conductivity of water it self in a small water volume, the water will however probably boil anyway. Three possible heat transfers from the beam power into the water, from the water out to surroundings are:

1. Pure heat conduction
2. Heat transfer by free convection
3. Heat transfer by boiling

According to earlier studies of water temperatures in targets [8] the two first are not sufficient to transfer the power put into the water of their target system (approximately the same properties for this target) out to surroundings. This is independent of intense cooling. The cooling efficiency of water it self can not provide enough heat conduction to prevent it from boiling through the two first ways. The heat transfer will therefore be performed effectively by water boiling i.e. movement of water molecules. A way to increase the boiling point is to increase the pressure inside the target chamber. A higher pressure results in a higher boiling point as is shown in figure 2.8.

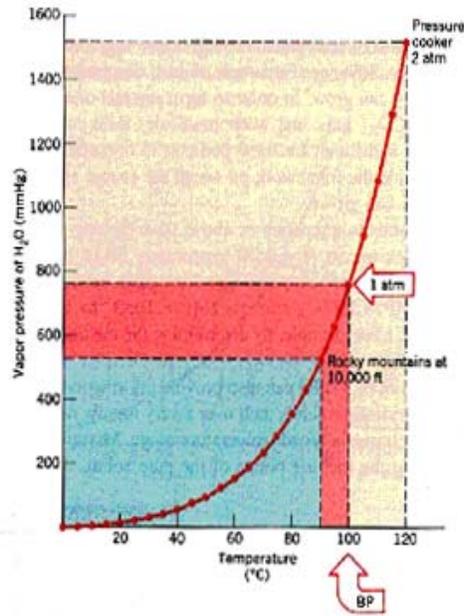


Fig 2.8 Boiling point of water as a function of pressure

With the conclusion that the target water will boil it is necessary to take that into consideration. This is done with an extra head space over the actual water level. When the water starts to boil, water molecules will convert to gases and by doing so they will expand a great factor. If there is no expansion volume, i.e. the headspace, a big pressure will be built up.

A successful target design includes the required thickness of the water layer for optimum beam degradation since the enriched water is expensive. The threshold value for the  $^{18}\text{O}(p,n)^{18}\text{F}$  is 2.5 MeV [15]. The optimum beam degradation i.e. the optimum water (the thinnest and therefore the cheapest) layer would be thick enough to degrade the beam to  $E = 2.5$  MeV at the end of the layer. Protons with a kinetic energy smaller than 2.5 MeV will not produce  $^{18}\text{F}$  so they are of no use. The energy acting upon the water is 15.9 MeV according to appendix A2.1.b. For a beam energy of 15.9 MeV the projected range<sup>2</sup> is approximately 2.8 mm [16]. This thickness is only valid for low beam currents. The actual thickness of the water layer under irradiation becomes smaller than the calculated due to air bubbles and cavitations. But on the other hand the depth will increase due to the outwards bending of the foil from the higher pressure inside the chamber. Even though the theoretical value is not correct it can however give a first estimation of the correct thickness.

To compensate for bubbles, cavitations and increased beam intensity the depth of the new target will be increased by a factor of approximately two compared to the old target (2.5 mm). The new depth will be 5.5 mm plus a silver ring (tightening) of 0.4 mm. During the irradiation the foil will bend outwards giving some “extra” depth to the water layer. The outwards bending is calculated in appendix A2.2. In Berridge’s work[13], the

<sup>2</sup> The projected range gives the depth of particle penetrations meanwhile the true length of particle tracks is described by the CSDA-range (Continuous Slowing Down Approximation).

investigation of different depths showed that a depth of approximately 6 mm was optimal (i.e. it gave same value as deeper targets at same beam intensity (high beam dose)).

## 2.4 Foils

The target will be connected to the cyclotron through a double foil system. The foils are supposed to be in Niobium or Havar material. The Niobium material has a high chemical resistance which will avoid contamination [12] of the target water. The Havar (see table A2.1 in appendix) is favorable because the pressures produced inside the chamber are high. According to earlier works, pressures up to 7 bars at 50  $\mu\text{A}$  have been produced [17] under similar conditions. The tensile strength of Havar foil is about three times stronger than that for Niobium (1860 MPa vs 585 MPa) [14]. When irradiation starts the pressure will rise in the target chamber. This pressure sets the limit on the thickness of the foil right next to the target. The foil system will consist of two foils flexible square window. The dimensions are 40x10 mm. The vacuum foil (see fig 2.1) will be 25 (Havar) or 50 (Nb) microns. Between the vacuum foil and the target foil, i.e. the one which encloses the water target, there will be a jet stream of cooled Helium-gas operating at 1 bar pressure. The foils will be pressed together with a thin wire of silver between the foils and the supporting material.

## 2.5 Construction of the target body

The target body is machined out from a Niobium piece 50 mm diameter and 6 mm thick, figure 2.10b, with help from a specialized stabilize tool, figure 2.10a. The half circle has a diameter of 40 mm. The pieces were ordered from Edstraco AB. The machined volume is approximately 9 cm<sup>2</sup> area time 0.55 cm depth (4.95 cm<sup>3</sup>) and the filling volume was determined to 3.5 ml. The rest of the volume is the head space. The filling volume was determined by closing the target with a transparent plastic plate. The level was adjusted to lie a little bit over the entrance windows edge, figure 2.10c. This match was easily done with the eyes. In the bottom and the top, two holes with 2 mm diameter were drilled for filling and evacuating of the <sup>18</sup>O-water and flushing of the target.



Fig 2.10a: Stabilizer for machining of Niobium piece

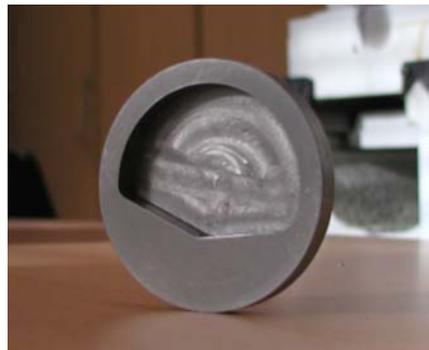


Fig 2.10b: Machined Niobium piece

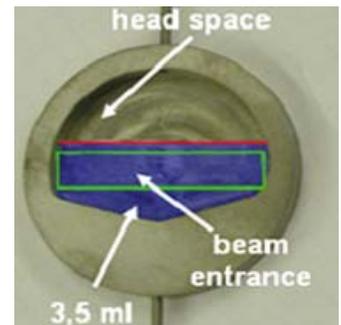
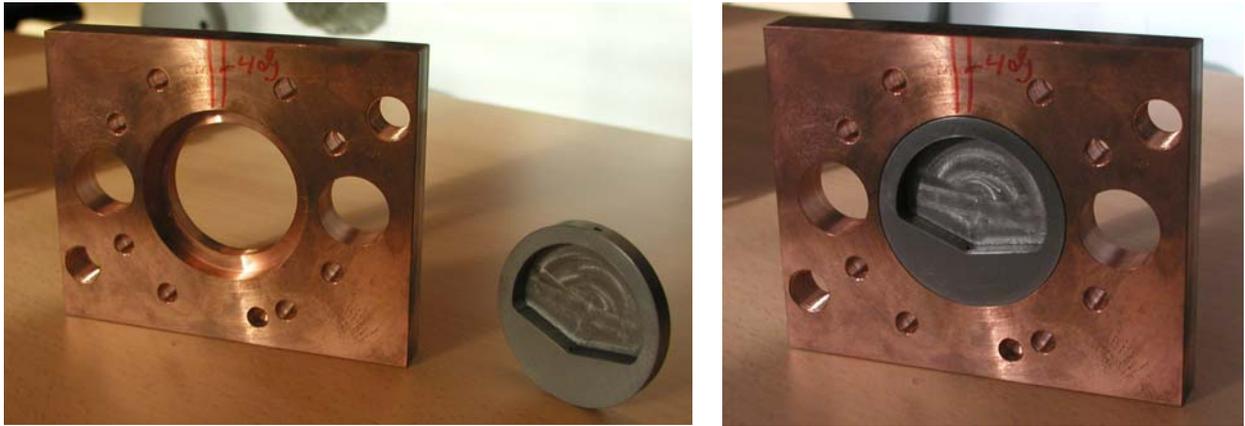


Fig 2.10c: Filling volume and head space

The slope of the bottom at the target chamber is there to avoid remains of the water when evacuating the chamber after an irradiation. To mount the target onto the cyclotron a flange of Copper was constructed, figure 2.11. The Copper flange was designed so that

the old Helium cooling part from Scanditronix could be mounted on it. Mounting holes and screws were also designed to be compatible with the cyclotron's target holder.

