Production of ^{18}F by proton bombardment of ^{18}O-enriched water targets under saturation conditions

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Abstract. Single-cavity, boiling-water targets are commonly used for the production of the PET radionuclide ^{18}F, utilizing the ^{18}O(p,n)^{18}F reaction on ^{18}O-enriched water. At iThemba LABS, two such target systems are in routine use. Monte Carlo simulations were performed by means of the radiation transport code MCNPX in order to investigate alternative target configurations with the aim to improve the production efficiency with a degraded beam from the separated-sector cyclotron. A configuration is presented where most of the energy degradation of a 66 MeV primary proton beam down to the recommended energy of 18 MeV is achieved in the cooling water. With the improved target system, 80% of the incident beam reaches the ^{18}O-water cavity, in contrast to the 25% typically obtained with the initial target configuration. A simple model is also presented that reproduces the pressure versus beam-current characteristics of several different closed-cavity boiling-water targets surprisingly well. An assumption of the model is that the majority of the system operates at saturation conditions as given by the standard steam tables. It has only one free parameter, namely an overall heat-transfer coefficient, which is also called an *h*-factor. This parameter can be determined by adjusting its value until a good agreement with measured data is obtained. In this way, values of *h* have been determined for a number of different targets with volumes ranging from 0.5 cm^3 to 5 cm^3. Even though the target sizes differ by an order of magnitude, the *h*-factor remains nearly constant. This gives the model predictive power.

1. Introduction

The radionuclide ^{18}F is extensively utilized in positron emission tomography (PET). Its use is not only limited to oncology but also important in fields such as neurology, cardiology and pharmacology. In addition, non-medical applications of PET are becoming increasingly important. A group at the University of Cape Town, for example, uses PET technology for positron emission particle tracking (PEPT) [1]. Fluorine-18 is routinely produced at iThemba LABS, mainly as a service to the South African medical community, utilizing the ^{18}O(p,n)^{18}F reaction on ^{18}O-enriched water targets.

Currently, iThemba LABS utilizes beams from both the separated sector cyclotron (SSC) and a dedicated 11 MeV PET cyclotron for ^{18}F production. The target system on the SSC became operational in 2005 and production with the PET cyclotron commenced in 2012. While the bulk of the production is nowadays done with the smaller PET cyclotron and its targetry, regular productions are still being scheduled on the SSC in order to keep both systems in good operational condition. This is done to prevent disruptions in the ^{18}F production programme (e.g. during shutdowns for accelerator maintenance or when unforeseen breakdowns occur) ensuring a continuous and reliable supply.

The main aim of this paper is to discuss two aspects of the targetry. The first deals with problems associated with radial beam spread, which is particularly severe when utilizing a degraded beam from the SSC to achieve the energies required for ^{18}F production. The second concerns the observation that saturation conditions are maintained inside the target cavities during bombardment. This, we believe, is relevant not only to water targets but also to other closed-cavity liquid targets. The focus is therefore on the SSC water targetry but the lessons learnt are relevant to other target systems as well, in particular where the incident beam has to be degraded to match the requirements of the target system.

The radial beam spread, which affects the ratio of protons penetrating the water cavity versus those lost upstream on degrader and collimator units, were studied by means of extensive Monte Carlo radiation transport calculations as well as separate beam current measurements on various electrically isolated targetry components. Simulations with the code MCNPX [2] assisted in improving the target geometry and significantly enhancing the efficiency of beam utilization, which will be shown below.

The conditions inside boiling-water cavities during bombardment were studied by measuring their pressure versus beam current characteristics. It is shown that a very simple model reproduces these characteristics quite well. In addition to the results for the SSC target in use at iThemba LABS, calculations are also presented for two other target systems described in the literature, giving results for cavity sizes ranging from 0.5 cm^3 to 5 cm^3.

