Abstract

ROBERTS, AMY NICOLE. Thermosyphon Targets Designed for the Production of $^{18}$F for use in Positron Emission Tomography. (Under the direction of Dr. Joseph M. Doster and Dr. Bruce W. Wieland.)

$^{18}$F is a radioisotope commonly used in Positron Emission Tomography. One way to produce $^{18}$F is to bombard $^{18}$O enriched water with protons, generated by a cyclotron. The production reaction is $^{18}$O(p,n)$^{18}$F. The purpose of this research was to model and build a more efficient target that can withstand higher proton beam currents and energies and produce greater yields of $^{18}$F. A thermosyphon was chosen as the basis for the target design. Thermosyphons transfer heat through evaporation and condensation. The concept is simple; the target water is allowed to boil and the water vapor rises out of the target chamber into a condenser region that is cooled along the outer surface. Condensation occurs along the condenser walls, and the condensate then runs down the length of the cylinder and back into the target chamber.

The thermosyphon target model showed that this form of target design was feasible, withstanding greater proton beam currents and energies and producing more $^{18}$F than conventional targetry. Three separate thermosyphon targets, each increasing in condenser heat transfer surface area and volume, were built and their performance was experimentally validated. The results of both the model and the experimental targets are discussed in detail.
THERMOSYPHON TARGETS
DESIGNED FOR THE PRODUCTION OF $^{18}$F
FOR USE IN POSITRON EMISSION TOMOGRAPHY

by

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Biography

Amy Roberts was born in Miami, Florida in September of 1978 to Pamela Robinson and Irvin Roberts and is the only child. She was educated in the public school system and graduated from Ledford Senior High School in Thomasville, NC in 1996. Following high school, she began her college career at North Carolina State University graduating in 2000 with a Bachelor of Science degree in Nuclear Engineering. She continued on at North Carolina State University to obtain her Master of Science degree in Nuclear Engineering, with a minor concentration in Biomedical Engineering. She is beginning her career with Knolls Atomic Power Laboratory in Schenectady, NY.
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Table of Contents

List of Figures ..................................................................................................................... v
List of Tables ....................................................................................................................... vi

1 Introduction .................................................................................................................... 1

2 Purpose ........................................................................................................................... 3
  2.1 Overall Purpose ....................................................................................................... 7
  2.2 Who this will benefit ............................................................................................... 7
  2.3 Previous Research/Target Designs ......................................................................... 8

3 Design Materials ............................................................................................................. 10

4 Model ............................................................................................................................. 14
  4.1 Design ................................................................................................................... 14
  4.2 Theory and Code Description ............................................................................... 16
  4.3 Results ................................................................................................................... 27

5 Experimental Target ..................................................................................................... 33
  5.1 Design ................................................................................................................... 35
  5.2 Experimental Apparatus and Procedure .............................................................. 42
  5.3 Theory and Problem Solution .............................................................................. 45
  5.4 Results ................................................................................................................... 50

6 Sources of Error ........................................................................................................... 63

7 Summary and Conclusions .......................................................................................... 66

References ......................................................................................................................... 68
List of Figures

Figure 1: $^{18}$O(p,n)$^{18}$F Cross Section .................................................................4
Figure 2: Saturation Yield in mCi/µA .................................................................5
Figure 3: Window/Sacrificial Grid Design .........................................................13
Figure 4: Thermosyphon Model .................................................................14
Figure 5: Detailed Thermosyphon Model ......................................................15
Figure 6: Flow Chart for solving the velocity on the secondary side .................20
Figure 7: Flow Chart for Thermosyphon Model Code ..................................26
Figure 8: Condenser Height vs. Beam Current ..........................................27
Figure 9: Heat Transfer Surface Area vs. Beam Current ...............................28
Figure 10: Pressure vs. Beam Current ..............................................................29
Figure 11: Void Fraction vs. Beam Current Fixed Height ...........................30
Figure 12: Void Fraction vs. Beam Current Fixed Pressure .........................32
Figure 13: Disassembled view of an Experimental Thermosyphon Target ........37
Figure 14: Thermosyphon 1 (TS-1) ...............................................................39
Figure 15: Thermosyphon 1 (TS-2) ...............................................................40
Figure 16: Thermosyphon 1 (TS-3) ...............................................................41
Figure 17: Instrumentation Schematic ...........................................................44
Figure 18: TS-1 Cross Sectional Vapor Flow Area .......................................49
Figure 19: TS-2 Cross Sectional Vapor Flow Area .......................................49
Figure 20: TS-3 Cross Sectional Vapor Flow Area .......................................50
Figure 21: TS-1 Sight Tube Data .................................................................51
Figure 22: TS-2 Sight Tube Data .................................................................52
Figure 23: TS-2 Sight Tube Data 2 ...............................................................52
Figure 24: TS-3 Sight Tube Data .................................................................53
Figure 25: TS-3 Sight Tube Data 2 ...............................................................53
Figure 26: Lower Bound Convective Heat Transfer Coefficient .................55
Figure 27: Heat Input vs. Heat Removed for TS-3 ......................................65
List of Tables

Table 1: Shows the relative size increase/decrease of the 3 experimental Thermosyphon targets................................................................. 42

Table 2: Calculated max beam current using the lower bound on convective heat transfer coefficient compared to the actual performance limit for each target......... 57

Table 3: Better estimate of target performance................................................................. 58

Table 4: Calculated versus Actual Convective Heat Transfer Coefficient ....................... 59

Table 5: Void Fraction at Performance Limit................................................................. 59

Table 6: TS-2 Pressure Fluctuation Data ...................................................................... 60
1 Introduction

Typical medical imaging techniques, such as CT scans and MRI’s, show the structure of the body being imaged. To diagnose a problem or disease it is also important to know what is taking place in that structure physiologically. Since all tissues operate on a chemical basis, knowing what chemical processes are occurring in the imaged tissue can provide information as to whether the tissue is diseased or not. One technique for imaging tissues physiologically is to attach a radioisotope to a biochemical that the body normally uses, and monitor the location of decay events. This type of imaging is known as positron emission tomography, hereafter called PET (Ollinger and Fessler, 1997).

PET has become a major diagnostic tool in determining the occurrence or stages of cancer because it shows on a fundamental level what is taking place within the tissues. One major problem with PET is the cost factor. The PET scanners themselves are very costly. In addition, the short half lives of radioisotopes normally used for these studies generally requires they be made on-site. A common method of generating radioisotopes used in PET scans is through the use of cyclotrons (Ollinger and Fessler, 1997).

Cyclotrons accelerate a beam of particles in a circular path, increasing their energy until the beam is deposited onto a target containing the element to be transmuted into the desired radioisotope. Cyclotrons were invented in the 1930’s and have been used since the 1950’s to produce radioisotopes for the medical industry.

The target consists of an attenuator and a beam window. The former reduces the energy of the beam to one that is favorable for the nuclear reaction to take place, the latter
creates the front barrier that houses the target material and is an area of reduced metal thickness that allows the beam to penetrate into the target material. The particle beam enters the target and collides with the atoms of the target material producing the desired radioisotope. Common production reactions used in positron emission tomography are: $^{18}$O$(p,n)^{18}$F, $^{16}$O$(p,\alpha)^{13}$N, $^{13}$C$(p,n)^{13}$N, $^{14}$N(d,n)$^{15}$O, and $^{14}$N$(p,\alpha)^{11}$C. The $^{18}$O$(p,n)^{18}$F reaction is the most often used and will be the focus of the remaining discussion.

The isotopes used in PET scans are positron emitters. The unstable radionuclide emits a positron, which looses energy through collisions with surrounding atoms and molecules. The positron then combines with an electron and annihilates producing a pair of 511 keV photons, emitted roughly 180° apart (Turkington, 1998). The photons are detected, allowing an image to be recorded. The image shows a map of the patient with the “hot spots” being the most physiologically active, and potentially cancerous cells.
2 Purpose

The purpose of this project is to model and design a more efficient target for the production of Fluorine-18, hereafter called $^{18}$F, for use in medical imaging scans. As stated before, bombarding $^{18}$O enriched water with protons generated by a cyclotron produces $^{18}$F. The protons deposit kinetic energy within the target resulting in direct heating of the water. Traditional target designs involve a stagnant water volume cooled by circulating water around the outside surface. Cooling fins surround the target body to enhance heat transfer. Due to the high cost of $^{18}$O enriched water it is desirable to keep the volume used in the target as low as possible. The risk of boiling the target water requires the cyclotron to operate at relatively low beam currents ($p^+/s$), which in turn equates to a lower beam density ($p^+/cm^3$). As the production rate of $^{18}$F is directly proportional to the beam current, lower beam density implies reduced $^{18}$F production. Higher beam currents would result in increased $^{18}$F production but would be accompanied by higher heat loads to the target. The heat input can be calculated from the beam current and proton energy as shown below:

$$1 \mu A = 10^{-6} \frac{C/s}{s} = \frac{10^{-6} \frac{C/s}{s}}{1.6 \times 10^{-19} \frac{C}{P^+}} = 6.25 \times 10^{12} \frac{P^+}{s}$$

$$\dot{Q} (\text{Watts}) = E (\text{MeV}/P^+) \times I (\mu A) \times \frac{6.25 \times 10^{12} \frac{P^+}{s}}{1 \mu A} \times \frac{1.6 \times 10^{-13} J}{\text{MeV}} = (J/s)$$
The terms \( \frac{6.25 \times 10^{12} \text{ } p^+/s}{1 \mu A} \times \frac{1.6 \times 10^{-13} J}{\text{MeV}} = 1.0 \left( \frac{J \cdot p^+}{\mu A \cdot \text{MeV} \cdot s} \right) \), show that the heat input to the target is merely the product of proton energy (MeV) multiplied by the beam current (\( \mu A \)), or \( \dot{Q} (\text{watts}) = E (\text{MeV}) \times I (\mu A) \).

