

Reliability Study of the Liquid Target Chamber for ^{18}F Production at the BATAN's Cyclotron Facilities

I. Kambali*, T. Heryanto, Rajiman, S. Ichwan

Center for Radioisotopes and Radiopharmaceuticals, National Nuclear Energy Agency
Puspiptek Serpong, Tangerang - 15314, Indonesia

ARTICLE INFO

Article history:

Received 25 June 2010

Received in Revised form 20 March 2011

Accepted 08 April 2011

Keywords:

Reliability study

Liquid target chamber

^{18}F production

Proton irradiation

Cyclotron

^{18}O -enriched water target

Heat transfer coefficient

ABSTRACT

The liquid target chamber for ^{18}F production at the Cyclotron Division, Centre for Radioisotopes and Radiopharmaceuticals (PRR) of the National Nuclear Energy Agency of Indonesia (BATAN) has been analysed for its reliability in enduring high pressures and heat transfer requirements during proton beam bombardment as well as the recommended irradiation parameters for effective ^{18}F production. The target chamber was subject to house the ^{18}O -enriched water bombarded with high energy proton beam to produce ^{18}F . A range of SRIM-computer simulations have also been conducted to calculate the ranges of several energetic proton beams (of up to 20 MeV) into pure water target. A study of radioactive impurities which might be produced from the proton-irradiated chamber's materials was also included based on some references. Due to concern over the heat produced during target irradiation, a heat transfer analysis - particularly for the target's cavity - was also included in the presented studies to obtain a brief preliminary calculation of the heating impacts prior to irradiation tests. The calculation was performed for various proton beam currents and energies of up to 30 μA and 20 MeV respectively. It was found that the chamber was reliable for production of ^{18}F from proton irradiated- ^{18}O enriched-water target by maintaining the chamber's pressure of up to 3.6 bar if the proton beam current was kept below 16 μA for all energies or the proton beam energy was kept to or below 10 MeV for any employed beam currents. The overall heat transfer coefficient was also found to depend on the power deposited into the water target.

© 2011 Atom Indonesia. All rights reserved

INTRODUCTION

Radionuclide ^{18}F has been widely used for Positron Emission Tomography (PET), particularly in various overseas hospitals for cancer scanning and mapping [1,2,3]. The positron-emitting radionuclide can be produced by bombarding a certain target (^{18}O -enriched water or Ne gas) with a sufficiently high energy particle such as proton and deuteron accelerated by a cyclotron or other typical accelerators. The most common nuclear reactions for these purposes are $^{18}\text{O}(p,n)^{18}\text{F}$ [3,4] and $^{nat}\text{Ne}(d,x)^{18}\text{F}$ [4,5].

Production of such a radionuclide requires a deep understanding and knowledge of threshold energy, nuclear cross-section data, target chamber requirements, as well as heat transfer process in the chamber during irradiation. Furthermore, it is also essential to deeply understand the knowledge of radiation protection, particularly from the radionuclides and other minor radioactive impurities

which might be produced during the target irradiation and potentially hazardous to radiation workers.

There has been a good agreement in recent findings related to threshold energies for positron-emitting radionuclide productions [4,6] and nuclear cross-section data which can be found elsewhere including the recent data released by the International Atomic Energy Agency (IAEA) [6]. However, on the other hand, there has been limited information regarding target chamber designs and requirements, particularly that of liquid chamber.

The liquid target chamber at the Cyclotron Division, Centre for Radioisotopes and Radiopharmaceuticals (PRR), the National Nuclear Energy Agency of Indonesia (BATAN) was initially designed by PRR and then made in Japan by The National Institute of Radiological Science (NIRS). However, the NIRS made some significant changes in the design, particularly the chamber's base materials due to the availability of the base materials in Japan. As a result, it was not immediately apparent if the chamber was reliable and suitable for ^{18}F production at the PRR's cyclotron facilities.

* Corresponding author.

E-mail address: imambatan@yahoo.com

Thus, a reliability study of the target chamber is essential to ensure that the heat deposited in the target during proton irradiation can be properly dissipated while maintaining the target pressure and temperature at the desired levels.

This paper discusses the structure of the liquid target chamber at the cyclotron division, the chamber's limitations and the heat transfer characteristics based on theoretical calculations and experimental data. The recommended irradiation parameters are also presented in the study.