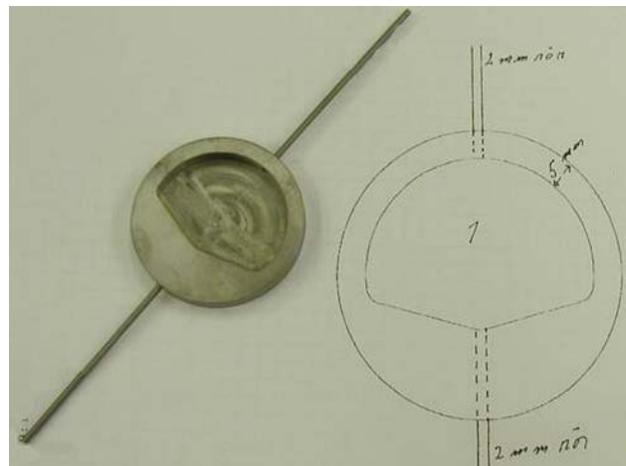


*Fig 2.11: The Copper holder for the Niobium piece*

Two tubes were connected to the filling/evacuation and flushing holes, 1/16" outer diameter and 1 mm inner diameter. In a first approach the tubes were supposed to be connected through electron beam welding. To weld niobium, temperatures over 2468 ° C are needed, so therefore Niobium tubes were used. At the welding facility there were problems in the rig up of the target and tubes. The welding is based on a rotational symmetry that results in a smooth circular weld. When the target was rigged up and rotated it started to flap. This problem resulted in a weak welding seam and that the tubes could not fit tight. Another reason why the electron welding failed was the differences in the amount of material. The target body consists of much more material than the tubes and therefore dissipates more heat. This results in that the tubes reach the melting point before the target body does.



*Fig 2.12a: Threads soldered on the tubes*



*Fig 2.12b: Tubes glued and screwed on the target body*

After the welding problems a new approach to connect the tubes was made. A hole was drilled in a 3 mm screw and then soldered onto a stainless steel tube, figure 2.12a. The holes in the target body were threaded. The connection was sealed with a two

compartment adhesive specialized for high temperatures (up to 250° C) to get a tight system, figure 2.12b.

Before mounting the tubes the target needed to be washed because of a black layer of burned oil, probably from the welding machine. This cleaning was performed by putting the entire target in to aqua regia (nitric acid and hydrochloric acid 1:3 mixture) and letting it boil for about one hour under supervision. The aqua regia totally cleaned the target from the oil. The target itself looked intact thus showing that there is no danger putting it into a bath of aqua regia. After the adhesive was mixed and applied the target and tubes were putted into an 80 ° C oven for approximately 5 hours. No stress was put onto the gluing joints for further a day. This solution worked very well and the construction of the target could proceed.

Next step in the construction process was to engineer an automatic system for closing and opening of the valve to which the tubes should be connected to. The valve system consist of a Reodyne valve with six ports, figure 2.13.

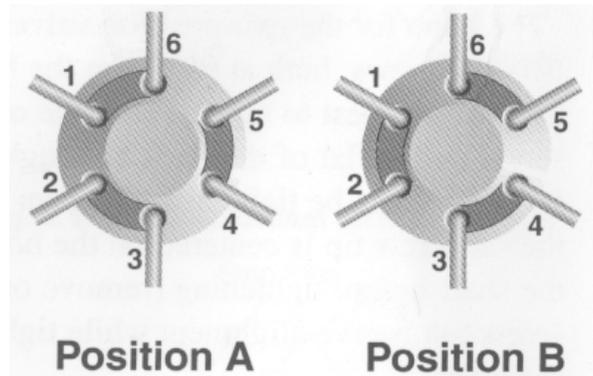


Fig 2.13: Valve system from Reodyne instruction manual

The lower (filling & evacuation) tube was connected to port number 1 and the upper tube (flushing) to port nr 2. This means that in position A (open position) port number 1 and 6 is united and thus letting the water enter the valve via port 6 and fill the target via port 1. By switching the valve to position B (closed position) port number 1 and number 2 gets united. In this situation the target system is isolated, it is a closed target. A pressure gauge will be mounted on the tube between the target and port number 2. This means that when the target is in closed position it is possible to measure the pressure development inside the target chamber during irradiation. The fitting between the stainless steel tube, the pressure gauge and the tube to the Reodyne valve was managed with T-cross from Swagelock. The fitting into the Reodyne valve was carried out with their own fittings. The Reodyne valve also gives the ability to test the fitting of the system by closing port 3 when filling the system with some gas (Helium, Nitrogen) instead of water in port 6. When closing the target after some gas filling it is possible to verify if the system fits tight by studying the pressure gauge. If a start pressure (i.e. the pressure existing after closing) is constant (at least not a fast decrease) it tells us that the system fits tight. The open position contra the closed position is changed by switching a spindle 90°. This spindle is connected to an electric engine that was mounted behind the Reodyne valve.

An engine with good radiation tolerance was used. The connection was achieved by gear-wheels on the motor and Reodyne spindles united by a flexible plastic toothed gearing. The opening and closing of the valve, i.e. turning the spindle 60° in opposite ways, was performed by switching the polarity of an electric engine. The exact location for the closed or open position of the valve is determined by two micro switches that cut the current in the circuit when activated. The micro switches are activated by a stick touching them when the spindle rotates. The position of the micro switches was calibrated to the correct place through changing their locations in a milled track that corresponds to activation from the stick when the stick is in open or closed position. All the parts were mounted onto a frame of aluminum, figure 2.14.