As stated above, the more protons that bombard the target (higher beam current) the more \(^{18}\text{F}\) one can produce. In addition to higher beam currents, higher incident proton energies also increase \(^{18}\text{F}\) yield, as can be seen in Figure 2. The trade off being a higher heat load in the target.

**Figure 1:** \(^{18}\text{O}(p,n)^{18}\text{F}\) Cross Section. Plot created from the data of Hess et. al. (2001).
One would think, by looking at Figure 1, that it would not be advantageous to operate at higher incident proton energies since the cross section, or probability for an interaction, is much lower at higher energies. However, Figure 2 shows that higher incident proton energies are advantageous and have a dramatic increase on the yield of $^{18}$F.

The following explains why the saturation yield increases with higher proton energy, even though the probability for an interaction is much lower at high proton energies. This discussion is relevant to understanding how the beam deposits its heat energy as it moves through the target and is also relevant to understanding the Bragg Peak Removal target discussed in 2.3.
The proton enters the target water with an initial kinetic energy. After undergoing Coulombic interactions and radiation losses (bremsstrahlung), the kinetic energy of the proton is reduced after traveling a distance x along its path. Stopping power is defined as incremental energy lost (dE) per unit distance traveled (dx), or (-dE/dx). As the proton decelerates the stopping power increases further back into the target, reaches a peak, and drops off to zero (Faw and Shultis, 1999). This also shows that the energy, or heat deposition, increases further back into the target. Consequently, the higher heat deposition reduces the target water density and also creates vapor voids further back into the target. Another important point to note is that the stopping power is lower at higher energies, for example a 22 MeV proton has a lower stopping power than a 15 MeV proton (Faw and Shultis, 1999). Therefore, a higher energy incident proton travels further into the target before reducing the target water density and creating vapor voids. A higher energy incident proton “sees” more $^{18}$O atoms to interact with at the beginning of the target, than a lower energy incident proton. For these reasons, it is easy to see how a higher energy proton yields more $^{18}$F, as can be seen in the saturation yield plot in Figure 2 (Hess, et. al., 2001). It is important to note that the research by Hess, et. al. was carried out using $^{18}$O enriched gas. The target designs discussed in this document use $^{18}$O enriched water. The interactions of the protons with hydrogen in the target water result in a 17% decrease in saturation yield compared to $^{18}$O enriched gas. Figure 2 has been corrected for this 17% decrease and shows the saturation yields expected for $^{18}$O enriched water.
2.1 Overall Purpose

The purpose of this project is to model and build a target that can withstand higher beam currents/higher incident proton energies and produce greater yields of F-18. A thermosyphon was chosen as the basis for the target design. Thermosyphons transfer heat through evaporation and condensation. The concept is simple; the target water is allowed to boil and the water vapor rises out of the target chamber into a condenser region that is cooled along the outer surface. Condensation occurs along the condenser walls, and the condensate then runs down the length of the cylinder and back into the target chamber.

The first objective of this research was to develop a model of the thermosyphon target in order to size the cylindrical condenser region, or equivalently to determine the heat transfer surface area needed to adequately cool the target, based upon the specific cooling capabilities of the PET facility at Duke University Medical Center. Once the thermosyphon design was established, the second objective of the project was to build and test a thermosyphon target to experimentally validate its performance.

2.2 Who this will benefit

The beam power available in most commercial cyclotrons exceeds the heat removal capacity of current target technology by a factor of two to four. If fully implemented, the new target systems would allow a substantial increase in effectiveness of the approximately 250 PET cyclotrons now operating in the United States. In January of 1998, Medicare approved insurance coverage on PET scans for diagnosing seven
major tumor types, with at least three more anticipated. This has translated into an increase in the number of PET scans done annually in the United States, as well as a 40% annual growth in sales of PET cyclotrons and scanners. A target that has a higher yield would lower costs and increase efficiency for the facilities providing the scans. The number of cyclotron runs per day, or the irradiation length of each run could be lowered without loss in $^{18}\text{F}$ production. Cyclotron manufacturers will probably sell 25-50 new cyclotrons per year in the next few years, and if more efficient/higher yield targets are available and utilized, they will offer a dramatic increase in $^{18}\text{F}$ production as well as lower the cost to the consumers of the radioisotope (hospitals, insurance companies, and individual patients) compared to what would be available from the same cyclotrons with conventional targetry.

2.3 Previous Research/Target Designs

Conventional targets are cylindrical, with a 11.1mm diameter and 10mm depth that contains a fixed volume of water (0.97 cc) and a top-pressurized reflux chamber, cooled by running water over the outside surface of the target body. Cooling fins surround the target body to enhance heat transfer. This target design can withstand up to about 22 MeV protons at 20 $\mu$A of beam current for a total heat deposition of 440 watts, based on Duke’s daily production target capabilities.

A variation on the basic cylindrical target, tested at Duke Medical Center, utilizes a stepping motor to move the back wall of the target in and out. The purpose of this target design is to remove the Bragg peak associated with the protons. As discussed previously, the protons decelerate with a corresponding increase in the stopping power
further back into the target, ultimately reaching a peak, and dropping off to zero. This peak causes a large amount of heat energy to be deposited in the back of the target, potentially leading to the creation of vapor voids which limit the beam current and consequently $^{18}\text{F}$ production. By creating a target that has a movable back wall, one can adjust the target depth for any given beam current such that the Bragg peak is deposited in the back wall rather than the target water. Cooling the metal back wall is much easier and much more efficient than cooling the target water. Unfortunately this Bragg peak energy removal technique was not successful, due to the large variations in the density of the boiling target water and its effects on beam penetration. The information conveyed above regarding the Bragg peak removal target is a result of personal communication with Dr. Bruce Wieland, Duke University Medical Center.

Other target designs considered include a pumped re-circulating target. In this design the target water is pumped out of the target, through a heat exchanger where it is cooled and then back to the target. The pumped re-circulating target design can withstand much higher beam currents and proton energies, but requires a significantly larger target volume of expensive $^{18}\text{O}$ enriched water. Another downfall of this design is the pump itself. A practical design requires a pump that has a high mass flow rate, but a very low dead volume. This design is still being considered with the current emphasis on the fabrication of a pump that meets the design criteria.
3 Design Materials

The basic target design contains three main components. The first structure the proton beam encounters is the degrader, or attenuator. The degrader reduces the proton beam energy from the cyclotron energy to the desired energy for the nuclear reaction of interest. The proton beam then strikes the target “window”, which serves as a barrier between the target water and the rest of the device. The window must be thin enough to allow the majority of the protons to penetrate without further attenuation, but thick enough to assure it can withstand “reasonable” water pressure created by heating the target. The third major component is the target body itself, which contains the target material. These three components are housed inside a support structure, or casing. Figure 12 in 5.1 shows a disassembled target allowing one to see the various components of the target and how they fit together.

Proper materials selection for the degrader is important and depends on the incoming energy of the proton beam and the desired energy for the reaction. The degrader must be able to absorb and dissipate large amounts of energy to efficiently attenuate the beam. Aluminum is attractive because it requires a small thickness to achieve the desired energy loss. Aluminum also has a reasonably high thermal conductivity, thus it dissipates heat well and can be easily cooled. Typically, degraders are water cooled through fins that surround the outer portion of the degrader body (Helus & Ruth, 1987).

The “window” is a metal foil that acts as a barrier between the target water and the rest of the apparatus. The proton beam passes through the window and into the target
water. For this reason the window material must be extremely thin, in the 1-200 µm range, but with no micro-holes. The window must have good mechanical strength, fail only at high pressures, a high melting point, good resistance to radiation damage, and good thermal conductivity. Since the window is in direct contact with the hot target material it too must be able to dissipate heat effectively and be cooled, making thermal conductivity an important consideration. Since the beam has already passed through the degrader, further attenuation is undesirable. The window material should cause little scattering of the proton beam. Common window materials include Havar (proprietary alloy), tantalum, titanium, aluminum, and beryllium copper. The window material used depends on whether the target is operated at high or low pressure (Helus & Ruth, 1987).

Sources of impurities in liquid targets include the target body as well as the target window. Metal ions can be introduced into the target material by interactions of the proton beam with the metal target body and target window. The $^{18}$F produced is in ionic form and tends to react with metal ions from the target body and window. It is for this reason that materials selection for the target body and window is of utmost importance so as not to pollute or decrease the yield of the $^{18}$F product (Helus & Ruth, 1987).

