A saturation boiling model for an elongated water target operating at a high pressure during ^{18}F production bombardments

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Abstract. An elongated boiling boiling water target for ^{18}F production is modelled by assuming the entire water volume to be at a constant temperature for a constant beam current and that a single overall convection heat-transfer coefficient applies to the target cooling. These approximations may seem severe for a system with complex boiling behaviour, however, it is justified due to the presence of fast mixing mechanisms in a relatively small liquid volume. It is shown that the measured pressure versus beam current curve of the target can be reproduced by such a simple model, assuming that the majority of the system operates at saturation conditions as given by the standard steam tables. Integral yields are also presented and the optimum incident beam energy is determined based on a thermal load point of view.

1. Introduction

The radionuclide ^{18}F is extensively utilized in positron emission tomography (PET). It is routinely produced at iThemba LABS as a service to the medical community in South Africa, utilizing the ^{18}O(p,n)^{18}F reaction on an enriched water target. Its use is not only limited to oncology but it also finds application 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, is using PET technology for positron emission particle tracking (PEPT) [1].

In 2005, iThemba LABS was tasked to produce and deliver ^{18}F in the Western Cape, utilizing beams from the separated sector cyclotron (SSC) as an interim measure until such time when a dedicated PET cyclotron facility would be established in this region. A commercial stand-alone target (SAT) and processing system for activated [^{18}O]H_2O was selected for this purpose, however, there was a compatibility problem concerning the proton beam energy which had to be solved. The SAT system required a proton beam of 18 MeV 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 a week (one energy change typically takes between 4 and 6 hours). Thus, the 66 MeV had to be degraded to an average beam energy of 18 MeV. Initially, a water-cooled aluminium degrader located upstream of the water target was used for this purpose, however, the radial beam spread was so severe that only about 25% of the beam reached the actual target cavity. A Monte Carlo simulation of the beam, degrader and target geometry quantitatively reproduced this low bombardment efficiency [2]. The problem could only be solved by completely rebuilding the SAT system, utilizing the cooling water for most of the energy degradation in close proximity to the target cavity. Figure 1 (a) shows the modified target while (b) depicts a simplified schematic diagram of the in-beam components. Following the degrader is a collimator to reshape the beam, before passing through a double-foil helium-cooled window into the $[^{18}O]H_2O$ target cavity. This modification immediately improved the beam entering the target cavity from 25% to 80%.

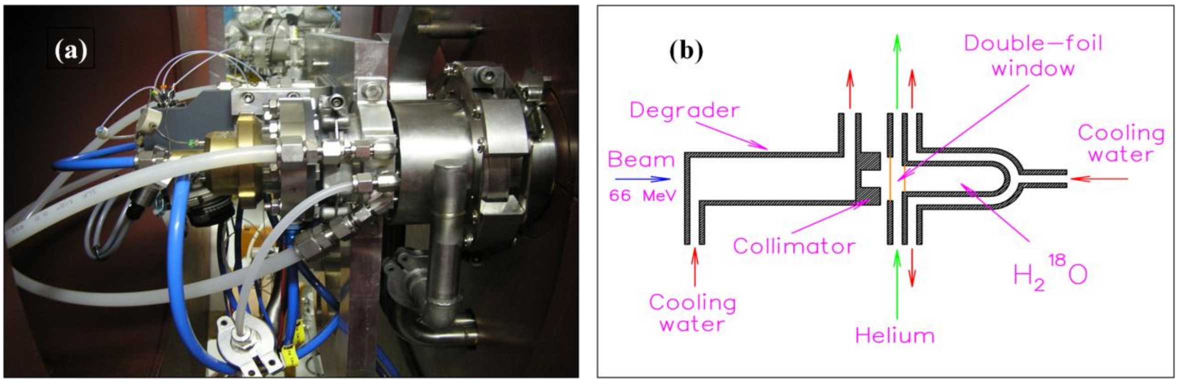

Figure 1. (a) The enriched ^{18}O –water target in use at iThemba LABS for the routine production of ^{18}F. **(b)** A simplified schematic diagram of the target system.

As the primary beam energy 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. The target is therefore well shielded during bombardment, similar to all the other radionuclide production targets at iThemba LABS. Figure 2 (a) shows the target mounted in the dedicated bombardment station (the radiation shield is in an open position) while (b) shows the station ready for bombardment (i.e. a closed shield).

Figure 2. (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.