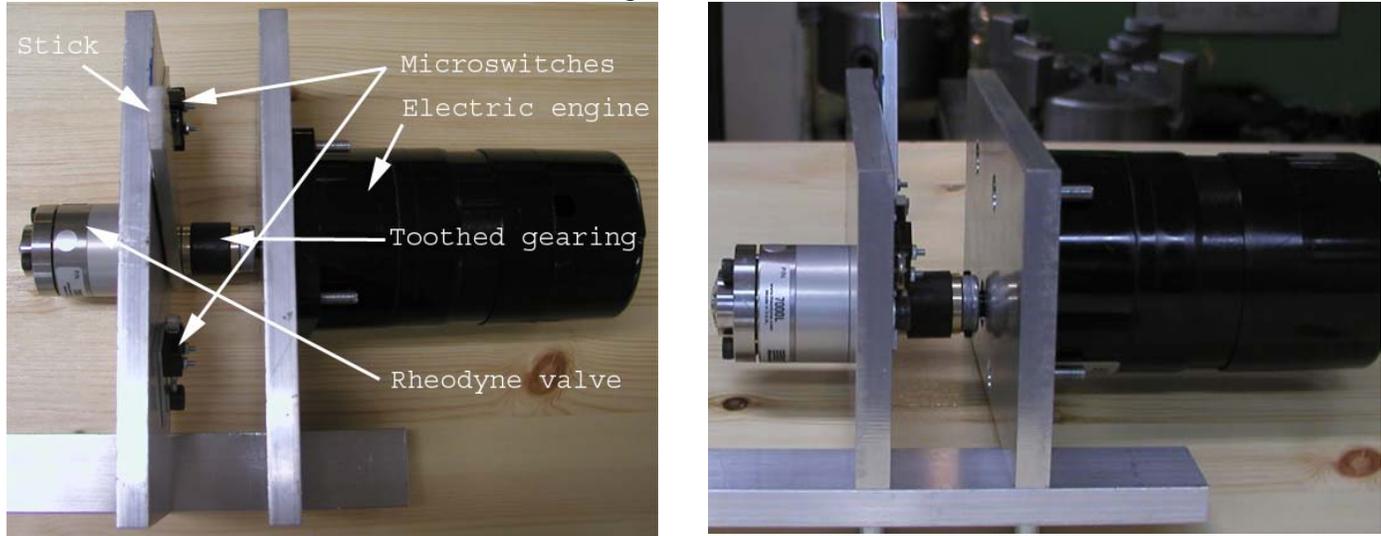


Fig 2.14: To left is the automated valve system viewed from above, to right it can be seen from the side. The frame is all made in Aluminum and screwed together with Allen screws. The stick that activates the micro switches is connected to the spindle and follows its rotations.

The house for the cooling water at the back of the copper plate (see figure 2.11) was connected through a solder seam with solder tin. The target body (Niobium piece) was then pushed in its position. At the joint between the target body and the copper a wire of Indium (1mm) was used as tightening material. The Niobium piece was pressured against the copper with a screw clamp. Because of the characteristics of Indium (very soft material) it floated out and tightened the joint. To seal the connection between the copper piece and the already existing Scanditronix helium cooling flange a thin wire (0.5 mm) of silver was used. The silver ring or the profile of the silver wire was manufactured simple by matching it by hands against the niobium piece. It was designed to fit in the middle of the niobium edge. The ends were welded together with a simple burner with Butane gas. The Havar foil was fitted to the Helium flange from Scanditronix and connected with a simple glue (conformal coating). The Helium cooling flange and the Niobium target with connected copper piece was screwed together with eight 5 mm Allen screws. At the other side of the Helium cooling flange a 25 micron Havar foil was already fitted and pushed together with the same principle. The target was now ready for tests.

The first test was to control if the cooling water system fitted tight. This was first done before the mounting of the copper piece and the Helium cooling part. That test was easily

done by connecting the water cooling house to the water system (6 bar). The test showed no tendency to leakage. After the test the rest of the parts were put together and this resulted in a leakage of approximately 1 deciliter per hour. The reason for the leakage was probably because of the stress applied on the copper flange when connecting it with the Allen screws against the helium flange. This stress probably cracked the solder seam of tin. The leakage was not that big and could temporarily be repaired with a two compartment adhesive. After two test runs the leakage increased and therefore the target was mounted off the cyclotron for better reparation. To tighten the solder, super steel from plastic padding was used. The old glue was removed with a steel brush. The new reparation succeeded and no leaking occurred.

The other test was to control if the target system was tight by filling it with gas and study the pressure. This was a critical test since there was no hundred percentage wisdom if the connection with the stainless steel tubes at the target body fitted tight. If they did not, a totally new approach had to be developed to attach these tubes. It was also a critical test because it was unknown if the silver ring could tight the joint between the foil and the target body.

The pressure gauge had an operating range of 1-5 volts which was equivalent to 200 psi. Therefore every volt corresponds to 50 psi (~ 3 bars).

When the system first was filled with Nitrogen gas to a pressure of 3 bars overpressure a big leakage resulted in a fast decrease when the target was closed. When it is closed it has ten possible joints for leakage: one around the periphery of the target foil, two at the target (adhesive and threads), five at the pressure gauge (four Swage lock, one with conical threads) and two at the Reodyne valve (Reodyne connection). The first step was to pull all screws a little bit tighter. Then the Nitrogen gas was switched to Helium gas since a tool to detect Helium gas was available. The system was again filled to 3 bars and the system was much tighter but a big leakage could be identified at the connection between the pressure gauge and the NPT connection that converted it to a 1/16 inch Swage lock fitting. This problem was solved by adding some sealing tape used by plumbers. The following test (again 3 bars) showed promising result and the leakage was ok to neglect. The following test subjected the system with a pressure approximately at 9 bars. At this pressure the leakage had increased a little bit but with a quick calculation the leakage was smaller than 0.1 ml per two hours.

## **2.6 Target operation**

The target is filled with 3.5 ml water through silicon tubes with a peristaltic pump. To verify that the filling/evacuation system worked, water was pumped in and then pumped out. The amount of water was measured before and after. The electronic pressure gauge was connected to an analogue voltmeter that can be visualized via a camera outside the cyclotron vault. The pressure can therefore be followed during the irradiation. Besides knowing the pressure there is another advantage with the meter. It tells the operator if the water has entered the chamber since if it has not, no pressure raise will occur. This meter helps to avoid irradiations with an empty target that sometimes can occur due to leakage or mistakes.

After an irradiation the target is flushed with 50 ml distilled water (20 minutes). The flushing is performed with the filling system. Approximately 5 % of the produced activity remains inside the chamber and is diluted into the flushing water. In the future a fully automated system will be installed in the control computer of the cyclotron. This will make the filling, evacuating and flushing of the target easier and executable outside at the operation desk with a control box.

The flushing is performed because the leftover seems to be aggressive [7]. When [<sup>18</sup>F]fluoride was produced from a tandem generator in Lund, a Pelletron accelerator was used. Since it only gave protons with energy of 6 MeV it was important to use thin foils (5 micron Ni). When the target was not flushed after a few runs, it started to leak. When the foil was dismantled several pin holes were noticed between the chamber and the foil. This rupture of the foil was probably due to galvanic ions produced from different (Silver and Nickel) chemical potentials and/or the aggressive solvents produced in the target chamber.

## 2.7 Irradiation experiments

The experiments for the target consisted of different types of irradiations.

1. *Pressure:* One of the first parameter that was examined was the pressure built up inside the target chamber. An individual pressure study was carried out where the pressure was examined for different beam intensities. To receive an equilibrium pressure every result with adherent beam intensity was noted after an irradiation of 5 min. This was performed for irradiations of 10, 20, 30, 40 and 50  $\mu\text{A}$ .
2. *One hour irradiation:* This is an irradiation of major interest since it is a common time of irradiation. The irradiation was performed with a beam intensity of 40  $\mu\text{A}$ . The activity produced from one hour corresponds to approximately 30 % of saturation activity. The irradiation was performed by using 10 % enriched  $^{18}\text{O}$ -water (95 %) mixed with distilled water. At end of bombardment (EOB) the activity was measured after one half life of  $^{18}\text{F}$ . This was done to get rid of the  $^{13}\text{N}$  originating from the activation of distilled water. A  $^{18}\text{F}$  half life corresponds to 109.8 min meanwhile 109.8 min correspond to approximately 11 half lives of  $^{13}\text{N}$ . Thus waiting one  $^{18}\text{F}$  half life gives less than 1/2000 of the  $^{13}\text{N}$  remains. The value was then extrapolated considering mixtures and decay of the  $^{18}\text{F}$ .
3. *Low intensity irradiation:* This test was performed to decide the saturation activity per  $\mu\text{A}$ . By decreasing the intensity the cavitations and boiling of the water also decreases thus giving a more correct value for the saturated activity. This is a base line test performed to verify how good the target can handle high beam intensities. This verification is done by comparing the values with each other.
4. *Two hour irradiation with FDG-synthesis:* This irradiation was a full scale experiment that tested the capacity of the target. 95 % enriched water was used. The production was pumped into the synthesis box to decide the reactivity of the produced [ $^{18}\text{F}$ ]fluoride for the synthesis of FDG.