The materials used in target bodies must provide good corrosion resistance, good resistance to activation, low contamination to target water, and must be easily cooled. Corrosion results from Oxygen in the materials forming metal oxides along surfaces of the body. The high amounts of heat generated and the interaction of the materials with radiation increase the corrosion rate. Activation is a major problem during irradiation, especially during long runs at high beam currents (Helus & Ruth, 1987).
A common choice for the target body is silver. Silver has a high thermal conductivity and therefore is easily cooled. On a radiochemical level, Silver has been shown to be more compatible with the production of $^{18}$F, leaching fewer ions into the solution than other metals (Helus & Ruth, 1987).

Current target designs operate at much higher pressures than standard low-pressure targets, with current target pressures as high as 500 psi. Operating at high-pressure means a higher boiling point and in the case of thermosyphons, smaller condenser heat transfer surface area. Higher pressures also allows for a higher proton beam current, directly resulting in more nuclear reactions and larger yields of $^{18}$F. One downside is that windows have a greater tendency to fail at these higher pressures. To combat this, a design proposed by Dr. Bruce Wieland at Duke Medical Center appears to circumvent this problem. Dr. Wieland has designed a grid structure on which the window rests. This grid is designed to provide strength and support to the window without significantly attenuating the beam. The grid uses 7 hexagon shaped holes that allow the beam to penetrate with little attenuation but also provides the needed support for the windows to stay intact at high pressures. Between the degrader and window grid, lies a sacrificial grid. The sacrificial grid matches the window grid identically, but precedes it and blocks out the 22 MeV protons that would otherwise deposit their heat energy in the window grid septa area, causing unnecessary heat addition to the target window and target water. Figure 3 shows the window/sacrificial grid design and was created by Robert Timberlake, Duke University machinist. The information conveyed
above regarding the window/sacrificial grid is a result of personal communication with Dr. Bruce Wieland, Duke University Medical Center.

Figure 3: Window/Sacrificial Grid Design
4 Model

4.1 Design

The first part of this project involves modeling a thermosyphon target and analyzing its heat transfer capabilities to determine if the design is feasible and warrants being built and tested. The equations and computer code written to model the thermosyphon will be discussed later. Figures 4 and 5 below show the modeled thermosyphon target (dimensions in inches).

Figure 4: Thermosyphon Model
The proposed thermosyphon target resembles two cylinders, one vertical (representing the condenser region) and one horizontal (representing the boiler region) that are joined to form the target. For the reasons stated previously, the target body material was chosen to be silver. 22 MeV protons enter the boiler region depositing their energy in the form of heat and boil the target water. The surge volume compensates for the thermal expansion of water when it is heated. Also, there is a volume increase due to the presence of both liquid and vapor (voiding in the beam strike) in the target water. Buoyancy forces cause the vapor bubbles to rise and enter the condenser region. The
The condenser region is cooled on the outside via cold water running through channels formed by the cooling fins. The cooling fins provide an increase in heat transfer surface area on the secondary side. Water vapor condenses when in contact with the cooler surface of the condenser region. Then condensate then runs down the length of the walls and back into the boiler region.

### 4.2 Theory and Code Description

A FORTRAN code was written to assess the feasibility of the thermosyphon target model, shown in Figure 4. This section will discuss the methodology and equations used to model the thermosyphon target. As discussed previously, the 22 MeV protons enter the target and deposit their heat energy. The target water begins to boil and the vapor rises into the condenser region where it condenses along the walls and flows back into the boiler region. The condenser region is cooled along its outer surface by coolant water flowing along the cooling fins.

Typically thermosyphons are initially evacuated, then filled with a liquid charge. The steady state pressure at which a thermosyphon operates is dictated solely by the cooling capacity of the system. For this reason the code is designed in such a way that the user inputs an operating pressure for the target along with the heat input (beam current and proton energy) and the code calculates the required condenser length. Physically, the code calculates how much heat transfer surface area is needed to adequately cool the target to maintain that operating pressure. Another variation of the code was written to allow the user to input a condenser length and heat input and the code will calculate the corresponding operating pressure. The code requires as input the inner...
diameter of the condenser region and thread form detail of the fins (refer to Figure 5).

Heat input into the target is given by $\dot{Q}(\text{watts}) = E(\text{MeV}) \times I(\mu A)$; therefore, the user must
input beam current ($\mu A$) and proton energy (MeV). The lengths and diameters of the
secondary side coolant lines connecting the target assembly to the inlet and outlet
manifolds need to be input, as does the secondary side inlet coolant temperature.

The user inputs a desired target operating pressure and the fluid properties are
determined by evaluating the steam tables at that pressure. Density is corrected to reflect
the fact that $^{18}$O water is denser than normal water. All other fluid properties must be
taken as those of normal water, as data on $^{18}$O specific water is unavailable.

On the secondary (coolant) side, cooling water from a supply manifold flows
along the cooling fins in the condenser region. Upon exiting the cooling fins, the coolant
flows to a retrieval manifold. The driving force behind this flow is a difference in
pressure of 72 psi between the two manifolds. The flow velocity can be obtained directly
from the pressure drop by (Todreas, 1990):

$$\Delta P = \frac{\rho v^2 A_x^2}{2} \left\{ \sum_i f_i L_i \frac{1}{D_{ei} A_{xi}} + \sum_j K_j \frac{1}{A_{xj}} \right\}$$  \hspace{1cm} (1)

For this design, there are three main forms losses on the secondary side. The
forms loss entering the condenser (contraction), the forms loss exiting the condenser
(expansion), and the forms loss associated with the spiral fins.
The forms loss associated with a sudden contraction is:

\[ K_{\text{con}} = 0.5(1 - \frac{A_{\text{small}}}{A_{\text{large}}})^{0.75} \] (Idelchik, 1996)

The forms loss associated with a sudden expansion is:

\[ K_{\text{exp}} = (1 - \frac{A_{\text{small}}}{A_{\text{large}}})^{2} \] (Idelchik, 1996)

The forms loss associated with the spiral fins is:

\[ K_{\text{fin}} = 0.0175 \times N \times \lambda_{el} \times \frac{R_{\text{curve}}}{D_{h}} \] (Idelchick, 1996)

Where \( N \) is the total angle traversed (\( N = 360^\circ \times \) number of encirclements) and:

\[ \lambda_{el} = \frac{A}{\text{Re}^{B}} \left( \frac{D_{\text{tube}}}{2R_{\text{curve}}} \right)^{C} \] (Idelchick, 1996)

A = 20, B = 0.65, C = 0.175 for \( \text{Re} \frac{D_{\text{tube}}}{2R_{\text{curve}}} < 600 \)

A = 10.4, B = 0.55, C = 0.225 for \( \text{Re} \frac{D_{\text{tube}}}{2R_{\text{curve}}} < 1400 \)

A = 5, B = 0.45, C = 0.275 for \( \text{Re} \frac{D_{\text{tube}}}{2R_{\text{curve}}} < 5000 \)

The forms losses for the spiral fins along with the frictional losses on the secondary side are dependent on Reynolds Number (Todreas, 1990):

\[ \text{Re} = \frac{\rho v D_{e}}{\mu} \] (2)
Where \( D_e \) is the equivalent diameter and \( \mu \) is the fluid viscosity. Functional forms of the friction factor have been developed for laminar and turbulent flow in smooth pipes (Todreas, 1990):

\[
f = \frac{64}{Re}, \quad \text{for } Re \leq 2300
\]

\[
f = 0.3164 Re^{-0.25}, \quad \text{for } 2300 < Re \leq 30,000
\]

\[
f = 0.184 Re^{-0.2}, \quad \text{for } Re > 30,000
\]

The first step in modeling the thermosyphon target is to determine the velocity of the coolant on the secondary side. Examination of the equations above shows that both the forms losses and friction losses are a function of Reynolds number, which is a function of velocity and fluid properties. Therefore, Equation 1 is transcendental in velocity. The fluid properties are taken to be those associated with the average bulk temperature on the secondary side. The following equations are also useful in calculating the secondary side velocity convergence criteria. See Figure 6 for a flow chart showing how the velocity was solved iteratively.

\[
\dot{m} = \rho v A_x
\]

\[
T_{\text{avg}} = \frac{T_{\text{inlet}} + T_{\text{outlet}}}{2}
\]

\[
\dot{Q} = \dot{m} C_p (T_{\text{outlet}} - T_{\text{inlet}})
\]

\[
\dot{Q} = 2\dot{m} C_p (T_{\text{avg}} - T_{\text{inlet}})
\]
Figure 6: Flow Chart for solving the velocity on the secondary side
Given the fluid velocity, the Dittus-Boelter correlation is used to determine the secondary side convective heat transfer coefficient (Todreas, 1990):

\[ Nu = C \text{Re}^{0.8} \text{Pr}^n \]  \( (3) \)

where:

\[ h_{sec} = \frac{Nu_{sec}k_{sec}}{De} \]  \( (4) \)

C = 0.23 and n = 0.4 when the fluid is heated or n = 0.3 when the fluid is cooled. Pr is the Prandtl number that is based on the specific fluid properties:

\[ Pr = \frac{C_p \mu}{k} \]

The inner wall temperature in the condenser section can be found from solution of the 1-D radial conduction equation:

\[ T_s = T_{avg} + \left\{ \frac{\ln \left( \frac{r_o}{r_i} \right)}{2\pi k L} + \frac{1}{h_{sec} A_{sec} \eta_{ov}} \right\} \]  \( (5) \)

Where \( T_{avg} \) is the average of the inlet and exit temperatures across the secondary side.