Second-generation targets, such as the one shown in figure 1, operate in the boiling regime. This is in contrast to first-generation targets which had much smaller cavities (due to the scarcity of enriched water at that time) and which were typically supplied with an external high pressure (usually helium) to suppress boiling. With the boiling targets an increase in yield by an order of magnitude could be achieved. The elongated shape of the target cavity is important as it prevents burn-through, a problem which sometimes happened in first-generation targets which were typically much thinner and thus would not stop the proton beam when a significant amount of steam bubbles formed. There are still some debates and open questions, however, in particular whether a second cavity for condensation cooling (i.e. a condenser) separated from the target cavity could enhance the target performance (see e.g. [3]). Note that the target of figure 1 has only a single, elongated cavity (i.e. no condenser or reflux cavity). It is certainly true that in large systems (e.g. boiling nuclear reactors) a condenser stage can be used with great efficacy. On the scale of a small target which contains only circa 2 – 5 mL of violently boiling water, however, the clean separation of the steam from the liquid and the subsequent return of the condensate to the target cavity are difficult to successfully implement. In contrast, targets with single cavities are simpler (see e.g. [4]) and seem to generally work well, therefore the type presently preferred at iThemba LABS. There are also other novel ideas e.g. thermosyphon [5] and recirculating targets [6], the development and progress of which one should certainly monitor as they may become important in future.

2. Target operational behaviour

At iThemba LABS, measurements of the target pressure versus the beam current were performed on a 2 mL single-cavity boiling target. A calibrated piezoresistive transducer with a response time of about 1 ms is a component of the beam interlock system and was used for the pressure measurements. An EG&G CD 1010 current digitizer was used to measure the beam current. These values were logged on a PC using the LabVIEW code for data acquisition. The results are shown in figure 3 (a). An attempt to interpret the results with a simple model was also made, as explained below.

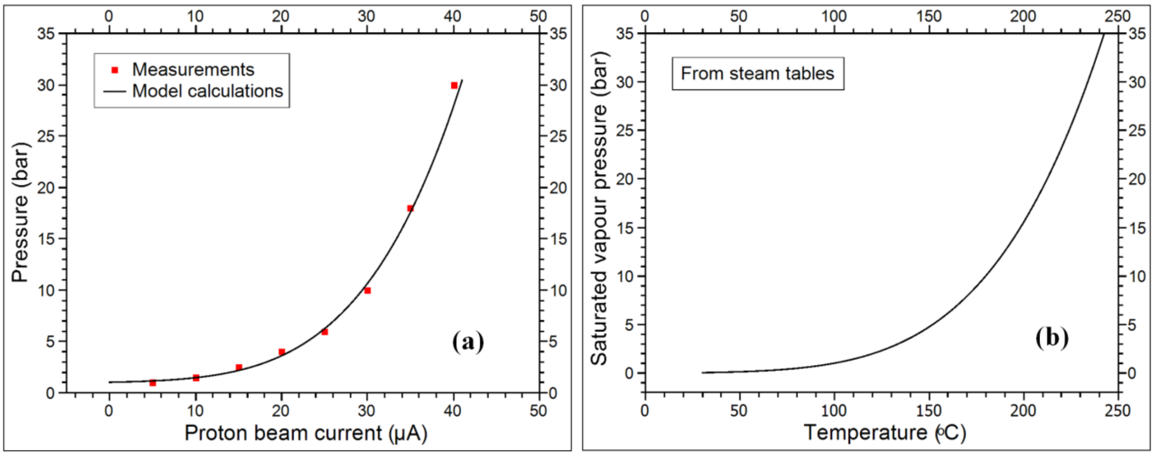

Figure 3. (a) Measurements and a calculation of the pressure inside the water target versus the proton beam current (see text). **(b)** Characteristic curve of the vapour pressure versus temperature of water under saturation conditions.

In a recent study by Alvord et al. [7], it was pointed out that elevated pressures and temperatures in excess of the saturation conditions may exist during bombardment, however, as long as the rate of condensation matches the rate of vaporization the bulk of the system should remain at saturation. Bulk boiling, enhanced by radiation-induced nucleation, provides a rapid mixing mechanism within the small target cavity. Superheated regions are therefore likely to form but also likely to disappear rapidly (on the scale of a few milliseconds). One concern may be unstable behaviour, e.g. rapid cycles of bulk vaporization and condensation, which my lead to large pressure fluctuations. This has been observed in boilers of various kinds and should be avoided in targets as it may destroy the thin foil entrance window. Due to the presence of fast mixing, our simple model assumes the target water to have a constant temperature. A second simplification is to neglect the temperature difference across the target chamber wall (1 mm thick niobium) which is justified because this wall is quite thin. A further assumption is that a single convection heat-transfer coefficient applies which is constant over the entire water-cooled surface. By writing down the energy balance between the beam heating and the convection cooling (Newton's law of cooling) and rearranging terms, one gets the following equation:

$$T_w = \left(\frac{\Delta E}{hA}\right) I_b - T_0,$$

where T_w is the target-water temperature, ΔE is the beam energy window, A is the water-cooled surface, h is the convection heat-transfer coefficient, I_b is the beam current and T_0 is the bulk temperature of the cooling water. The only unknown is the convection heat-transfer coefficient, however, h^{-1} only acts as a scaling factor and does not affect the shape of the T_w versus I_b curve. Assuming saturation conditions, the relationship between the pressure, P, of the target water and its temperature, T_w, is uniquely known from the steam tables [8]. Figure 3 (a) shows a plot of P versus I_b, where h has been adjusted until a good fit with the measurements was obtained. Figure 3 (b) shows the characteristic curve of pressure versus temperature for water at saturation. The good agreement in shape is taken as evidence that the majority of the system operates at saturation conditions.

3. Integral yield

Production yields obtained with the target system described above are in good agreement with expectations based on evaluated integral yield data [9], shown in figure 4(a), which is further evidence that bulk vaporization does not cause negative effects such as burn-through of the beam. In figure 4 (b) the same information is shown but plotted in units of kBq/J (i.e. the yield per unit energy deposited by the beam in the target water.) This curve shows a maximum at 14 MeV, which would be the optimum incident energy from a thermal load point of view. It is interesting to note that most commercial PET cyclotrons operate in the proton energy region 11–18 MeV.

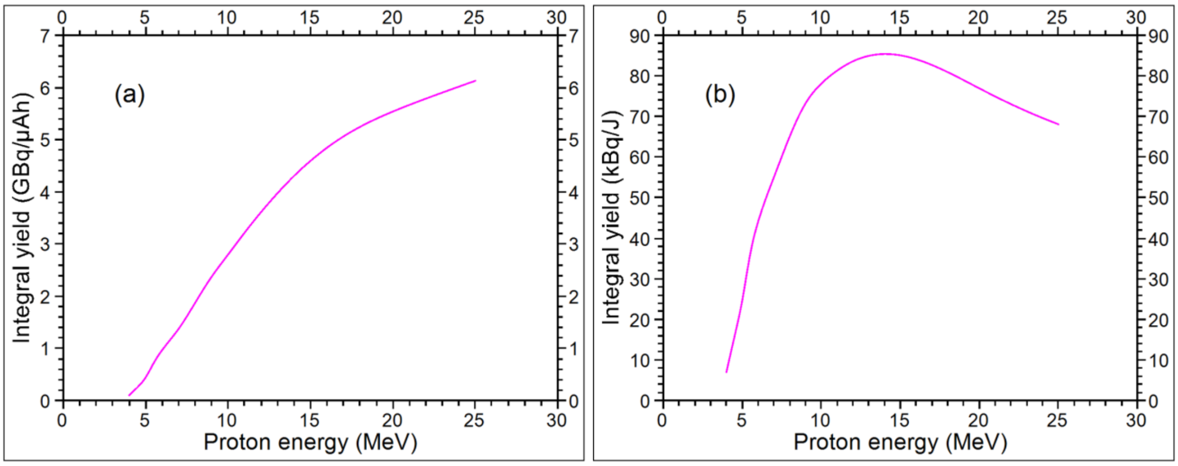

Figure 4. (a) Integral yield curve for the production of ^{18}F in the bombardment of enriched ^{18}O-water with protons. **(b)** Integral yield per unit energy deposited by the beam in the water target.

4. Summary and conclusion

The 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. No instabilities due to superheating have been observed.

The use of a 66 MeV primary proton beam delivered by the separated sector cyclotron (SSC) for this purpose is not ideal as the beam energy is much higher than required. Beam time from the SSC is also at a premium. In addition, the use of a degraded beam leads to higher residual activation, more radioactive waste and more radiation damage to targetry components. This arrangement was never envisaged to be permanent, however, but rather to stimulate the establishment and use of PET in South Africa. It was foreseen that a dedicated, small PET cyclotron would eventually be established in the Western Cape.

A commercial 11 MeV PET cyclotron is currently being installed at iThemba LABS and is expected to take over all ^{18}F productions by about September 2012. This will free some much needed beam time on the SSC (about 12 hours of the 54 hours per week allocated for radionuclide production) that will be used for the production of other radionuclides such as the relatively long-lived isotopes ^{22}Na, ^{68}Ge and ^{82}Sr which remain in high demand. The existing water target system will, however, be retained as a backup facility. The year-round provision of ^{18}F will thus be secured.

References

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