THEORETICAL REVIEW

The heat transfer process in the ion beam bombardment is significantly affected by the beam's energy and current. The power (W , in Watts) deposited into a thick target sample where the proton beam is expected to transfer its entire energy depends on the ion beam current (I , in μA) and energy loss in the sample (E , in MeV), which is given by [7]:

$$W = IE \quad (1)$$

The total heat transfer by conduction (Q , in Watts) for a given material of thickness x (in m) is dependent on the thermal conductivity (k , in W/mK) of the material, heat transfer surface area (A , in m^2) and temperature difference (ΔT) across material, which follows Fourier's law of conduction [8]:

$$Q = \frac{kA}{x} \Delta T = \frac{A\Delta T}{R} \quad (2)$$

where $R = \frac{x}{k}$, is the thermal resistance of the material.

To simplify the calculation of heat transfer through n numbers of materials which are designed in sequence such as insulations, the overall heat transfer coefficient or thermal transmittance (h , in $\text{W/m}^2\text{K}$) is then introduced in which,

$$h = \frac{1}{R_1 + R_2 + \dots + R_n} \quad (3)$$

So the overall heat transfer becomes:

$$Q = hA\Delta T \quad (4)$$

Since the irradiation-induced power deposited into the materials used in the liquid target chamber equals to the total heat transfer, then combining equations (1) and (4) results in:

$$W = Q \quad (5a)$$

$$IE = hA\Delta T \quad (5b)$$

EXPERIMENTS

Computer simulations

A range of computer simulations using Stopping and Range of Ion in Matter (SRIM) version 2008 package [9] was conducted in order to initially calculate the ranges of up to 20 MeV protons in pure water target and to ensure that the protons would not pass through the target which could reach the cooling water.

Theoretical and empirical calculations

Calculations of power deposited into the water target were done by theoretical equation for various proton beam currents of up to 30 μA and variable energies of up to 20 MeV. The predicted power was then employed to calculate the overall heat transfer coefficient of the target chamber in combination with the experimental data.

The heat transfer analysis was also performed to experimentally determine the overall heat transfer coefficient and eventually to empirically formulate and predict the overall heat transfer coefficient for different beam parameters, particularly for various proton beam currents and energies (by assuming that the heat transfer is only through conduction). The empirical formula was deduced by means of polynomial regression.

Target irradiation

The target irradiation was intended to test the reliability of the liquid target chamber in confining ^{18}O -enriched water during routine ^{18}F production in the future as well as to measure the changes in the chamber's pressure and temperature during irradiation. In the irradiation test, an 18 MeV proton beam was perpendicularly incident into 1.35 ml water instead of the ^{18}O -enriched water target for 10 minutes, with variable proton beam currents of up to 5 μA . The initial pressure in the cavity and temperature of the target were 0.04 bar and 25°C respectively. The range of proton in water and ^{18}O -enriched water is expected to be insignificantly different, so that an attempt to employ a pure water target in the experiments should give similar results in the heat transfer process compared to that of using ^{18}O -enriched water. In accordance with the reliability analysis of the target chamber in enduring

high pressure and temperature during proton irradiation, the experimental data were plotted and extrapolated to obtain further proton beam current dependence of the target's pressure and temperature for various proton energies and beam currents.

RESULTS AND DISCUSSION

The PRR's liquid target chamber

While maintaining the target pressure and temperature at the desired levels during proton irradiation, the target thickness also has to be well presented to sufficiently stop a proton beam of up to 20 MeV. As well, the thickness of the target's rear window is required to be designed so that the proton beams are not able to enter the cooling water in the event of irradiation without an ^{18}O -enriched water target being present in the chamber.

The current target chamber consists of 3 main bodies made of stainless steel 304 (SS-304) which houses the cooling air, target cavity and cooling water respectively. The liquid target chamber designed for ^{18}F production is depicted in Fig. 1.

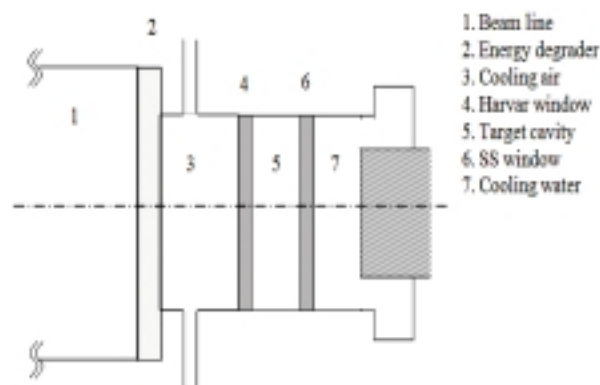


Fig. 1. Schematic design of the Liquid Target Chamber for ^{18}F Production at the Cyclotron Division of Centre for Radioisotopes and Radiopharmaceuticals.