The \( \eta_{ov} \) term is the fin efficiency associated with the external cooling fins. The efficiency accounts for the increase in heat transfer area, while correcting for the increase in resistance to heat transfer from conduction within the fin itself. The overall fin efficiency is (Incropera, 1996):

\[ \eta_{ov} = 1 - \frac{NA_{fin}}{NA_{fin} + A_b (1 - \eta_{fin})} \]
N is the number of fins present, \( A_{\text{fin}} \) is the surface area of a single fin, \( A_0 \) is the surface area in the absence of any fins, and \( \eta_{\text{fin}} \) is the fin efficiency of a single fin, given by (Incropera, 1996):

\[
\eta_{\text{fin}} = C2 \frac{K_1(mr_1)I_1(mr_{2c}) - I_1(mr_1)K_1(mr_{2c})}{I_0(mr_1) + K_1(mr_{2c}) + K_0(mr_1)I_1(mr_{2c})}
\]

\[
C2 = \frac{(2r_1/m)}{r_{2c}^2 - r_1^2}
\]

\[
A_{\text{fin}} = 2\pi(r_{2c}^2 - r_1^2)
\]

\[
r_{2c} = r_2 + (t/2)
\]

\[
m = \left(\frac{2h}{kt}\right)^{1/2}
\]

Where \( r_1 \) is the outer radius of the condenser, \( r_2 = r_1 + \) thread depth, \( t \) is the fin thickness at its base, and \( k \) is the thermal conductivity of the fin material. \( I_0 \) and \( I_1 \) are zero and first order modified Bessel functions of the first kind. \( K_0 \) and \( K_1 \) are zero and first order modified Bessel functions of the second kind.

Equations 6, 7, and 8 are the governing equations used to model heat transfer on the primary side of the thermosyphon target and were derived for condensation along a flat wall (Incropera, 1996):

\[
\overline{h}_L = \frac{4}{3} 4^{-1/4} \left[ \frac{g\rho_f (\rho_f - \rho_g) k_i^3 h_{fg}}{\mu(T_{\text{sat}} - T_s) L} \right]^{1/4} = 0.943 \left[ \frac{g\rho_f (\rho_f - \rho_g) k_i^3 h_{fg}'}{\mu(T_{\text{sat}} - T_s) L} \right]^{1/4}
\]  

(6)

where: \( h_{fg}' = h_{fg} + 0.68C_p(T_{\text{sat}} - T_s) \)

Applying Newton’s Law of Cooling:

\[
Q = \overline{h}_L A(T_{\text{sat}} - T_s)
\]  

(7)
\[
\dot{m}_{\text{cond}} = \frac{Q}{h_{fg}} = \frac{h_L A (T_{\text{sat}} - T_s)}{h_{fg}} \tag{8}
\]

Knowing the surface temperature along the inner wall of the condenser it is now possible to solve for the condensation heat transfer coefficient. Substituting Equation 5 into Equation 7 gives:

\[
Q = \frac{1}{h_L A_{\text{prim}}} + \ln\left(\frac{r_o}{r_i}\right) \frac{1}{2\pi k L} + \frac{1}{h_{\text{sec}} A_{\text{sec}} \eta_{ov}} \left( T_{\text{sat}} - T_{\text{avg}} \right)
\]

The only unknown is the condenser length, \(L\), which is computed using Brent’s Algorithm to solve the transcendental equation (Atkinson, 1978).

It is possible for turbulence to exist in the condensate film, which greatly enhances heat transfer at the walls. Under these conditions, the condensation heat transfer coefficient is determined from the following correlations (Incropera, 1996):

\[
\dot{m} = \rho_f u_m b \delta
\]

\[
\text{Re}_\delta = \frac{4 \Gamma}{\mu} = \frac{4 \dot{m} \rho_f u_m \delta}{\mu} \tag{9}
\]

For laminar waves:

\[
\text{Re}_\delta = \frac{4 g \rho_f (\rho_f - \rho_g) b^3}{3 \mu^2}
\]

Express in terms of a modified Nusselt #:

\[
\frac{h \left( v_f^2 / g \right)^{1/3}}{k_f} = 1.47 \text{Re}_\delta^{-1/3} \quad \text{Re}_\delta \leq 30
\]
Kutateladze formed the correlation for the wavy laminar regime:

\[
\frac{h_i (v_i^2 / g)^{1/3}}{k_i} = \frac{Re_s}{1.08 Re_{\delta}^{1.22} - 5.2} \quad 30 \leq Re\delta \leq 1800 \quad (10)
\]

For the turbulent regime Labuntsov determined the correlation:

\[
\frac{h_i (v_i^2 / g)^{1/3}}{k_i} = \frac{Re_{\delta}}{8750 + 58 Pr^{-0.5} (Re_{\delta}^{0.75} - 253)} \quad Re\delta \geq 1800
\]

After solving for a condenser length, Equation 8 can be used to determine the condensate mass flow rate. Once the mass flow rate of the condensate is determined, Equation 9 can be used to find the Reynolds Number. A Reynolds number test is performed to see in which region the film lies, laminar, wavy laminar, or turbulent. The condensation heat transfer coefficient is recomputed, and a new condenser length determined. A new condensate mass flow rate and Reynolds Number can be calculated for the new condenser height and the process is repeated. A convergence loop was set up with the convergence criteria being a L2 norm on the Reynolds Number. See Figure 7 for a flowchart.

The void fraction within the beam strike area is required to insure the beam is completely stopped within the target volume. At steady state, the condensation mass flow rate is equal to the vapor flow rate out of the target volume. The vapor mass flow rate is related to the vapor volume fraction through (Wallis, 1969):

\[
\dot{m}_g = \alpha_g \rho_g v_g A_x \quad (11)
\]

where \( v_g = (1 - \alpha_g Co)^{-1} \left[ \frac{\sigma_{gg} c (\rho_f - \rho_g)}{\rho_f^2} \right]^{1/4} \), \( Co = 1.2 \)
Substituting (11) into (12):

\[
\frac{\dot{m}_g}{\alpha_g \rho_g A_s} = (1 - \alpha_g \mathrm{Co})^{-1} 1.53 \left[ \sigma_{gg} \frac{(\rho_f - \rho_g)}{\rho_f^2} \right]^{1/4}, \mathrm{Co} = 1.2
\]

Which is a transcendental equation in $\alpha_{gs}$ and may be solved iteratively.

A surge volume is required to allow for the thermal expansion of the target water when heated. Also contributing to that surge volume is the increase in volume due to the mixture of liquid and vapor in the boiler region. Given the liquid density and vapor volume fraction in the target region, the surge volume may be computed from

\[
\rho_m = \alpha_g \rho_g + (1 - \alpha_g) \rho_f
\]

\[
V_m = \left( \frac{\rho_f}{\rho_m} \right) V_o
\]

\[
\Delta V_{\text{increase}} = V_m - V_o
\]

where $V_o$ is the liquid charge at zero-power input.
BEGIN

Input:
Geometry
Fluid Props
System Char.

Brent’s
Algorithm
Root Solver

Output:
Condenser height/heat
transfer surface area

Function determining the heat
transfer capacity of
the condenser

Function determining the heat
transfer capacity of
the condenser with
wavy laminar
approx.

Calculate mass flow rate & Reynolds
Number of Condensate Film

Brent’s
Algorithm
Root Solver

Wavy
Laminar

Reynolds #

NO

YES

Convergence
Criteria

End

Function determining
the void fraction in
boiler region

Output:
Final Condenser Height & Void Fraction

Calculate new mass flow rate & Reynolds Number of Condensate Film

Figure 7: Flow Chart for Thermosyphon Model Code
4.3 Results

A code was written that allowed the user to input a desired operating pressure and heat input with the resulting output being the length of the condenser necessary to cool the target. Figure 8 is a plot of condenser height versus beam current for an operating pressure of 500 psi. This operating pressure was chosen because of the higher boiling point and the target window’s ability to withstand that pressure.

![Condenser Height vs. Beam Current](image)

**Figure 8: Condenser Height vs. Beam Current**

The enhancement in the heat transfer rate resulting from turbulence in the condensate film is clearly seen in Figure 8. Since this plot is at a constant pressure of 500 psi, the enhanced heat transfer from turbulence results in a smaller condenser height, or heat transfer surface area. To design a target to withstand 60 micro amps of 22 MeV protons (1.32 kilowatts) would require a condenser height of 0.8 inches.
Figure 9 is a variation of Figure 8, this time plotting heat transfer surface area versus beam current. The inner diameter of the condenser modeled was 10mm, or 1cm, therefore the heat transfer surface area is $A_{\text{surf}} = \pi DH$, where H is the height shown in Figure 8. Plotting the output in this manner helps to predict the performance of the condenser region in the experimental targets.

![Heat Transfer Surface Area vs Beam Current](image)

Figure 9: Heat Transfer Surface Area vs. Beam Current

Another variation of the code was written to allow the user to input a condenser height and heat input with the resulting output being the target operating pressure. Two condenser lengths were examined, 0.7 inches and 0.6 inches. Figure 10 shows the results of pressure versus beam current for a fixed condenser height.
At low beam currents up to about 20 micro amps, there is very little pressure difference for the two heights and film condensation models. Above 20 micro amps, the target with the greater condenser length (and heat transfer area) operates at lower pressures for a given heat input. This is as expected. Again, it is important to note the dramatic effects of a wavy condensate film. The waves in the film result in greater heat transfer at the wall of the condenser, therefore the target operates at a lower pressure.