### 3. Results

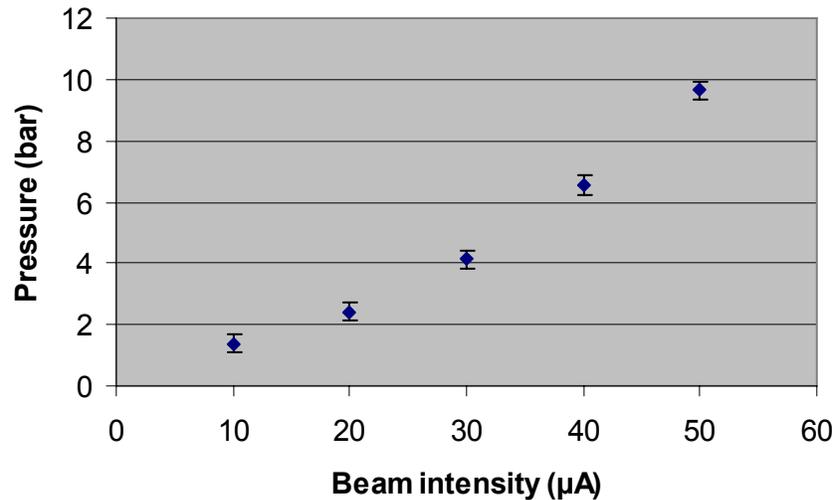
#### 3.1 Foil tolerances

Material	Thickness ( $\mu\text{m}$ )	Tolerance (Bar)
Nb	100	6
Nb	150	9
Havar	25	6.5
Havar	50	13

Table 3.1: Calculations are made for flexible material rectangular windows, held not fixed in the Jeffrey I. Western guideline [19]

#### 3.1 Pressure test

Target pressure as function of beam current



The errors are estimated to be 10 % of noted voltage value and this corresponds to a factor of 0.3 bar

#### 3.3 One hour irradiation

1 hour, Beam current = 40  $\mu\text{A}$  (n=2)

1	Produced Activity	A_sat/ $\mu\text{A}$
GBq	93.00	6.72
Curie	2.51	0.18

2	Produced Activity	A_sat/ $\mu\text{A}$
GBq	92.00	6.86
Curie	2.49	0.19

No discoloration of water.

### **3.4 Low intensity irradiation**

30 min-Low intensity (n=1), Beam current = 10  $\mu$ A

<b>1</b>	<b>Produced Activity</b>	<b>A_sat//<math>\mu</math>A</b>
<b>GBq</b>	12.00	6.93
<b>Curie</b>	0.32	0.19

No discoloration of water.

### **3.5 Two hour irradiation with FDG synthesis**

2 hour, Beam current =40  $\mu$ A, (n=3)

<b>1</b>	<b>Produced Activity</b>	<b>A_sat//<math>\mu</math>A</b>	<b>Amount FDG</b>	<b>Yield</b>
<b>GBq</b>	64.00	Se discussion	39	61 %
<b>Curie</b>	1.72	-	1.10	-

<b>2</b>	<b>Produced Activity</b>	<b>A_sat//<math>\mu</math>A</b>	<b>Amount FDG</b>	<b>Yield</b>
<b>GBq</b>	138.00	6.57	80	58 %
<b>Curie</b>	3.73	0.18	2.16	-

<b>3</b>	<b>Produced Activity</b>	<b>A_sat//<math>\mu</math>A</b>	<b>Amount FDG</b>	<b>Yield</b>
<b>GBq</b>	142.00	6.65	87	62 %
<b>Curie</b>	3.81	0.18	2.14	-

No discoloration of water.

## **4. Discussion**

### **4.1 Material Choice**

The material choice, Niobium is good because the fact that it fulfils most of the demands set on the target. The induced activity diminishes fast (approximately 14 hours). The mechanical properties are good i.e. they allow high tension on the material. The melting point is high (2468 °C). The only drawbacks with Niobium are the poor heat conductivity and the welding problem. Those issues were solved with threads and glue and compensating of back plate material. Titanium was not chosen mostly because the long half life of the activation products. It can also be mentioned that the mechanical strength

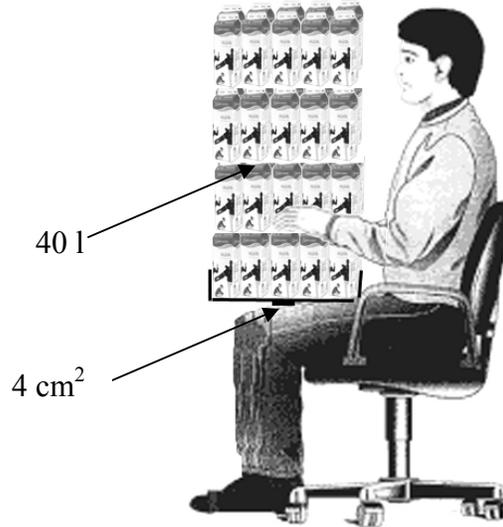
of Titanium are not as good as those for Niobium. Comparisons between Silver, Niobium and Titanium can be visualized schematically in table 4.1 below:

Material	Mechanical Properties	Heat conduction	Chemical resistance	Activation
Silver	3	1	3	1.2
Niobium	1	2.3	1.2	1.2
Titanium	2	2.3	1.2	3

Table 4.1: 1=good, 2=ok, 3=bad, 1,2 & 2,3=Hard to differ them apart

## 4.2 Foils and pressure

The test showed a pressure of approximately 10 bars at 50  $\mu\text{A}$ . It is interesting to notice that a 10 bar pressure corresponds to a water pillar of 100 m on an area of four times one centimeter. This is equivalent of 40 liters of milk bottles putting pressure on the same area.



The intensity that will mostly be used, 40  $\mu\text{A}$ , gives a pressure of around 6.5 bars. For that intensity a 150 micron Niobium foil would have been sufficient but since the target is constructed to benefit the full beam intensity capacity (50  $\mu\text{A} \approx 10$  bar) the Havar foil (13 bar) seems better. It is also unclear whether the Niobium foil has soft or hard mechanical properties and therefore even a better reason to use a Havar foil.

If regular maintenance of the target shows to be unnecessary, the Niobium foils with low activation, is even more unfavorable due to higher energy degradation (comparing to Havar). The drawback with Havar is the induced activity in the foil which can be high when maintenance is performed at the target. If one operates fast, approximately some minutes, when removing the Havar foil the equivalent doses have been estimated to 0.030 mSv whole body and 0.21 mSv for the skin [18].

### 4.3 One hour irradiation

The results for the first irradiation were extrapolated (10 % H<sub>2</sub><sup>18</sup>O) and corrected for decay. The results are high, 93 respectively 92 GBq, and with a coherent normal FDG-synthesis, a production of approximately 55 GBq can be synthesized.

### 4.4 Low intensity irradiation

This irradiation was performed to investigate how much the beam penetrates or misses the water when the intensity increases. This is carried out by comparing the A<sub>sat</sub> values per micro amperes for the low intensity irradiation with high intensity irradiations. In Theory the value (GBq/μA) for the low intensity should be higher since a beam with higher intensity penetrates more water due to greater cavitations and boiling. With a low intensity irradiation of approximately 10 μA the maximum saturated activity can be decided since the water is believed to degrade most of the incoming particles i.e. they will all produce <sup>18</sup>F.

The mean beam intensity is calculated from the accumulated charge and exact irradiation time since there is a variation in the beam intensity during an irradiation.

When comparing the values from table 4.2 it can be seen that they are all almost the same.