2. Targetry and Monte Carlo simulations

A commercial stand-alone target (SAT) and processing system for activated [^{18}O]H_2O was purchased for use on the SSC. There was, however, a compatibility issue concerning the proton beam energy that had to be resolved. A proton beam of 18 MeV was specified for the SAT system but the routine radionuclide production programme was based on proton beams of 66 MeV, shared with neutron therapy. The cyclotron schedule could not accommodate energy changes between 66 MeV and 18 MeV several times per week (one energy change typically takes between 4 and 6 hours). Thus, the beam energy had to be degraded from 66 MeV down to an average of 18 MeV.

A simplified schematic diagram of the initial target configuration is shown in figure 1. The target cavity for [^{18}O]H_2O is cylindrical and elongated with a volume of 2.3 mL. The beam is stopped in the target water. A helium-cooled, double-foil Havar window is employed to contain the target water at the beam entrance side of the cavity. Just upstream of the cavity is a conical collimator, supplied by the commercial manufacturer. Its function is to stop scattered beam particles emerging from the aluminium degrader on a relatively large water-cooled surface and to allow only a well-defined degraded beam into the target cavity. Note that, for the sake of clarity, figure 1 does not show certain details such as the cooling channels of the degrader and collimator units.

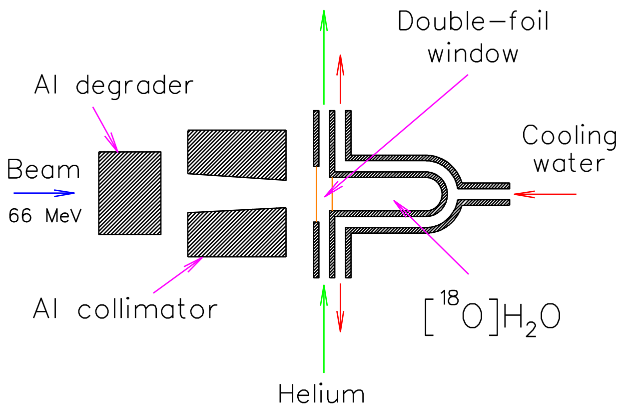

Figure 1. A simplified schematic diagram of the initial configuration of the ^{18}O–enriched water target for ^{18}F production with a 66 MeV primary proton beam delivered by the SSC.

The performance of this beautifully constructed target was rather disappointing, however, as it turned out to be impossible to get more than 25% of the protons into the Nb target cavity under well-optimized beam conditions, i.e. three quarters of the beam was stopped by the collimator. Simulations of the beam and targetry were subsequently performed by means of the Monte Carlo code MCNPX in an attempt to better understand this behavior and to improve the targetry.

MCNPX Version 2.7.0 [2] was used for the Monte Carlo simulations. The beam was modelled with realistic values of the emittance in two orthogonal directions normal to the beam axis downstream of the last focusing quadrupole (ε_x = 14 π-mm-mrad and ε_y = 5 π-mm-mrad), drift length upstream of the target (150 cm) and beam spot at the target position (2 to 3 mm FWHM). It was found that the beam profile at the entrance to the target cavity was rather insensitive to the exact choice of beam parameters upstream of the degrader as long as the values remained realistic. By means of the Monte Carlo simulations, various alternative target configurations were investigated as well as the effect of small changes in the degraded beam energy. Concerning the latter, it is interesting to look at the relevant integral yield curve [3] for the production of ^{18}F but expressed as the yield per unit energy deposited by the beam in the target water. This is shown in figure 2. A maximum is evident at 14 MeV. In principle, this should be the optimum incident energy from a thermal point of view because the challenge is to remove a significant amount of heat from quite a small volume with very limited heat-transfer surfaces. As the 66 MeV beam had to be downgraded by a rather a large amount, it was eventually decided to retain the nominal energy of 18 MeV as specified by the commercial supplier.