Figure 10 is especially important when discussing start up conditions. Start up involves gradually increasing the beam current incident on the target. This creates several problems. As can be seen in Figure 10, the target will start off at a low pressure and increase to the desired operating pressure as the beam current incident on the target increases to the operating conditions. At low beam currents/low pressures, the fixed
condenser length/cooling capacity exceeds what is needed to cool the target. When the pressure the in the target is low the boiling point too is very low. This causes increased boiling in the target at low incident beam currents. Because of this, the void fraction in the boiler is very high. This causes the proton beam to penetrate through the vapor voids and deposit into the back wall of the target. If a large portion of the proton beam is deposited in the back wall of the target, that leaves only a small amount of protons to react with a small amount of target water to produce $^{18}$F. Not only is the yield decreased, but also a large amount of heat is being deposited into the back wall, and therefore one runs the risk of physically damaging the target. Figure 11 below shows how the void fraction behaves for the fixed condenser heights of 0.7 inches and 0.6 inches.

![Void Fraction vs. Beam Current](image)

Figure 11: Void Fraction vs. Beam Current
At start up conditions, typically 5 micro amps, 81% of the initial water charge is vapor. Because vapor is much less dense than water, the majority of the proton beam would pass through the boiler region and deposit its heat in the back wall of the target. Damage to the target body is probable in this case. As Figure 10 shows, as the beam current increases the pressure increases and results in a higher boiling point. A higher boiling point causes the void fraction to decrease, as observed in Figure 11. One thing to note again is the effect of the wavy condensate film. As stated earlier, a wavy film causes a decrease in target pressure for a fixed height. This lower pressure causes the void fraction to increase. From Figures 10 and 11 it is apparent that in order to avoid issues associated with start up, it is necessary to apply an over pressure at the desired operating pressure to prevent the target from voiding at low power inputs.

Figure 12 shows the behavior of vapor voiding at a constant pressure. These void fractions are independent of condenser length, and plotted for a constant pressure of 500 psi. If a target was desired to operate at 40 micro amps and 500 psi, the condenser height should be approximately 0.5” (Figure 8). The void fraction at steady state operation of that target would be 50% (Figure 12). The thermosyphon target modeled had a target depth of 10mm, the same depth/volume of the typical targets in use. If 50% of the boiler region is vapor, that leaves an effective water thickness of 5mm. The range of a 22 MeV proton is approximately 7mm. Therefore the target depth (or volume) needs to be increased by 50% in order to stop all the protons. Current targets reach their performance limits at around 20 micro amps of beam current. By utilizing a thermosyphon design, one can double the incident beam current to 40 micro amps and double the yield of $^{18}\text{F}$.
Figure 12: Void Fraction vs. Beam Current

produced. The trade off is a 50% increase in target water volume necessary to stop the protons.
5 Experimental Target

Since a thermosyphon target allows the target water to boil and form a vapor region, it was necessary to determine how targets behave at the onset of boiling. It is difficult if not impossible to characterize the boiling behavior of a target through standard correlations for boiling heat transfer. Those correlations work under the assumption of an externally applied heat source that cause vapor bubbles to form along nucleation sites on the material being heated. Once the vapor bubbles grow large enough, buoyancy forces cause the bubble to depart the heated surface. The correlations used to determine the heat transfer coefficients for this type of boiling phenomenon are well documented. A target behaves very differently. The incident proton beam deposits its heat energy while traveling through the target water. Therefore the heat is internally generated and boiling takes place in the beam strike.

It was hypothesized that two possible scenarios existed when a target began to boil. The first is that the target may be taking advantage of turbulence that would be the result of vapor bubbles being formed in the beam strike. When the vapor bubbles form, enhanced turbulence would allow the bubbles to migrate towards the outer wall of the target. The bubbles would encounter cooler fluid outside of the beam strike or at the wall and the bubbles would collapse. The collapse would cause fluid to rush in to fill the void. This process would create turbulence and produce internal mixing that would greatly enhance heat transfer. The second scenario involves vapor bubbles forming in the beam strike and buoyancy forces causing the vapor to rise to the top forming a condenser
If the latter were found to be true, then a thermosyphon target design would definitely be advantageous.

To investigate these two scenarios, first a cylindrical target was used with no additional space above the beam strike for a condenser region to form. A port was placed at the bottom of the target that allowed for water to be displaced out of the target due to thermal expansion and vapor voiding, this port will hereafter be referred to as the “sight tube”. Since there was no additional space above the beam strike, any vapor void formed at the top of the target would allow a large fraction of the beam to pass through the target and deposit all of its heat energy into the back wall of the target. With the onset of the vapor void at the top of the target, water is expelled from the target into the sight tube, and a large fraction of the beam passes through the vapor and deposits its energy into the back wall, not the water. Subsequently, the vapor void along with the water remaining in the target is cooled causing the vapor void to collapse and the expelled water to rush back into the target. If the first scenario were true and vapor bubbles were forming in the beam strike then moving out into cooler water surrounding the beam strike and collapsing, one would expect more of a “jitter” in the sight tube as opposed to the large fluctuations in displaced water. The jitter would represent the bubbles forming, moving out into cooler water, collapsing, and water rushing in to fill the void left by the bubble creating turbulence.

After running experiments on the cylindrical target with no additional space above the beam strike, it was determined that both scenarios are in fact valid. At very low power throughput, or very low beam currents, the jitter effect was evident in the sight
tube representing vapor bubbles forming in the beam strike then collapsing in the cooler surrounding fluid. This is nothing more than subcooled boiling, caused by local vapor voiding in the beam strike followed by bubbles moving out into the surrounding subcooled target water and collapsing. At higher power throughput, or higher beam currents, the target was heated to the saturation point and net voiding began which generated a large vapor space at the top of the target. This was evident from the large fluctuations in volume expelled from the target suggesting that a vapor void was forming at the top of the target and the beam strike was being invaded by vapor. With this determined it was known that a thermosyphon target design was in fact a viable option. The thermosyphon design creates a separate region above the beam strike that allows a vapor volume to form at the top of the target and puts into practice the efficient condensation mode of heat transfer. The limitation on target performance is determined to be when the heat input into the target causes the vapor volume to exceed what is allotted in the condenser region, or in other words, the volume of vapor in the condenser region invades the beam strike.

5.1 Design

The data obtained from the cylindrical target with no additional space above the beam strike implied that a vapor void does form at the top of the target, which would make a thermosyphon target design practical. Also, the model of the thermosyphon target showed the target was able to withstand higher beam currents/proton energies. To confirm the model results, three different thermosyphon targets were designed, built, and
tested. Modifications to the as built targets from the modeled target, included the ability to cool the condenser section, as well as the target body, with the goal to improve target performance even further. In addition, provisions were made to operate the target with a controlled over pressure.

Machining a target with the geometry shown in Figures 4 and 5, would create major design changes in the existing target assembly. The most notable changes would occur in the flanges, or casing, that house the various parts of the target, see Figure 13 and its descriptions. In order to minimize facility changes, it was decided to use standard designs for the target assembly and only modify the target body, or Part F, by machining a condenser region above the beam strike.

The major difference between the three experimental targets is an increase in volume and heat transfer surface area in both the boiler and condenser regions. An increase in condenser volume is particularly advantageous because that allows the target to withstand higher beam currents before the vapor starts to invade the beam strike. Figure 13 gives a view of how a total target system looks when disassembled.
Figure 13: Disassembled view of an Experimental Thermosyphon Target

- **Part A:** The front flange is the casing that holds the collimator, degrader, and sacrificial grid in place. It contains the inlet and exit ports for the cooling water to cool the collimator, degrader, and sacrificial grid assembly. These inlet and exit ports contain thermocouples to monitor the temperature rise in the cooling water across this piece.

- **Part B:** The collimator, degrader, and sacrificial grid are shown here. The collimator serves to focus the proton beam after it leaves the cyclotron. The degrader as discussed in the previous section attenuates the beam from 27 MeV to 22 MeV. The sacrificial grid blocks out the 22 MeV protons that would otherwise deposit their heat energy in the window grid septa area. This piece is made entirely out of aluminum for the reasons discussed in the previous section. This piece is also finned to enhance heat transfer surface area for the coolant water.

- **Part C:** The insulator flange houses the port for the vacuum pump.

- **Part D:** The window grid is designed to provide strength and support to the window without significantly attenuating the beam. The grid uses 7 hexagon shaped holes that allow the beam to penetrate with little attenuation but also provides the needed support for the windows to stay intact at high pressures. This piece is also made out of aluminum and finned to enhance heat transfer surface area.
• Part E: The target flange provides casing for the window grid, window, and target body. It also contains the inlet and exit ports for the cooling water to cool the window grid, window, and target body. These inlet and exit ports contain thermocouples to monitor the temperature rise in the cooling water across this piece.
• Part F: This piece is the thermosyphon, or target body, and is shown in detail in Figures 14, 15, and 16. The target body houses the $^{18}$O enriched water and is where the reaction takes place to form $^{18}$F. The window is a metal foil that acts as a barrier between the target water and the rest of the apparatus. The proton beam passes through the window and into the target water. This piece is made out of fine silver. This piece is also finned to enhance heat transfer surface area for the coolant water.
• Part G: This piece is the back flange. It forms the back wall of the target body. It contains the fill and drainage ports that are used to fill the target with the $^{18}$O enriched water and drain the target at the end of the run. It also contains the inlet and exit ports for the cooling water to cool the back wall of the target. These inlet and exit ports contain thermocouples to monitor the temperature rise in the cooling water across this piece.