Production		A <sub>sat</sub> (95 % H <sub>2</sub> O)GBq/μA	Corrected values*
1	1 hour at ~40 μA	6.72	7.43
2	1 hour at ~40 μA	6.86	7.58
Low intensity 3	30 min at ~10 μA	6.93	7.66
5	2 hour at ~40 μA	6.57	7.26
6	2 hour at ~40 μA	6.65	7.36
Theoretical value at 14 MeV	T.J Ruth & P. Wolf [15]	-	7.99
Theoretical value at 16 MeV	S. Takács [10]		12.80
Theoretical value at 16 MeV	Water target E.Hess [19]		10.31

Table 4.2: \*To compare the values with the literature they have to be corrected for leftovers in the target (~5 %) and 95 %- enriched water (5 %). The correct value to compare is the one presented by Hess et. Al since it is calculated for enriched water(17 % lower) instead of pure oxygen-18-gas

The results are approximately two units below the values presented by Hess et. Al. The low intensity value is the highest test value as expected. Since the high intensity values are almost the same as the low intensity value it tells us that the depth of the target is extremely well decided. The range for 15.9 MeV's protons in water is 2.8 mm [16]. The total depth of the water layer under irradiation has a maximum of 5.5 mm (target chamber) plus 0.4 mm (silver ring) and 1.2 mm (deflection from pressure) ~ 7 mm. The 1.2 mm from deflection is the maximum elongation so it can be hard to tell the average depth. The target is 6-7 mm deep which approximately double the theoretical value for the range is and that is sufficient to manage beam intensities of 40 μA.

#### **4.5 Two hour irradiation with FDG synthesis**

The first result of the two hour tests was not satisfying since the production (64 GBq) only corresponded to approximately half the expected value. The chemist noticed that only about half of the water volume had entered the FDG-synthesis box and therefore the low value could be explained. It is still today unclear what happened to the rest of the water volume but a valid explanation can be that volume was blown away through the flushing exit. It was noticed that a high pressure remained inside the chamber after the machine was shut off. When the valve was opened the pressure could maybe have blown the water volume away in the wrong direction. To prevent this from happening in the future, a bottle is now connected to the end of the flushing tube. The next test showed a more pleasing result (138 GBq) and the good trend was followed up with the result of the third test (142 GBq). The results are very satisfying. No water leftovers were seen. The first setback was hopefully only one time accident. The reactivity of the produced [<sup>18</sup>F]fluoride was tested and showed to be normal. A dip in the yield is seen for the second run but it is not too low to be regarded as abnormal.

#### **4.6 General conclusions**

The results from this Niobium target are very good. It is very satisfying to know that for a one hour irradiation it is possible to produce around 90 GBq with an appurtenant FDG production of approximately 55 GBq. This is roughly a 300 % improvement comparing to the old target (~30 GBq). The biggest gain is due to the beam intensity increase. The target can benefit the full beam intensity potential of the cyclotron thanks to the increase of the water layer depth and the chemical inertness of Niobium. There is also an increase due to higher energy into the water and the removal of bars that steals intensity. The target is capable to produce even larger amounts of FDG, approximately 87 GBq (142 GBq into the FDG-box) for a two hour irradiation.

To the best of our knowledge the results put this target in a leading position for MC 16 and MC 17 cyclotrons. The chemical inertness of Niobium material has been verified. There was no discoloration of water from any irradiation and the reactivity was normal at the FDG synthesis. The capacity of the target will provide the facility with good margins when the demands will increase.

Since this target has worked as a prototype next generation will have some improvements. One major enhancement is to try to connect the filling and flushing tubes via PEEK connections and to use silicon or plastic tubes. These tubes should be compatible with Scanditronix original filling and evacuating system since other facilities has shown interest in this target (Uppsala, Groeningen, Toronto and Taiwan). Another improvement is to use a smaller electric engine since the capacity of the used is unnecessary big. A new mechanical pressure gauge should be installed since the electronic one suffers major damage from radiation. It is also better if the holding flange could be machined from one piece, preferably aluminum, because then the risk of cooling water leakage decreases.

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# Appendix 1

## A1.1. Production of radioactivity

### A1.1.a. Thin layer

To calculate the number  $R$  of nuclear reactions one needs to know the reaction probability between the impinging particle and the target. This probability is expressed in terms of a cross section,  $\sigma$ , which is an effective area that the incoming particle can see i.e. react with. The cross section is expressed in barns or milibarns.

$$1 \text{ barn} = 10^{-24} \text{ cm}^2 = 10^3 \text{ milibarns (mb)}$$

The cross section varies for different nuclei and different energies. It often shows isolated maxima (resonances). The excitation function,  $\sigma(E)$ , for the  $^{18}\text{O}(p,n)^{18}\text{F}$  reaction [15] is shown in figure A1.1 where there is an isolated maxima around 5.5 MeV.

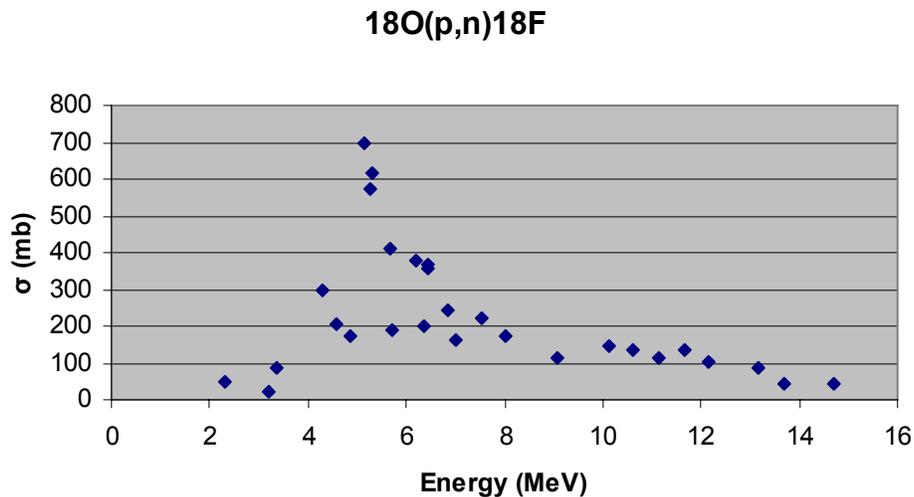


Fig A1.1: Absolute cross sections for the production of  $^{18}\text{F}$  via the  $^{18}\text{O}(p,n)^{18}\text{F}$  reaction

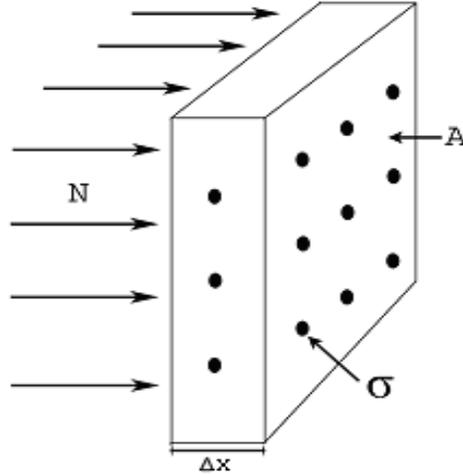


Fig A1:2 activation of target material

If the target sample contains  $n$  target nuclei per unit volume, then will fig A1:2 above contain  $A \cdot \Delta x \cdot n$  target nuclei ( $A$ =area  $\Delta x$ =slice width). The total cross section is the number of target nuclei times their cross sections i.e.  $A \cdot \Delta x \cdot n \cdot \sigma$ . The reaction probability for an incoming particle and a target nucleus is then described by the quota between the total cross section area and the total area<sup>3</sup>:

$$p = \frac{A \cdot dx \cdot n \cdot \sigma}{A} = \Delta x \cdot n \cdot \sigma \quad (\text{A1.1})$$