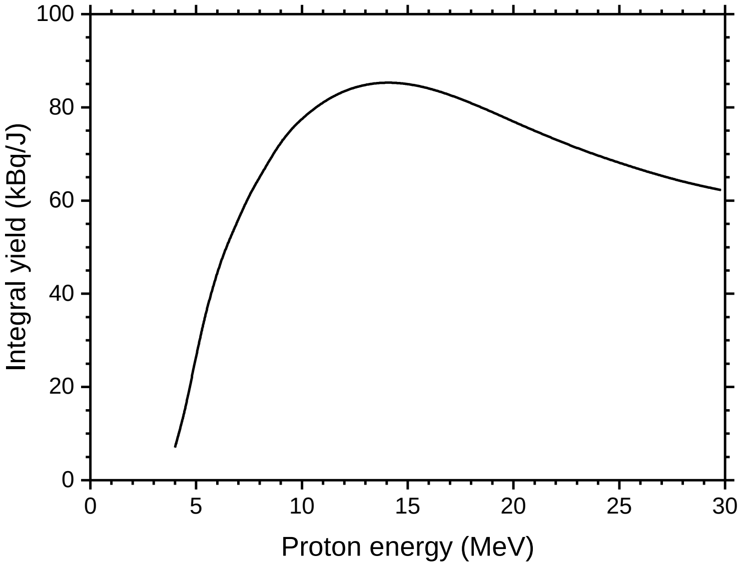

Figure 2. Integral yield per unit energy deposited by the beam in the water cavity, plotted as a function of incident energy. Note that the beam is stopped inside the target water.

A full account of all the simulations performed will not be given here. Various possibilities exist to improve the configuration of figure 1, in particular by reducing the length of the elongated collimator. Finally, a configuration was adopted where most of the beam degrading is done in the cooling water (at a high flow rate of 30 L/min) in close proximity to the target cavity. Also, the collimator was reduced considerably in length and machined to be an integral part of a single degrader/collimator unit. The final configuration is shown schematically in figure 3. The collimator has a 10 mm diameter and that of the target cavity is 11.5 mm. Using the cooling water for degrading purposes has an additional advantage, namely that a large fraction of residual radioactivity induced in the degrader can be transported away with the water to an ion exchanger located in a convenient location outside the vault. This helps to reduce the radiation dose to staff doing routine target maintenance in the vault.

Figure 4 shows the fraction of the incident beam at a penetration depth corresponding to the entrance of the target cavity, as a function of radius, obtained by means of MCNPX simulations. In these particular simulations, the degraders were retained but the collimators were removed in order to study the overall radial beam spread. In both cases, 1 000 000 primary protons were tracked. Since no secondary photons and neutrons needed to be tracked, the execution time of the code was quite short (< 30 min) on a standard high-end PC, in spite of creating a large number of event histories. The improvement in beam efficiency of the modified target configuration is evident. Excellent agreement between Monte Carlo simulations and results from beam current measurements on the major targetry components was obtained. In a typical production run, about 20% of the beam is usually measured on the combined degrader/collimator unit and 80% on the Nb target cavity. Also, by utilizing a mesh tally on the inside surface of the Nb cavity, it was possible to determine that > 99% of the collimated proton beam entering the cavity stopped in the target water without escaping to the Nb wall. (Concerning figure 4, note that the inner radius of the cylindrical part of the target cavity is 5.75 mm).

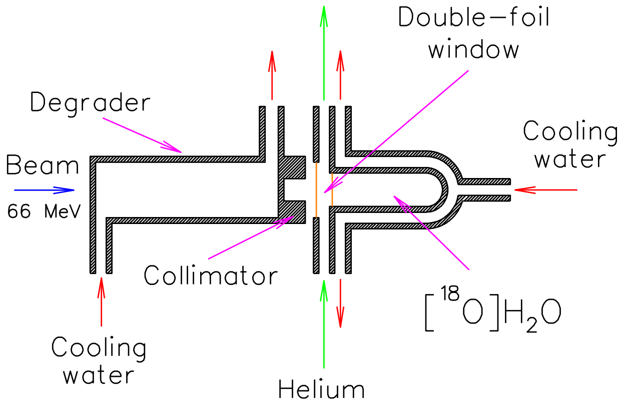

Figure 3. Simplified schematic diagram of the final configuration of the ^{18}O–enriched water target for ^{18}F production with a 66 MeV primary proton beam delivered by the SSC.