The following are the CAD drawings for Thermosyphon 1 (TS-1), Thermosyphon 2 (TS-2), and Thermosyphon 3 (TS-3) created by Robert Timberlake, Duke University machinist. The dimensions are given in inches. Table 1 shows the relative size increase/decrease between the three experimental targets.
Figure 14: Thermosyphon 1 (TS-1)
Figure 16: Thermosyphon 3 (TS-3)
<table>
<thead>
<tr>
<th>Target</th>
<th>Total Volume (cc)</th>
<th>Condenser Volume (cc)</th>
<th>Boiler Volume (cc)</th>
<th>Percent Increase Condenser</th>
<th>Percent Increase Boiler</th>
</tr>
</thead>
<tbody>
<tr>
<td>TS-1</td>
<td>1.33</td>
<td>0.25</td>
<td>1.08</td>
<td></td>
<td></td>
</tr>
<tr>
<td>TS-2</td>
<td>1.55</td>
<td>0.45</td>
<td>1.10</td>
<td>TS-1 $\rightarrow$ TS-2 = 80%</td>
<td>TS-1 $\rightarrow$ TS-2 = 1.85 %</td>
</tr>
<tr>
<td>TS-3</td>
<td>1.54</td>
<td>0.59</td>
<td>0.95</td>
<td>TS-1 $\rightarrow$ TS-3 = 136%</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>TS-1 $\rightarrow$ TS-3 = -12%</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>TS-2 $\rightarrow$ TS-3 = -13.6%</td>
<td></td>
</tr>
</tbody>
</table>

Table 1: Shows the relative size increase/decrease of the 3 experimental Thermosyphon targets.

5.2 Experimental Apparatus and Procedure

An evaporative thermosyphon works by transferring heat from a boiler section to a condenser at a higher elevation. To fill the target, a vent line at the top of the target is open, and the target is completely filled from the bottom. The vent line is then sealed and the target is pressurized to 500 psi by applying a Helium gas overpressure to a water column connected to the fill line at the bottom of the target. When beam current is applied, the target water thermally expands out of the bottom of the target against the imposed pressure. As a vapor void is formed at the top of the target in the condenser region, additional water is forced out of the bottom against the imposed pressure. The device is self-regulating in that the vapor void formed is only large enough to transfer the heat introduced by the beam. The vapor condenses when power is removed and water flows back into the target to refill the target volume with liquid, or in other words, it goes back to the original condition at zero power input.
The experiments on the thermosyphon targets were run without sacrificial and window grids, which avoids having to correct the data for the beam lost in the grid septa. Figure 17 shows the instrumentation schematic for the thermosyphon target experiments and was created by Claudio Illan, Duke University Medical Center. The sight tube was used to monitor water expelled from the bottom of the target due to thermal expansion of the heated liquid and also due to formation of a vapor void in the condenser region when under beam bombardment. A TV camera was placed inside the cyclotron vault and used to observe the water level in the sight tube while the target was operating. The water level along with the incident beam current was recorded by hand. Valves and pumps were operated via field point modules and Labview software, with programs written to load with natural or $^{18}$O water, deliver, rinse, and blow dry. Thermocouple data along with pressure transducer data was recorded and displayed on line with Labview.
5.3 Theory and Problem Solution

The data acquired during the experiments included the volume of water expelled from the target, the target pressure, temperatures across the secondary side components, and the mass flow rate of the coolant on the secondary side. Since this target has internally generated heat, standard correlations for boiling heat transfer cannot be used; and boiling heat transfer coefficients must be inferred from the experimental data. From a steady state energy balance it can be shown that for the experimental target:

$$Q = Q_{\text{convection}} + Q_{\text{condensation}}$$

This simply states that the total heat removed from the target is a combination of the of the heat being removed from the boiler region, due to convection, and the heat removed from the condenser region, due to condensation. Another way of writing the energy balance is:

$$Q = h_{\text{boiler}} A_{\text{boiler}} (T_{\text{sat}} - T_s) + h_{\text{condenser}} A_{\text{condenser}} (T_{\text{sat}} - T_s)$$

The primary side surface temperature, $T_s$, is determined from:

$$T_s = T_{\text{avg}_\text{sec}} + Q \left\{ \frac{\ln \left( \frac{r_o}{r_i} \right)}{2\pi k L} + \frac{1}{h_{\text{sec}} A_{\text{sec}} \eta_{ov}} \right\}$$

where:

$$\eta_{ov} = 1 - \frac{N A_{\text{fin}}}{N A_{\text{fin}} + A_b} (1 - \eta_{\text{fin}})$$
\[ \eta_{\text{fin}} = C_2 \frac{K_1 (mr_1) I_1 (mr_{2c}) - I_1 (mr_1) K_1 (mr_{2c})}{I_0 (mr_1) + K_1 (mr_{2c}) + K_0 (mr_1) I_1 (mr_{2c})} \]

\[ C_2 = \frac{(2r_1 / m)}{r_{2c}^2 - r_1^2} \]

\[ A_{\text{fin}} = 2\pi (r_{2c}^2 - r_1^2) \]

\[ r_{2c} = r_2 + (t / 2) \]

\[ m = \left( \frac{2h}{kt} \right)^{1/2} \]

As shown in figures 14, 15, and 16 the experimental target closely resembles condensation in a horizontal pipe, or cylinder, rather than flat wall condensation as was derived for the model above. For this reason it is necessary to use an empirically derived correlation for the heat transfer coefficient in horizontal tubes/cylinders.

\[ \bar{h}_{\text{condenser}} = 0.555 \left[ \frac{g\rho_\ell (\rho_\ell - \rho_v) k_v h'_{fg}}{\mu_1 (T_{\text{sat}} - T_s) D} \right]^{1/4} \quad \text{(Incropera, 1996)} \]

Where for this case the modified latent heat of vaporization is:

\[ h'_{fg} = h_{fg} + \frac{3}{8} c_{p,\ell} (T_{\text{sat}} - T_s) \quad \text{(Incropera, 1996)} \]

When analyzing the heat transfer coefficient for the condenser region the diameter (D) was taken to be the equivalent diameter of the target, or \( D_{eq} = \frac{4A_x}{P_w} \), where \( A_x \) is the cross sectional area and \( P_w \) is the wetted perimeter.

The experimental data was used to estimate the convective heat transfer coefficient in the boiler. A 1cm rise in the sight tube level equates to 70.685 micro liters of target water expelled. Assuming that the thermal expansion in the target is linear it can determine how much of the expelled water is due to thermal expansion and how
much is due to a vapor void formation. Of additional interest was the amount of water
volume expelled that would cause the vapor void to invade the beam strike, thus causing
the target to fail. This volume could be estimated as indicated below.

\[ \beta = \text{Volumetric thermal expansion coefficient} \]

\[ \beta = -\frac{1}{\rho_i} \frac{\rho_f - \rho_i}{T_f - T_i} \]

\[ \rho_i = 998.66 \text{ kg/m}^3, \rho_f = 811.48 \text{ kg/m}^3, T_i = 18^\circ C, T_f = 467.01^\circ F = 241.672^\circ C \]

\[ \Delta T = 241.672^\circ C - 18^\circ C = 223.672^\circ C \]

\[ \beta = -\frac{1}{998.66} \frac{811.48 - 998.66}{241.672 - 18} = 8.37973 \times 10^{-4} \text{ } ^\circ C \]

\[ \Delta V = V_i \beta \Delta T \]

**TS-1**

Thermal Expansion:

\[ \Delta V = 1.33cm^3 \left( \frac{8.37973 \times 10^{-4}}{^\circ C} \right) 223.672^\circ C = 0.24928cm^3 = 249.28\mu L \]

Volume Condenser Region = 251.8\mu L

Total to invade beam strike = 249.28 + 251.8 = 501.1\mu L

**TS-2**

Thermal Expansion:

\[ \Delta V = 1.55cm^3 \left( \frac{8.37973 \times 10^{-4}}{^\circ C} \right) 223.672^\circ C = 0.290518cm^3 = 290.52\mu L \]

Volume Condenser Region = 449.67\mu L

Total to invade beam strike = 290.52 + 449.67 = 740.19 \mu L
TS-3
Thermal Expansion:
\[ \Delta V = 1.54 \times 10^{-3} \left( \frac{8.37973 \times 10^{-4}}{\circ C} \right) \times 223.672 \circ C = 0.28864 \times cm^3 = 288.64 \mu L \]

Volume Condenser Region = 591.16 \mu L

Total to invade beam strike = 288.64 + 591.16 = 879.8 \mu L

This information was used to experimentally determine the convective heat transfer coefficient for the boiler region and will be discussed in a later chapter. Also computed was the void fraction in the boiler region at the performance limit (max heat input). This was carried out in exactly the same manner as in the model:

\[ \dot{m}_g = \alpha_g \rho_g v_g A_x \]

\[ v_g = \left(1 - \alpha_g Co\right)^{-1} 1.53 \left[ \frac{\sigma_{gg} (\rho_f - \rho_g)}{\rho_f^2} \right]^{1/4}, Co = 1.2 \quad \text{(Wallis, 1969)} \]

\[ \frac{\dot{m}_g}{\alpha_g \rho_g A_x} = \left(1 - \alpha_g Co\right)^{-1} 1.53 \left[ \frac{\sigma_{gg} (\rho_f - \rho_g)}{\rho_f^2} \right]^{1/4}, Co = 1.2 \]

Figures 18, 19, and 20 show the cross-sectional vapor flow areas for TS-1, TS-2, and TS-3 at the performance limit. This limit is determined to be the maximum vapor volume that can form before invading the beam strike.
Figure 18: TS-1 Cross Sectional Vapor Flow Area = 0.137906 sq. in.