The rate,  $R$  ( $s^{-1}$ ), at which we will have nuclear reaction per second per unit volume ( $cm^3$ ) of a target with thickness  $\Delta x$  (cm) can be expressed as equation A1.1:

$$R = p \cdot N_p = n \cdot \sigma \cdot \Delta x \cdot N_p \quad (\text{A1.2})$$

where:

$$N_p (s^{-1}) = \left( \frac{1}{Z} \right) \cdot I = \text{particle current (elementary charges } s^{-1}) \quad Z = \text{charge}$$

$\sigma (cm^2)$  = probability for one incident particle to react with a single target nucleus

$$\text{with } n = \left( \frac{\rho}{M} \right) \cdot N_L \quad (\text{A1.3})$$

where:

$\rho (g/cm^3)$  = density of target material

$M (g)$  = molecular weight, mass number in grams

$N_L$  = Avogadro's number ( $6,023 \cdot 10^{23} \text{ atoms/mole}$ )

<sup>3</sup> This is valid under the assumption that  $\Delta x$  is so thin that, seen from the incoming particles, no overlapping of the individual cross sections occurs.

equation A1.1 becomes:

$$R = N_p \cdot \left( \frac{\rho}{M} \right) \cdot N_L \cdot \sigma \cdot \Delta x \quad (\text{A1.4})$$

if one prefers numbers per gram just divide with  $\rho$  :

$$R = N_p \cdot \frac{N_L}{M} \cdot \sigma \cdot \Delta x = N_p \cdot N_T \cdot \sigma \cdot \Delta x \quad (\text{A1.5})$$

where:

$$N_T = \frac{N_L}{M} = \text{number of target atoms per gram} \quad (\text{A1.6})$$

Meanwhile we are having the reactions the number of the target nuclei,  $n$ , will decrease since they are converted to a new nucleus. This decrease is however of such a small amount that it is safe to regard  $n$  as constant. A typical flux in a cyclotron (40  $\mu\text{A}$ ) might be of the order  $10^{14} / (s \cdot \text{cm}^2)$  according to a simple calculation:

1 A corresponds to 1 C and 1 C is the same as  $\frac{1\text{C}}{1.602 \cdot 10^{-19}}$  elementary charges, i.e.

protons. The number of protons per second is easy to calculate when the beam intensity is known:

$$n = \frac{40 \cdot 10^{-6}}{1.602 \cdot 10^{-19}} \approx 2,5 \cdot 10^{14} / s$$

The typical cross sections are of the order  $10^{-24} \text{cm}^2$ . From this it follows that the probability for a reaction to occur is  $10^{-10} / s$  per particle. This means that say after an hour of production the number of converted particles are small ( $\propto 10^{-7}$  of the original number).

If  $N$  is the formed radioactive product, then one can describe its change with time through following differential equation:

$$dN = Rdt - \lambda Ndt \quad (\text{A1.7})$$

Where the increase,  $Rdt$  is due to the production at rate  $R$  and the decrease,  $\lambda Ndt$  is due to radioactive decay.

The differential equation is easily solved:

$$N = \frac{R}{\lambda} \cdot (1 - e^{-\lambda t}) \quad (\text{A1.8})$$

And the activity is given as:

$$A = \lambda \cdot N = R \cdot (1 - e^{-\lambda \cdot t}) \quad (\text{A1.9})$$

$$\text{or with } R = N_p \cdot \frac{N_L}{M} \cdot \sigma \cdot \Delta x \quad (\text{A1.10})$$

$$A = N_p \cdot \frac{N_L}{M} \cdot \sigma \cdot \Delta x \cdot (1 - e^{-\lambda \cdot t}) \quad \text{Activation formula} \quad (\text{A1.11})$$


---

If we have a short irradiation time compared with one half-life, equation 1.8 can be Taylor expanded. Only keeping the linear term (a good approximation) thus gives us:

$$A \approx R \cdot \lambda \cdot t \quad t \ll t_{1/2} \quad (\text{eq.A1.11}) \quad \text{this is the part with a constant production rate.}$$

For long time compared with one half-life the exponential term will become zero. This means that equation (?) becomes:

$$A \approx R \quad t \gg t_{1/2} \quad (\text{A1.12})$$

This is known as the saturation activity,  $A_s$

$$A_s = R = N_p \cdot \frac{N_L}{M} \cdot \sigma \cdot \Delta x \quad (\text{A1.13})$$

If the irradiation is stopped at  $t_s$  the activity for  $t > t_s$  can be described as:

$$A = N_p \cdot \frac{N_L}{M} \cdot \sigma \cdot \Delta x \cdot (1 - e^{-\lambda \cdot t_s}) \cdot e^{-\lambda \cdot (t - t_s)} \quad (\text{A1.14})$$

### A1.1.b. Thick layer

These equations are only valid for a thin layer. For a thick layer one has to calculate the summation of many of these thin layers. For one single thin layer the fluens rate and energy on impinging particle is approximately constant (if not it should be regarded as a thick target) meanwhile for a thick layer (several of thin layers) the fluens rate and energies varies with the depth of the target sample. With different energies follows variable cross-sections, se excitation function. The previous model is so thin that seen from the incident particles no change of the cross section occurs.

If we start with the activation formula eq.A1.11:

$$A = N_p \cdot \frac{N_L}{M} \cdot \sigma \cdot \Delta x \cdot (1 - e^{-\lambda \cdot t})$$

where:

$$\Delta x = \frac{\Delta E}{(dE/dx)}$$

$$\lambda = \frac{\ln 2}{T_{1/2}}$$

To calculate the entire produced activity one has to integrate over the whole energy range;  $E_s \leq E \leq E_{\max}$  where  $E_s$  = starting energy, threshold, for the reaction and  $E_{\max}$  is the maximum energy.

$$A = N_p \cdot \frac{N_L}{M} \cdot (1 - e^{-\lambda \cdot t}) \int_{E_s}^{E_{\max}} \frac{\sigma(E)}{(dE/dx)} dE \quad (\text{A1.15})$$

when  $t \rightarrow \infty$  ( $t \gg t_{1/2}$ ) we will again get the saturation activity:

$$A_s = N_p \cdot \frac{N_L}{M} \cdot \int_{E_s}^{E_{\max}} \frac{\sigma(E)}{(dE/dx)} dE \quad (\text{A1.16})$$

Next step is to develop the formula for the Yield. This is measured in  $\mu\text{Ci}/\mu\text{A}$  or  $\text{GBq}/\mu\text{A}$ .  $A/I=Y$  ( $\mu\text{Ci}/\mu\text{A}$  or  $\text{GBq}/\mu\text{A}$ ) is the Thick Target Yield

For:

$$A_s = 1 \mu\text{Ci} = 3.7 \cdot 10^4 \text{ s}^{-1}$$

$$N_p = 1 \text{ elementary charge } e / \text{s} = 1.602 \cdot 10^{-27} \mu\text{A}$$

$$\sigma = 1 \text{ milibarn (1mb)} = 10^{-27} \text{ cm}^2$$

we can calculate:

$$Y = A/I = \frac{\overbrace{1}^{N_p} \cdot I \cdot \frac{N_L}{M} \cdot \int_{E_s}^{E_{\max}} \frac{\sigma(E)}{(dE/dx)} dE}{I} =$$

$$\frac{1}{Z \cdot M} \cdot \frac{6.023 \cdot 10^{23}}{3.7 \cdot 10^4} \cdot \frac{10^{-27}}{1.602 \cdot 10^{-13}} \cdot \int_{E_s}^{E_{\max}} \frac{\sigma(E)}{(dE/dx)} dE =$$

$$1.016 \cdot 10^5 \cdot \frac{1}{Z \cdot M} \cdot \int_{E_s}^{E_{\max}} \frac{\sigma(E)}{(dE/dx)} dE \left( \frac{\mu\text{Ci}}{\mu\text{A}} \right) \quad (\text{A1.17})$$

## A1.2. Nomenclature

The Standard unit of activity is the Becquerel (Bq). The Bq is equal to one decay per second. Since a production is in the region of several GBq, it can sometimes be convenient to use the old unit Curie (1Ci = 37GBq). When comparing results from different facilities the produced activity can be presented as the amount of activity, in Bq and/or Ci, with an appurtenant time for the production.