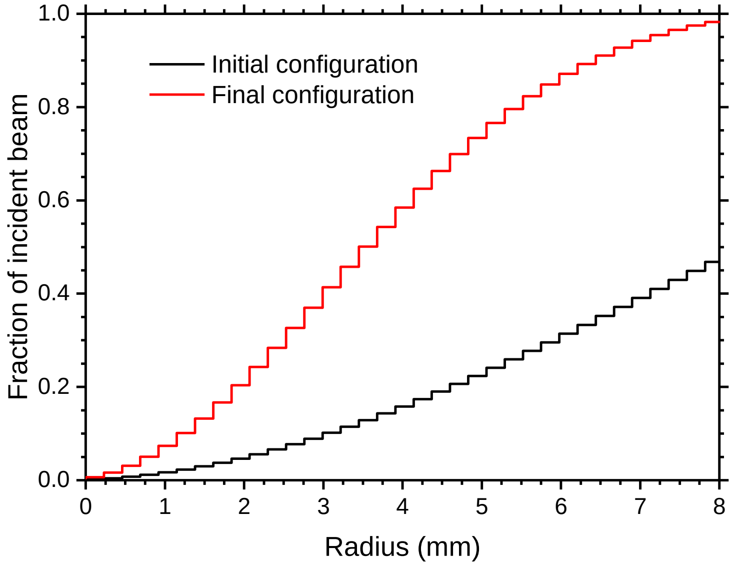

Figure 4. MCNPX predictions of the fraction of the incident beam reaching the entrance plane of the ^{18}O–enriched water target cavity, plotted as a function of the radius around the beam axis. Here the mesh tally was placed on the plane of the inner Havar foil and the degraders were removed.

3. Boiling and saturation conditions

At iThemba LABS, measurements of the target pressure versus the beam current were performed on a 2.3 mL single-cavity, boiling-water target. A calibrated piezoresistive transducer with a response time of about 1 ms was used for the pressure measurements. An EG&G CD 1010 current digitizer was used to measure current on the target cavity (Nb insert) and a Keithley 485 electrometer to measure current on the degrader/collimator. These values were logged on a PC using the LabVIEW code [4] for data acquisition. The results are shown in figure 5. Also shown in figure 5 are published measured results for a much smaller cylindrical target cavity with a volume of 0.5 mL, operating at a proton beam energy of 13 MeV [5], as well as for a larger conically-shaped cavity with a volume of 5 mL [6]. The measurements are compared with calculations based on a simple model, described below.

The model assumes that the bulk of the ^{18}O-enriched target water has a constant temperature, which is the same as the inner wall temperature of the cavity, T_w. It is further assumed that saturation conditions are being maintained during bombardment. Both assumptions are being justified by referring to the work of Alvord et al. [7]. These authors pointed out that elevated pressures and temperatures in excess of saturation conditions may exist in a water target during bombardment. However, as long as the rate of condensation matches the rate of vaporization, the bulk of the system should remain at saturation conditions. Superheated regions are therefore likely to form but also likely to rapidly disappear, typically on the scale of a few milliseconds for such small volumes. The boiling process is generally quite complex, enhanced by radiation-induced nucleation. This, however, introduces fast mixing mechanisms in the water volume. Large temperature gradients can therefore only briefly exist under these very ebullient boiling conditions.