Figure 19: TS-2 Cross Sectional Vapor Flow Area = 0.171963 sq. in.
5.4 Results

Two main conclusions were drawn from the thermosyphon model. The first was that by applying an over pressure the problems associated with start up could be avoided. The second was that the thermosyphon target design was feasible if only condenser region were cooled. It would seem reasonable then that by cooling both the boiler and condenser regions a gain in target performance would result. Both of these points were applied to the experimental targets. The entire target body along with the back wall was cooled in the experiments. A helium gas over pressure was applied to the water column connected to the bottom of the target. The first step in analyzing the data was to experimentally determine the convective heat transfer coefficient in the boiler region. These boiling heat transfer coefficients are important for estimating target performance for conditions outside of the test set. Since these targets have internal heat generation, typical surface heated boiling heat transfer correlations are not applicable. The following
figures show the volume of water expelled versus incident beam current for all three thermosyphon targets. The blue line in each figure represents how much water should be expelled to thermally expand room temperature water to saturation conditions at 500 psi assuming linear thermal expansion. The yellow line represents how much water must be expelled for the vapor at the top of the target to invade the beam strike, and thus cause the target to reach its performance limit.

Figure 21: TS-1 Sight Tube Data
Figure 22: TS-2 Sight Tube Data

Figure 23: TS-2 Sight Tube Data 2

52
Figure 24: TS-3 Sight Tube Data

Figure 25: TS-3 Sight Tube Data 2
As discussed previously, when acquiring sight tube data an oscillation in the water expelled was observed. Figures 21-25 show both the high and the low swing of the water expelled. Where the high swing is the maximum and the low swing is the minimum water expelled for a given beam current. At low beam currents the volume expelled remains steady until the onset of subcooled boiling. Vapor bubbles displace water out of the target. When the vapor bubbles move out into cooler fluid they collapse and water rushes in to fill the void left by the bubble. This can be observed as a “jitter” (small high to low swing) in the sight tube. After the water has reached the saturation point at 500 psi, a much more pronounced high to low swing, due to the rapid boiling can be observed.

Thermosyphon 1 reaches the thermal expansion limit at around 18 micro amps, Thermosyphon 2 at 22 micro amps, and Thermosyphon 3 at 35 micro amps. Thermosyphon 1 reaches its performance limit at 26.4 micro amps, based on linear extrapolation. Thermosyphon 2 reaches its performance limit at 34.63, based on linear extrapolation. Finally the cyclotron was unable to produce enough beam current to determine the performance limit of Thermosyphon 3. At 45 micro amps, or 990 watts, the target was still performing below its maximum heat input.

The data obtained from TS-1 and TS-2 were used to predict the performance of TS-3. At any beam current below the thermal expansion line, the target was considered to be a full liquid target. There are vapor bubbles in the beam strike that move out to cooler surrounding fluid and collapse, but the primary mode of heat transfer is assumed
to be convection at the target walls. From the sight tube data, one can predict the bulk target water temperature. The equations governing the heat transfer are:

$$\Delta T = \frac{\Delta V}{V_i \beta}$$

$$T_{\text{bulk}} = \Delta T + T_{\text{initial}}$$

$$UA = \frac{1}{h_{\text{boiler}} A_{\text{prim}} \ln\left(\frac{r_o}{r_i}\right) \frac{1}{2\pi kL} + \frac{1}{h_{\text{sec}} A_{\text{sec}} \eta_{\text{av}}}}$$

$$Q = UA \left(T_{\text{bulk}} - T_{\text{bulk sec}}\right)$$

The heat input is known via \( \dot{Q} \text{(watts)} = E \text{(MeV)} \times I \text{(\muA)} \). Everything in the equation is either known or can be calculated therefore one can extract \( h_{\text{boiler}} \). This gives a lower bound estimate on the heat transfer coefficient, since the heat transfer coefficient will be much higher when the target is rapidly boiling. Figure 26 shows the lower bound estimate of convective heat transfer coefficient vs. beam current.

![Estimate of Convective Heat Transfer Coefficient vs. Beam Current](image.png)

Figure 26: Lower Bound Convective Heat Transfer Coefficient
convective heat transfer coefficients for each of the three thermosyphon targets.

The next step was to use the lower bound convective heat transfer coefficient to determine the max heat input into the target at the performance limit, where the vapor void in the condenser region invades the beam strike. The equations below are used to perform that calculation and it is assumed that the surface temperature on the inner wall of the target is the same in both the boiler and condenser regions.

\[
Q = Q_{\text{convection}} + Q_{\text{condensation}}
\]

\[
Q = h_{\text{boiler}} A_{\text{boiler}} (T_{\text{sat}} - T_s) + h_{\text{condenser}} A_{\text{condenser}} (T_{\text{sat}} - T_s)
\]

\[
T_s = T_{\text{bulk}} + Q \left[ \ln \left( \frac{r_o}{r_i} \right) \frac{1}{2\pi k L} + \frac{1}{h_{\text{sec}} A_{\text{sec}} \eta_{ov}} \right]
\]

\[
\eta_{ov} = 1 - \frac{N_{A_{\text{fin}}}}{N_{A_{\text{fin}}} + A_b} (1 - \eta_{fin})
\]

\[
\eta_{fin} = C2 \frac{K_1(mr_i)I_1(mr_{2c}) - I_1(mr_i)K_1(mr_{2c})}{I_0(mr_i) + K_1(mr_{2c}) + K_0(mr_i)I_1(mr_{2c})}
\]

\[
C2 = \frac{(2r_i/m)}{r_{2c}^2 - r_i^2}
\]

\[
A_{\text{fin}} = 2\pi(r_{2c}^2 - r_i^2)
\]

\[
r_{2c} = r_2 + (t/2)
\]

\[
m = \left( \frac{2h}{kt} \right)^{1/2}
\]

\[
\overline{h}_{\text{condenser}} = 0.555 \left[ \frac{g \rho_v (\rho_i - \rho_v) k_i^3 h'_{fg} \mu_{i} (T_{\text{sat}} - T_s) D}{\mu_{i} (T_{\text{sat}} - T_s) D} \right]^{1/4}
\]
Table 2 shows the calculated max beam current using the lower bound on convective heat transfer coefficient compared to the actual performance limit for each target.

![Table 2](https://example.com/table2.png)

One can see that using the lower bound convective heat transfer coefficient results in conservative values for maximum beam current, compared to the actual target performance.

As an alternative method for estimating the performance limit, we again use the assumption that any beam current below the thermal expansion line results in a solid liquid target being cooled by convection. This allowed for $Q_{\text{convection}}$ to be bound. For TS-1 $Q_{\text{convection}}$ was bound at $18 \mu A*22\text{MeV}$, TS-2 $Q_{\text{convection}}$ was bound at $22 \mu A*22\text{MeV}$, and TS-3 $Q_{\text{convection}}$ was bound at $35 \mu A*22\text{MeV}$. Using the same equations listed above, but now substituting in each bound for $Q_{\text{convection}}$ allowed for a better estimate of target performance. This is shown in Table 3.
Target | $Q_{\text{convection bound}}$ | Beam Current max Calculated (µA) | Beam Current max Actual (µA)
--- | --- | --- | ---
TS-1 | 18 µA*22MeV | 25.7 | 26.4
TS-2 | 22 µA*22MeV | 32.7 | 36.4
TS-3 | 35 µA*22MeV | 47.4 | Not reached

Table 3: Better estimate of target performance

Analyzing the experimental data in this way yields much closer results to the actual target performance of TS-1 and TS-2. This also predicts that TS-3 can withstand up to 47.4 micro amps. Looking at Figure 24 shows that at 45 micro amps TS-3 appears to be nowhere near its performance limit, so it would appear that this estimate is still conservative.

One final attempt was made at finding a better estimate of the convective heat transfer coefficient. For TS-1 and TS-2, the performance limit is known from Figures 21-23 (26.4 µA and 36.4 µA respectively). Therefore everything in the following equation is known or can be calculated, and $h_{\text{boiler}}$ at the performance limit can be extracted:

$$Q = h_{\text{boiler}} A_{\text{boiler}} (T_{\text{sat}} - T_s) + h_{\text{condenser}} A_{\text{condenser}} (T_{\text{sat}} - T_s)$$
The ratio of the actual convective heat transfer coefficient over the one calculated using the sight tube data to get a lower bound are similar for TS-1 and TS-2. Applying that to TS-3 yields a convective heat transfer coefficient at the performance limit of $7277.494 \times 1.4 = 10,188.5$ w/m$^2$ K. Using that as the convective heat transfer coefficient instead of the lower bound yields a max beam current input of 46.85 micro amps, which is close to 47.4 micro amps calculated in Table 3.