With the time known it is easy to calculate the saturation activity. The saturation activity corresponds to a production run for an eternal time (or a time  $t \gg t_{1/2}$ ). The saturation activity is presented as  $A_{sat}$  in GBq or Ci with belonging beam current that was used under production ( $\mu A$ ) or as saturation activity divided with adherent beam intensity (Gbq/ $\mu A$  or Ci/ $\mu A$ ).

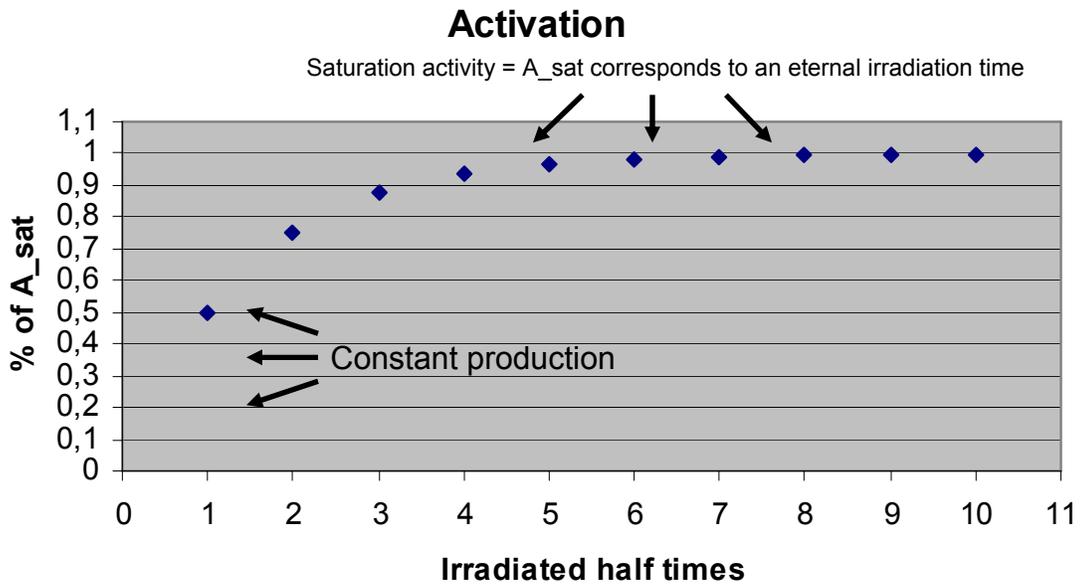


Fig A1.3: Percentage of saturation activity as a function of number of irradiated half lives

A closer look at the activation formula, which is sketched in figure A1.3 shows that an irradiation for one half life gives us 50 % of the saturation activity. An irradiation of two half lives results in 75 % of the saturation activity. Another half lives added will result in an 87.5 % of saturation activity. In our FDG-production we only run the cyclotron for one to two hour. This corresponds to approximately one half half-life to one half-life of the [ $^{18}F$ ]fluoride ( $t_{1/2}=109.8$  min).

If the cyclotron has produced 1 Ci while working one hour which corresponds to approximately 30 % of saturation activity. The saturation activity can then be calculated by simply extrapolating the 1 Ci to a corresponding 100 % equivalent value which is 3.3Ci.

For an isotope like [ $^{18}F$ ]fluoride it is too time consuming to irradiate longer than 0.5-1 half life because the actually half life of [ $^{18}F$ ]fluoride is 109.8 min. The gain is not worth

the effort in time. For isotopes with a shorter half life say,  $^{15}\text{O}$  ( $t_{1/2}= 2.04$  min) or  $^{13}\text{N}$  ( $t_{1/2}=9.96$  min) an irradiation of more half lives is reasonable.

### ***A1.3. Detailed table of results***

<b>Date</b>	<b>Integrated time</b>	<b>Integrated time (min)</b>	<b>% of A_sat</b>	<b>A_sat</b>	<b><math>\mu\text{Ah}</math></b>	<b><math>\mu\text{A}</math></b>	<b>A (GBq)</b>	<b>FDG</b>	<b>Yield FDG</b>	<b>A_sat GBq/<math>\mu\text{A}</math></b>	<b>% <math>\text{H}_2^{18}\text{O}</math></b>
<b>2005-12-08</b>	01:01:00	61	0.32	290.63	44	43.28	93	-	-	6.72	10
<b>2005-12-08</b>	01:01:21	61.4	0.32	286.32	42.7	41.73	92	-	-	6.86	10
<b>2006-01-10</b>	00:28:53	28.9	0.17	71.96	5	10.38	12	-	-	6.93	10
<b>2006-01-11*</b>	-	-	-	-	-	-	64	39	0.61	-	95
<b>2006-02-08</b>	01:58:47	118.8	0.53	261.55	78.8	39.80	138	80	0.58	6.57	95
<b>2006-02-14</b>	01:58:31	118.5	0.53	269.59	80.01	40.51	142	87	0.61	6.65	95

*Fig A1.4 detailed table of results*

\*See discussion 4.5 in thesis

## Appendix 2

### A2.1. Calculations of foil thickness

As discussed earlier one must take the pressure into consideration when deciding the thickness of the target foil. Such kind of calculations can be performed using an already existing guideline which is to be regarded as an established model for designing of foils. The guideline [20] starts to set up some demands on the allowable stress that will act on the foil:

$$S = 0.5F_U \quad (\text{A2.1})$$

$$S = 0.9F_Y \quad (\text{A2.2})$$

Where:

S = allowable stress (psi, pounds per square inch)

F<sub>U</sub> = ultimate tensile strength (psi)

F<sub>Y</sub> = yield strength or stress to produce 5 % elongation (psi)

For a rectangular flexible window, held not fixed, the allowable stress shall exceed:

$$S > K_1(E(qa/t)^2)^{1/3} \quad (\text{A2.3})$$

Where:

t = thickness of window (inch)

K<sub>1</sub> = stress constant based on ratio a/b

a = short side of rectangular window measured at O-ring (inch)

b = long side of rectangular window measured at O-ring (inch)

q = uniform pressure on window (psid)

E = Young's modulus of window material (psi)

Y = window deflection (inch)

It can be tough to estimate the actual pressure that will be generated in the chamber. So another way to look at the situation is to calculate the allowable pressure for different foil thicknesses. By doing so the pressure, q, must be solved out from the equation above. This results in the equation:

$$q \leq \frac{t}{a} \left( \left( \frac{S}{k_1} \right)^3 \cdot \frac{1}{E} \right)^{1/2} \quad (\text{A2.4})$$

With this formula it is easy to predict the tolerance of different foil thicknesses i.e. how much pressure a specific foil thickness will tolerate before it brakes down.

### A2.1.a. Niobium foils

Mechanical properties for Niobium are fetched from table 2.1 in the thesis.

The first pressure tolerance is calculated for a 100 micron foil.  
According to equations A2.1 and A2.2:

$$S = 0.5F_U$$

$$S = 0.9F_Y$$

Where

$$F_U = 84847 \text{ psi}$$

$$F_Y = 79771 \text{ psi}$$

$$\left. \begin{array}{l} S = 0.5F_U = 0.5 \cdot 84847 \approx 42424 \\ S = 0.9F_Y = 0.9 \cdot 79771 \approx 71794 \end{array} \right\} \Rightarrow S = 42424 \text{ psi}$$

It is always the smallest quantity from the two equations above that sets the limit of the allowable stress.