A high energy proton will enter the whole chamber by passing through an aluminum window of few millimeters thickness whilst depositing a quantity of its energy into the aluminium energy degrader. The thickness of the degrader can be changed depending on the required proton energy. The cooling air which is employed mainly to cool the target cavity then dissipates the proton energy when the beam travels through the coolant, just before it reaches the havar window (0.1 mm thick) which separates the cooling air and the liquid target.

Havar is a non-magnetic cobalt-base alloy which comprises of 42.5%Co, 20%Cr, 13%Ni and minor elements such as Fe, W, Mo, Mn. After transferring some energy to the entrance window, the proton beam will finally strike the liquid target in the cavity of 21 mm diameter and 4 mm thick. For instance, based on the SRIM simulation, a 20 MeV proton beam is expected to penetrate into the water target with a projected range of 3.18 mm so that the whole proton energy will be deposited into the pure water of 4 mm thick in the extant chamber without having any excessive energy to reach the cooling water. The range of proton in water and ^{18}O -enriched water will not be significantly different.

Another consideration which should be taken into account with regards to radiation protection and the radiochemical impurities of the liquid target is the possible radionuclides produced during energetic proton irradiation that can potentially complicate the chemical handling and treatment of the produced ^{18}F . Therefore, a study of possible radioactive impurities that might be produced during the proton irradiation in the designed liquid target chamber has been concluded. Typical possible radionuclides produced by a proton beam of above 5 MeV are listed in Table 1.

As shown in Table 1, radioactive impurity produced by $^{27}\text{Al}(p,n)^{27}\text{Si}$ nuclear reaction will not necessarily be taken into account since the aluminum window is placed outside the cavity and the half live of ^{27}Si is only 4.2 seconds [10]. On the other hand, the radioactive impurities such as ^{52}Mn , ^{57}Co which might be produced when the proton beam comes through the havar entrance window will be expected to recoil off the front havar surface outside the target cavity instead of penetrating through the entrance window or recoiling off the rear havar surface. The SRIM-2008 computer simulations confirmed the more favourable front surface recoils rather than the rear ones. Moreover the impurities such as ^{56}Co , ^{183}Re and ^{95m}Tc are expected to be negligibly small as they are only minor elements in the havar alloy. Similarly, the predicted impurities (^{56}Co , ^{52}Mn and ^{57}Co) will not be of major concern since the intensity of the proton beam able to hit the SS-304 is not significant due to its energy loss during its penetration through the water target. Moreover, an energy fraction of much less than 5 MeV is due to hit the havar windows and the SS-304 containment which means that the probabilities of the nuclear reactions between the proton beam and the elements are reasonably negligible since the threshold energies for nuclear reactions between proton and the materials are above 5 MeV.

Table 1. Possible radioactive impurities produced during proton irradiation in the liquid target chamber [10, 11].

Materials	Impurities	Nuclear Reactions	Threshold Energies (MeV)	Half Lives	Major Gamma Energies (MeV)
Aluminum	²⁷ Si	²⁷ Al(p,n) ²⁷ Si	~6	4.2 seconds	2211
Havar	⁵² Mn	⁵² Cr(p,n) ⁵² Mn	~5	5,6 days	744, 936, 1434
	⁵⁷ Co	⁵⁷ Ni(p,n) ⁵⁷ Co	~5	271.7 days	122
SS-304	⁵⁶ Co	⁵⁶ Fe(p,n) ⁵⁶ Co	~5	78.8 days	847, 1238
	¹⁸³ Re	¹⁸³ W(p,n) ¹⁸³ Re	~9	70 days	162.32
	^{95m} Tc	⁹⁵ Mo(p,n) ^{95m} Tc	~7	61 days	203.9, 582.2, 835.1
	⁵⁶ Co	⁵⁶ Fe(p,n) ⁵⁶ Co	~5	78.8 days	847, 1238
	⁵² Mn	⁵² Cr(p,n) ⁵² Mn	~5	5.6 days	744, 936, 1434
	⁵⁷ Co	⁵⁷ Ni(p,n) ⁵⁷ Co	~5	271.7 days	122

Deposited power dependence of proton beam current and energy

It is, in fact, complicated to determine the heat transfer coefficient of each material and fluid involved in the process since it is primarily dependant on the medium’s phase, temperature (as a result of proton beam bombardment), density, molecular bonding as well as proton beam current, though experimental data may help determine the overall heat transfer coefficient as there are some limitations in the theoretical calculations. From the experimental data Fig. 2, the overall heat transfer coefficient (*h*) for the extant liquid target chamber can be empirically approximated by a polynomial of order 6 (with $R^2 = 0.9997$):

$$h = 1 \times 10^{-7}W^6 - 3x10^{-5}W^5 + 0.0047W^4 - 0.3275W^3 + 12.837W^2 - 274.34W + 2825.2 \quad (6)$$

where *W* is the deposited power into the water target.