Also calculated was the void fraction for TS-1 and TS-2 at the performance limit. The results are shown in the following table:

<table>
<thead>
<tr>
<th>Target</th>
<th>Max Beam Current at Performance limit ($\mu$A)</th>
<th>Void Fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>TS-1</td>
<td>26.4</td>
<td>0.46</td>
</tr>
<tr>
<td>TS-2</td>
<td>36.4</td>
<td>0.47</td>
</tr>
</tbody>
</table>

Table 5: Void Fraction at Performance Limit
Pressure fluctuations within the target measured by the pressure transducer were recorded during the experiments. These occurred mainly at higher beam currents. Table 6 shows the pressure fluctuations in TS-2 for 7-10-01 data.

<table>
<thead>
<tr>
<th>Beam Current (µA)</th>
<th>Pressure (psi)</th>
</tr>
</thead>
<tbody>
<tr>
<td>18</td>
<td>524.2-527.8</td>
</tr>
<tr>
<td>20</td>
<td>522.8-528.9</td>
</tr>
<tr>
<td>22</td>
<td>521-531</td>
</tr>
<tr>
<td>24</td>
<td>520-528</td>
</tr>
<tr>
<td>26</td>
<td>520-528</td>
</tr>
<tr>
<td>28</td>
<td>520-528</td>
</tr>
<tr>
<td>30</td>
<td>514-540</td>
</tr>
<tr>
<td>30 (isolate target)</td>
<td>1100 broke target window</td>
</tr>
</tbody>
</table>

Table 6: TS-2 Pressure Fluctuation Data.

At 30 micro amps the target was isolated, meaning it was valved off from the helium supplying the overpressure. At steady state a thermosyphon should go to a pressure dictated by the cooling capacity of the system. If there is more than enough cooling capacity for that heat input, the pressure should drop and vice versa. In the case of TS-2, 30 micro amps was less than its performance limit of 36.4 micro amps, leading one to predict that there is plenty of cooling capacity and the pressure should drop. As shown in Table 6, the opposite happened. The pressure skyrocketed and broke the window.

Recent experiments performed on TS-3 on 5-20-02 showed similar behavior. TS-3 was
isolated at 5 micro amps, and the pressure oscillated between 200-500 psi. When isolated at 10 micro amps severe pressure oscillations were also observed. Isolating TS-3 at any higher beam currents proved to be too risky because of possible window failure. In the absence of the helium over pressure, the system is then unstable to minor perturbations in pressure. The Helium gas over pressure damps these perturbations and is therefore necessary for stable operation.

Due to the high cost of $^{18}$O enriched water, most of the experimental data was collected using normal water. One set of experiments was run on TS-2 using $^{18}$O enriched water to perform yield calculations. The target was irradiated for 70 minutes with 22 MeV protons at 19.54 micro amps of beam current. From Figure 2 in Chapter 2, 22 MeV protons in water produce a saturation yield of 333 mCi/$\mu$A. After draining and rinsing the target 2,193 mCi of $^{18}$F were produced. The fraction of the saturation yield one would expect from irradiating for only 70 minutes can be determined by:

$$fraction = 1 - e^{-\lambda t}$$

$$\lambda = \frac{\ln(2)}{110\text{ min}}$$

$$t = 70\text{ min}$$

$$fraction = 0.3567$$

$$\frac{2193\text{ mCi}}{0.3567 \ast 19.54\mu A} = 314.7\frac{mCi}{\mu A}$$

Which is then corrected for the enrichment of the $^{18}$O water:

$$\frac{314.7\frac{mCi}{\mu A}}{0.95} = 331\frac{mCi}{\mu A}$$
Based on the experimental data one would expect to get 331 mCi/µA if the target was irradiated for an infinite period of time. This is very close to the theoretical yield of 333 mCi/µA from Figure 2.
6 Sources of Error

This section serves as a general discussion of the sources of error. In terms of the model, the main sources of error arise in the use of the various correlations used to calculate the convective heat transfer coefficient on the secondary side, the friction and forms losses on the secondary side, fin efficiency, and the correlations used to describe the wavy laminar film condensate. Every effort was made to match the model to the experimental targets in terms of conduction thickness, materials, and secondary side cooling capacity.

The sources of error for the experimental data are numerous. Since it is impossible to be in the cyclotron vault while the target is operating, a TV camera was used to monitor the water expelled from the target. A ruler marked off in centimeters was placed beside the sight tube so that the height change due to the volume of water displaced could be recorded via the TV camera. The poor resolution on the TV camera allowed the data collector to only record the height change in the sight tube in 0.5 cm increments. The calibration of the thermocouples and pressure transducer contain a certain amount of error. Any of the readouts on the cyclotron control panel, such as beam current, contain some error due to the fact that they are analog dials. The cyclotron produces 27 MeV protons, which are attenuated down to 22 MeV, via the degrader, that introduces a source of error in the incident proton energy. The Havar target window also attenuates the beam a small amount. The target window is also cooled, but due to the low thermal conductivity of Havar foil, its contribution to the heat transfer was neglected. When reviewing the experimental data it is important to note the high swing and the low
swing in terms of the volume of water expelled from the target. At higher beam currents the swing is larger, this allows hot target water to get mixed with the cooler water in the sight tube. This can add some error to the experimental data. Also any cooling due to radiation to the surrounding air was not included. There are also errors in the measurement of secondary side mass flow rate used in the calculations. Figure 27 for TS-3 shows the discrepancy between the amount of heat input into the target \( Q = \mu A * \) MeV) and the amount of heat removed based on thermocouple data across the various parts of the target as shown in Chapter 3 \( Q = \dot{m} C_p \Delta T \). This could be due to several factors; the most obvious is an error in beam current read off the cyclotron panel. For example the data collector might think that the beam current is at 20 micro amps when in fact it may be higher or lower than 20 micro amps. Another error could be with the thermocouples or measured mass flow rate.

It has been recently discovered that the valve at the bottom of the target used when isolating the target from the helium over pressure restricts the flow of water in and out of the target. After removing this valve such that there was a direct port from the sight tube into the bottom of the target, pressure fluctuations within the target became much smaller and increased target performance has been observed.
Figure 27: Heat Input vs. Heat Removed for TS-3
7 Summary and Conclusions

The goal of this project was to investigate the feasibility of thermosyphon targets for the increased production of $^{18}$F in cyclotrons. A more efficient target design would cut down the cost of PET as a form of medical imaging.

A computational model was developed to simulate the performance of prototypical thermosyphons. The model showed that a thermosyphon target was in fact feasible and would out perform the targets currently used. Current targets operate at around 20 micro amps of beam current. One of the simulated models showed that the beam current could be doubled to 40 micro amps, which would double the $^{18}$F yield. The only trade off was increasing the target volume by 50% to stop all the protons. The model also showed that the target would need to have a gas over pressure to avoid start up problems.

Based on the model results, three separate thermosyphon targets were designed, built, and tested, each having an increase in condenser volume/heat transfer surface area over the previous version. By increasing the condenser volume 80% in TS-1→TS-2, the resulting target performance increased 37.9%. The performance limit for TS-3 was not reached, so the data from TS-1 and TS-2 were used to predict a max performance for TS-3 of 47 $\mu$A. However, this seems to be on the conservative side since TS-3 operates at 45 $\mu$A and appears to be well below its maximum capabilities at that beam current.

The experimental data also showed that is was necessary to maintain a helium gas over pressure at all times while the target is operating. Eliminating the gas over pressure
by isolating the target, resulted in severe internal pressure oscillations which can cause catastrophic failure by rupturing the target window.

The yield experiments showed that the target performed as expected. The increase in $^{18}\text{F}$ yield is directly proportional to beam current. Once again, the thermosyphon targets out perform any of the targets currently in use. This will help to lower the cost and increase the efficiency of the production of radioisotopes used in medical centers all over the country. Companies that mass-produce radiopharmaceuticals would also benefit from a target that could withstand higher beam current, equating with higher yields. The decrease in cost of radionuclide production could be passed down to the individual patients by lowering the cost of a PET scan.

Some suggestions for future work include, exploring the pumped re-circulating target. Developing a pump with a low dead volume that can produce the mass flow rates necessary to properly cool the target shows great promise as another alternative to $^{18}\text{F}$ water target design. Another modification would be in developing a sidestream system to capture $^{18}\text{F}$ fluoride ions on-line and remove the trapped activity without having to drain the target water. This would apply mainly to a pumped re-circulating type target, but it would allow tailoring the quantity of the recovered activity based on the amount of doses required. Another suggestion for future work would be to find a cyclotron that can output more than 1 kilowatt of beam power so that the true performance limitation on TS-3 can be experimentally validated. Last but not least, with the recent development in improved target performance by removing the valve at the bottom of the thermosyphon targets, it would be beneficial to perform more experiments without the valve for all three targets.
References


Wieland, Bruce W., et.al. Self-regulating thermosyphon water target for the production of F-18 Fluoride at proton beam power of one kw and beyond. Accepted for publication in the Proceedings of the Ninth International Workshop on Targetry and Target Chemistry. Turku, Finland. May 23-25, 2002.