Now it is possible to calculate the allowable pressure with help from equation A2.4 with:

$$t = 0.003937 \text{ ''}$$

$$K_1 = 0.346$$

$$a = 0.472 \text{ ''}$$

$$b = 1.575 \text{ ''}$$

$$E = 15228962 \text{ psi}$$

$$q \leq \frac{t}{a} \left( \left( \frac{S}{k_1} \right)^3 \cdot \frac{1}{E} \right)^{1/2} = \frac{0,003937}{0,472} \left( \left( \frac{42424}{0,346} \right)^3 \cdot \frac{1}{15228962} \right)^{1/2} \approx 92 \text{ psi}$$

92 psi is approximately the same as 6 bar

This pressure tolerance seems a little bit to low considering those reports that have shown pressures around 7 bar at beam intensities around 50  $\mu\text{A}$  [17].

A way to perk up the tolerance is to increase the thickness of the foil. A 150 micron foil will increase the tolerance 1.5 times since the thickness increases the term linear in the equation. This gives a tolerance of approximately 9 bar (133 psi).

Increasing the foil thickness will result in a greater energy degradation which will give a lower yield. An investigation of how much the degradation depends on the thickness of the Niobium foil is performed in fig A2.1:

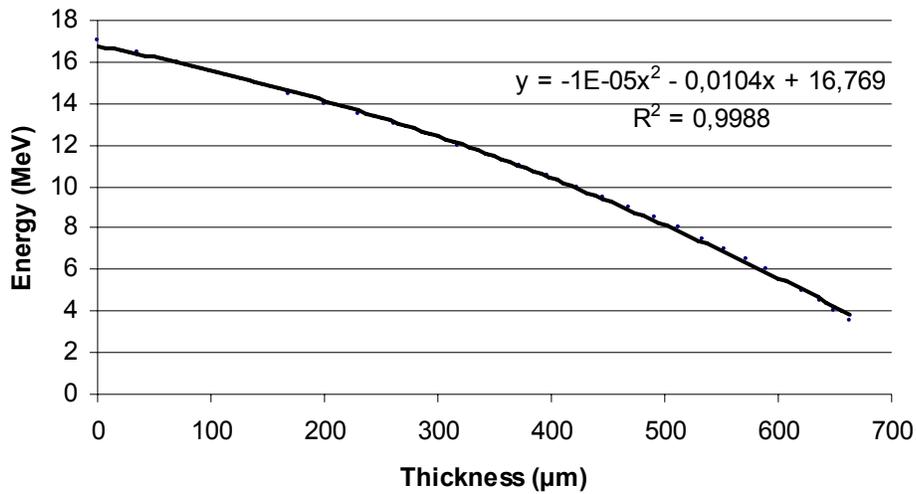


Fig A2.1: Energy degradation from 17 MeV for different Niobium foils thicknesses. Data from NIST [16]

The figure provides information of how much a specific foil thickness will degrade the energy. The first 50 micron foil will lower the impinging energy from 17 MeV to approximately 16.2 MeV. Another foil with a thickness of 100 microns will then degrade the energy to approximately 15.2 MeV. For a 150 micron target foil the total (200 micron) degradation will result in an energy of 14.7 MeV.

### A2.1.b. Havar foils

An alternative to the Niobium foils is to use a Havar foil which gives higher pressure tolerance but also a more non favorable activation. The tensile strength of Havar is very high according to table A2.1:

Modulus of elasticity (GPa)	200
Tensile strength (MPa)	1860
Density (g cm <sup>-3</sup> )	8.3
Thermal conductivity at 23C (Wm <sup>-1</sup> K <sup>-1</sup> )	14.7
Composition	Co 42.5%, Cr 20%, Mn 1.6%, Mo 2%, Ni 13%, W 2.8%

Table A2.1: Havar properties [12]

According to equation A2.1

$$S = 0.5F_U$$

Where

$$F_U = 269800 \text{ psi}$$

$$S = 0.5F_U = 0.5 \cdot 269800 \approx 134900 \text{ psi}$$

Since the tensile strength is so high the thickness of the foil can be decreased thus giving us a smaller energy degradation of the beam. A 50 micron foil (0.0019685”) gives the tolerance:

$$q \leq \frac{t}{a} \left( \left( \frac{S}{k_1} \right)^3 \cdot \frac{1}{E} \right)^{1/2} = \frac{0,0019685}{0,472} \left( \left( \frac{134900}{0,346} \right)^3 \cdot \frac{1}{29010000} \right)^{1/2} \approx 188,51 \text{ psi}$$

$$t = 0.00119685 \text{ “}$$

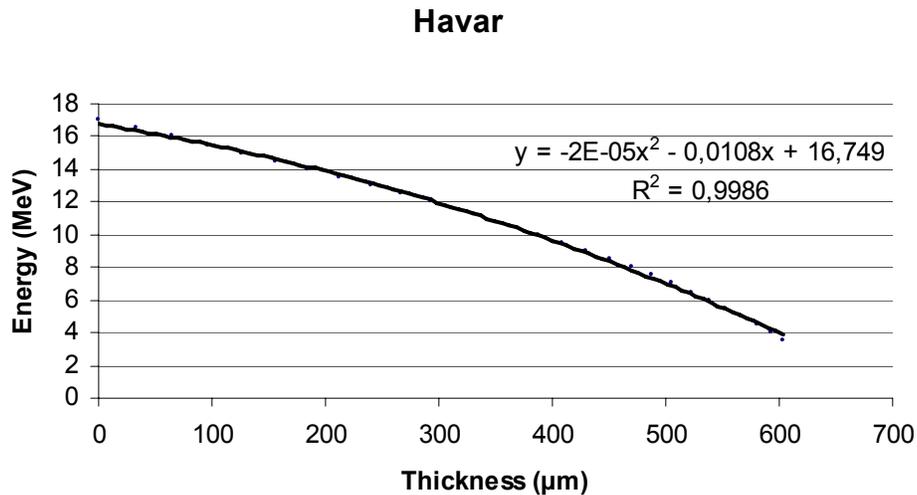
$$K_1 = 0.346$$

$$a = 0.472 \text{ “}$$

$$b = 1.575 \text{ “}$$

$$E = 29010000 \text{ psi}$$

188.51 psi is almost the same as 13 bar. With a 25 micron foil we will have a tolerance of 6.5 bar.



*Fig A2.2: Energy degradation from 17 MeV for different Havar foils thicknesses. The data are taken from NIST[16] for Iron, an element close to the Co (42.5 % of Havar). This assumption is ok since the variations in the data are very small in the energy region of 16-17 MeV.*

A 50 micron Havar foil plus a 25 micron at the machine will degrade the incoming energy to approximately 15.9 MeV according to fig A2.2.

## **A2.2. Foil deflection**

Because of higher pressure inside the chamber in contrast to the He-cooling pressure, the foil will bend outwards. This results in a thicker layer. This elongation can be calculated with equation from the guidelines. The calculation is made for 147 psi which corresponds to 10 bar.

$$y = k_2 \left( \frac{qa^4}{Et} \right)^{1/3} \quad (\text{A2.5})$$

y = window deflection (inch)

with :

$$t = 0.00119685''$$

$$E = 29010000 \text{ psi}$$

$$a = 0.472''$$

$$k_2 = 0.360$$

$$q = 147 \text{ psi}$$

equation A2.5 become:

$$y \approx 0.045 \text{ inch} \approx 1.2 \text{ mm}$$

If the elongation is approximated to the height of a triangle the volume can be calculated with equation:

$$V = \frac{A \cdot h}{2} = \frac{4 \cdot 1 \cdot 0.12}{2} = 0.24 \text{ cm}^3 = 0.24 \text{ ml} \quad (\text{A2.6})$$

An important parameter to regard is the fact that all calculations are made for temperatures of approximately 20 °C. When the temperature rises the mechanical properties will change. A benefit is the He-cooling from the other side of the foil which will give a counter pressure of 1 bar thus resulting in an “extra one bar allowance” for the chamber foil.