A second simplification is to neglect the temperature difference across the target chamber wall, which is only justified if the wall is thin (e.g. 1 mm thick Nb). A further assumption is that a single, overall convective heat-transfer coefficient can be applied, which is considered to be constant over the entire water-cooled surface. Here we take as the heat-transfer surface the inner cavity surface opposite the outer surface of the Nb insert directly in contact with the cooling water. The energy balance between the beam heating and the convection heat transfer (Newton's law of cooling) is given by

$$I_b \Delta E = hA(T_w - T_0), \tag{1}$$

where I_b is the beam intensity, ΔE is the energy window of the target (which is the same as the incident energy if the beam is stopped in the target water), h is the convective heat-transfer coefficient, A is the inner cavity surface through which the heat has to be transferred from the target-water volume to the cooling water, and T_0 is the cooling-water temperature.

The saturated vapor pressure versus temperature of water is a characteristic curve, given by the steam tables [8]. It can be written as

$$P = f(T) \quad \text{or} \quad T = f^{-1}(P). \tag{2}$$

Assuming the bulk of the system at saturation conditions, substitution of (1) into (2) gives

$$P = f(T_w) = f\left[\left(\frac{\Delta E}{hA}\right) I_b + T_0\right]. \tag{3}$$

The function f can be conveniently represented by a polynomial fitted through the tabulated values. The only unknown in equation (3) is the overall convective heat-transfer coefficient, h. Our approach was to adjust h until a good visual fit with a set of measured data was obtained. The results are shown in figure 5 and the values of h as well as other relevant quantities are presented in table 1.

A very good agreement between the measurements and calculations is evident in figure 5 for the 2.3 mL and 5 mL targets. In the case of the 0.5 mL target of reference [5], only one measured value was available, nevertheless, the extracted value of h is in good quantitative agreement with the other two cases, as shown in table 1. The extracted h-factors are indeed remarkably similar, even though the target volumes differ by an order of magnitude.

It is evident from equation (3) that the target performance in terms of the maximum operational beam current for a given beam energy and a maximum pressure (and hence the corresponding yield) can be improved by increasing the heat-transfer surface (A), the heat-transfer coefficient (h) and by reducing the cooling-water temperature (T_0). At a typical operational pressure of 30 bar, the saturated water temperature is 234 °C, thus by using chilled cooling water at, say, 15 °C only a small performance increase of about 7% will be obtained compared to using cooling water at an average temperature of 30 °C (i.e. room temperature). It will also be difficult to significantly increase the overall heat-transfer coefficient by increasing the cooling-water flow rate. High flow rates are normally employed and care must be taken as further increases may cause adverse effects, e.g. too large forces on components and cavitation with damage potential. Thus, the only viable way to significantly increase target performance is to increase the surface from which the heat can be removed. This leads to an increase in the target volume. As the enriched ^{18}O-water is quite expensive, some judgment call needs to be made, depending on operational needs. This is not the same for all producers. For this reason, commercial suppliers nowadays offer their clients a range of target sizes with volumes typically ranging between 2 and 10 cm^3.