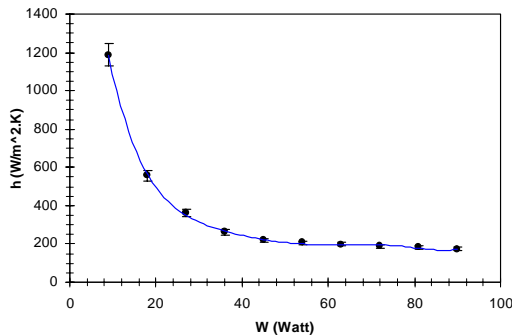


Fig. 2. Overall heat transfer coefficient (*h*) as a function of power (*W*) deposited into the water target.

The experimental data and the empirical formula indicate that the heat transfer coefficient gradually decreases with increasing deposited power up to ~40 W. The coefficient then levels off at higher deposited power. This empirical formula of the overall heat transfer coefficient applied to the extant liquid target chamber is important for further design when a larger capacity of ¹⁸F production is required.

Target pressure and temperature dependence of proton beam current

The pressure and temperature dependence of proton beam current based on the experimental data can be seen in Fig. 3(a) and (b) which demonstrate that an 18 MeV proton beam which is incident into the specified water target is expected to cause the water to boil completely when the beam current is increased to any value above 5 μA. The localised boiling water and bubble formation are of important concerns in the design of the liquid target chamber, particularly to determine the thickness of the target body, the heat transfer and removal as well as the capacity and reliability of the system.

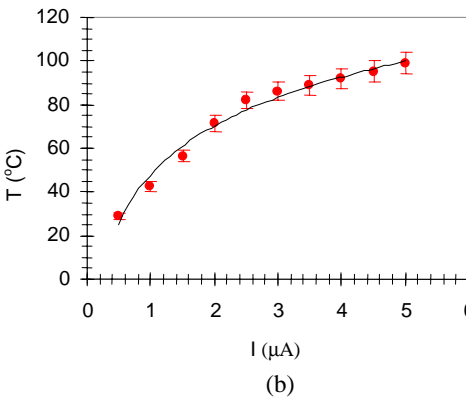
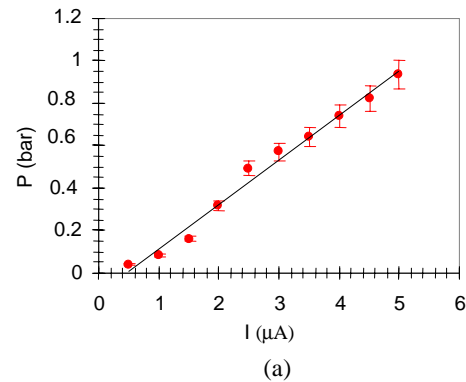


Fig. 3. Experimental data indicating the relationships between proton beam current and (a) target pressure and (b) target temperature for which an 18 MeV proton beam was incident into 1.35 ml water target.

The irradiation parameters for future ^{18}F production can be managed based on these simulations and experimental data as well as the empirical formula, particularly to set up the proton beam energy and current so that the heat produced during target irradiation can be removed properly by the available cooling fluids, and the number of radioactive impurities can be minimised for easier treatment. For this purpose, some extrapolated plots of the relationship between proton beam current and target water pressure as well as temperature obtained from the experimental data are made for studying the reliability and limitations of the extant liquid target chamber (see Fig. 4).

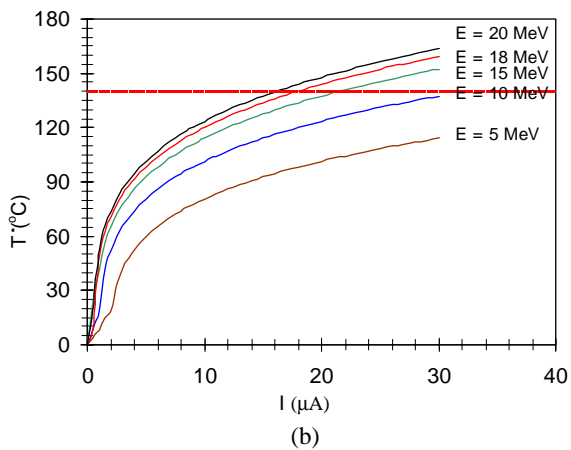
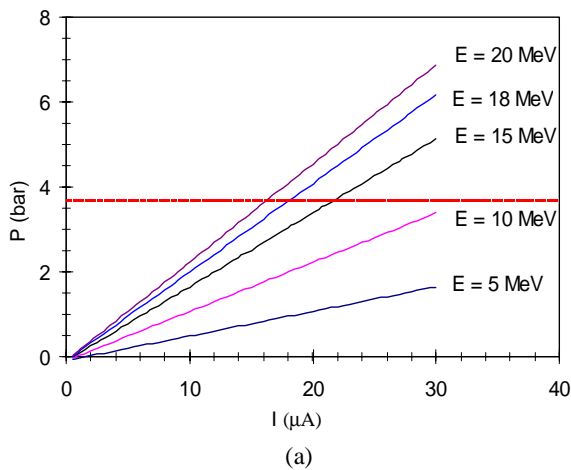


Fig. 4. Extrapolated plots of the target (a) pressure (P) and (b) temperature (T) as a function of proton beam current (I) for several proton energies. The horizontal dashed lines indicate the maximum pressure or temperature endured by the target chamber at given beam currents.

The extrapolated data show that the liquid target chamber is able to endure a pressure (as a result of proton beam bombardment) of up to 3.67

bar or a temperature of up to 140.2°C, as indicated by the horizontal dashed line in Fig. 4(a) and (b). The relationship between saturated pressure (P , in Torr) of water and the corresponding temperature (T , in °C) was calculated from the Antoine equation [12]. At this maximum pressure and temperature, the target system was still able to confine the situation; however, during the target irradiation, experimental data indicated that the water target began to spill over above this pressure (3.67 bar), which raised safety concern.

Furthermore, the maximum proton beam currents and energies recommended to be employed in the target system can be determined from the extrapolated plots. For instance, a bombardment of over 30 μA proton beam into ^{18}O enriched-water is possibly applied to the system if the incident proton energy is set to or below 10 MeV. The ^{18}F production is also reliable for which the proton beam current is set to or below 16 μA for higher energies. Energy-current-related limitations are briefly summarised in Table 2.

Table 2. Maximum proton beam current recommended for a given proton beam energy

Proton Energy (MeV)	5	10	15	18	20
Maximum proton current (μA)	30	30	21.5	18	16

CONCLUSION

Based on this study, the liquid target chamber at the BATAN's cyclotron facilities is reliable for ^{18}F production from proton irradiated- ^{18}O enriched-water target by maintaining the chamber's pressure of up to 3.6 bar if the proton beam current is set to or below 16 μA for all energies or the proton beam energy is kept to or below 10 MeV for any employed beam currents. The radioactive impurities in the target chamber should not complicate the ^{18}F production since their yields are expected to be insignificant. The calculated overall heat transferred presented in this paper can be used to design further liquid target chamber if a larger capacity of the radionuclide production is needed.

ACKNOWLEDGEMENTS

The writers would like to gratefully thank to the Indonesian Government for financially supporting this research. Technical assistance by cyclotron staff at the Centre for Radioisotopes and Radiopharmaceutical, National Nuclear Energy Agency is also sincerely acknowledged.

REFERENCES

1. T. Ishizumi, U. Tateishi, S. Watanabe, T. Maeda and Y. Arai, *Ann. Nucl. Med.* **21** (2007) 299.
2. C. Chen, B. Lee, W. Yau, P. Wu, W. Chen, S. Peng and N. Chiu, *Korean J. Radiol.* **10** (2009) 194.
3. M.S. Berridge, K.W. Voelker and B. Bennington, *Appl. Radiat. Isot.* **57** (2002) 303.
4. S.W. Kiatwanga, P. Leleux and P. Lipnik, *Phys. Rev. C* **42** (1990) 748.
5. M. Guillaume, A. Luxen, B. Nebeling, M. Argentini, J.C. Clark and V.W. Pike, *Appl. Radiat. Isot.* **42** (1991) 749.
6. D.L. Aldama, *Technical Report on Formatting of Cross Sections for Production of Diagnostic Radionuclides in ENDF-6 Format*, IAEA-NDS 210, Cuba (2003).
7. T.A. Cahil, D.W. McColm and B.H. Kusko, *Nucl. Instr. Meth. B* **14** (1986) 38.
8. http://www.en.wikipedia.com/wiki/Heat_transfer. Retrieved March 10 (2009).
9. <http://www.srim.org>. Retrieved March 2 (2009).
10. Anonymous, *The Thin Layer Activation Method and Its Applications in Industry*, IAEA-TECDOC 924 (1997).
11. <http://ie.lbl.gov/toi/nuclide.asp?iZA=140027>. Retrieved March 26 (2009).
12. http://www.en.wikipedia.org/wiki/Antoine_equation. Retrieved March 10 (2009).