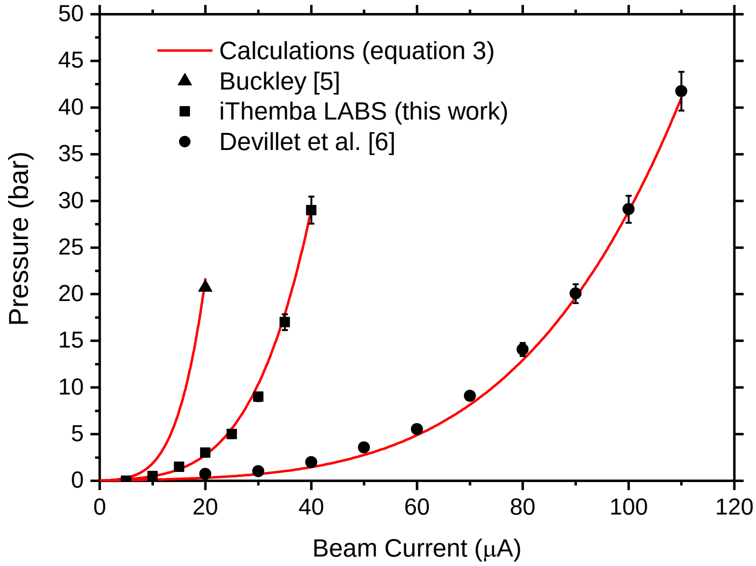

Figure 5. Pressure versus beam current for several ^{18}O-enriched water targets, as indicated. Error bars are shown where they exceed the symbol size.

Table 1. Values extracted for the overall heat-transfer coefficient (h) and other relevant quantities of the targets investigated.

Target	Energy window [ΔE] (MeV)	Cavity volume (cm^3)	Heat-transfer surface [A] (cm^2)	Cooling-water temperature [T_0] (°C)	h (W cm^{-2} °C^{-1})
Reference [5]	13	0.5	2.70	20	0.49
This work	18	2.3	6.56	30	0.54
Reference [6]	18	5.0	18.59	30	0.48

4. Other considerations

Single-cavity, boiling-water targets are vulnerable to sudden increases in beam current as bulk vaporization can lead to destructive pressure spikes. An interlock condition set on the target pressure is therefore essential. The calibrated piezoresistive transducer used for the pressure measurements at iThemba LABS has a rapid response time of about 1 ms and is a necessary component of the beam interlock system.

As the primary beam energy from the SSC is 66 MeV, the primary activation of the targetry as well as the secondary neutron activation of the immediate vicinity is far more severe than what is typical for a PET facility based on a small medical cyclotron. The target is therefore well shielded during bombardment, similar to all the other radionuclide production targets on SSC beamlines at iThemba LABS. Figure 6 shows the target and a dedicated bombardment station for semi-permanent targets.

Figure 6. (a) The shielding of the bombardment station in an open position, showing the location of the target on the beamline. **(b)** The bombardment station in beam-ready state.

5. Summary and conclusion

The SSC target system described above has now been in routine use for the production of ^{18}F at iThemba LABS for several years. It operates in the boiling regime with the majority of the system at saturation conditions. By keeping it in routine use on the SSC conjointly with a new 11 MeV PET cyclotron and its dedicated targetry, a year-round supply of ^{18}F can be maintained.

The Monte Carlo radiation transport code MCNPX has proved to be an extremely useful tool in gaining an understanding on target systems under irradiation conditions. It is possible to simulate the proton beam very precisely with the latest version of the code (Version 2.7.0) and the radial beam spread as a function of penetration depth is found to be in excellent agreement with experimental observations. Simulations of the 66 MeV proton beam from the SSC and the entire ^{18}O-enriched water-target system allowed us to investigate alternative configurations of the degrader and collimator. Finally, it proved to be advantageous to combine them in a single unit and use cooling water for most of the required energy degradation (down to an average energy of 18 MeV).

A study of the pressure inside a water target versus the applied beam current indicated that saturation conditions are indeed maintained. This made it possible to reproduce the pressure versus beam-current characteristics of a number of different targets, under their respective operational conditions, with a rather simple model. The values extracted for the overall heat-transfer coefficient, which is the only free parameter of the model, are quite similar in magnitude for a number of target systems with volumes differing by an order of magnitude. This is interpreted as as evidence that the model has predictive power. It may be useful to apply to other liquid targets as well.

The monitoring of the pressure inside a closed-cavity, boiling-water target, by means of a transducer with a fast response time, is strongly advisable for inclusion in the beam interlock system. If an upper pressure limit is exceeded, the bombardment should terminate immediately. Lastly, the use of a degraded beam for the purpose of ^{18}F production, as implemented on the SSC of iThemba LABS, requires local shielding in close proximity to the target. This is not a requirement for a production facility based on a dedicated low-energy PET cyclotron.

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