Computer simulation techniques to design Xenon-124 solid target for iodine-123 production

T. Kakavand¹, M. Sadeghi^{2*}, K. Kamali Moghaddam², S. Shokri Bonab¹, B. Fateh²

¹ Department of Physics, Zanjan University, Zanjan, Iran

² Cyclotron Accelerator and Nuclear Medicine Department, Nuclear Research Center for Agriculture and Medicine, Karaj, Iran

Background: lodine-123 (1231) is regarded as one of the best radionuclides for in vivo medical studies using single-photon emission computed tomography (SPECT) due to its suitable physical property. Materials and Methods: To design a new system in order to replace cryogenically solidified xenon target by the gas one, some necessary calculations are needed to be done such as finding the excitation functions variation of the production reactions, thick target yield of 123 production, etc. The computer codes Alice91 and SRIM have been used as a calculation tools. Results: According to the suggested design, a conical shaped irradiation vessel made of copper with thickness of 1mm, outlet diameter of 1 cm, 5 cm length and 12° angle at summit can be fixed inside a liquid nitrogen housing chamber. The 124Xe gas was sent to the inside of this very cold conical trap and eventually deposited on its surface in solid form. Calculation showed that during bombardment with 17-28 MeV proton energy, the thickness of solidified xenon layer remained about 0.28 mm. Conclusion: The production yield of 123 can be predicted to be around 150 mCi/µAh. Iran. J. Radiat. Res., 2008; 5 (4): 207-

Keywords: Cyclotron Accelerator, Iodine-123, Xenon-124 gas, Radiopharmaceutical, Alice91 code, SRIM code.

INTRODUCTION

¹²³I in the form of sodium iodide is used most commonly for the measurement of thyroid uptake and thyroid imaging. In addition, radioiodinated compounds have been widely employed in the detection of cardiological, neurological and oncological diseases ⁽¹⁾. Among the various types of nuclear reactions for producing ¹²³I, the following reactions are favored due to the absence of ¹²⁴I and ¹²⁵I impurities:

$$_{54}^{124}Xe(p,2n)_{55}^{23}Cs\frac{5.9\,min}{\beta^{+}}_{54}Xe\frac{2.08h}{\beta^{+}}_{53}I$$

$$^{^{124}}_{^{54}} Xe(p,pn)^{23}_{^{54}} Xe \frac{2.08\ h}{\beta^{^{+}}} ^{^{123}}_{^{53}} I$$

¹²³I production system via ¹²⁴Xe gas target technology has been constructed and installed in Cyclotron and Nuclear Medicine Department of Nuclear Research Centre for Agriculture and Medicine (NRCAM). One of the major problems in this system is the highly expensive cost of the enriched 124Xe gas. Therefore, saving this gas inside the system is very important. Unfortunately, by accidental rupture of the window foil or bad function of O-rings, the whole Xenon gas will escape from the system immediately. In this paper, by using computer codes, Alice91, SRIM and doing some calculations, it has been tried to demonstrate the latest effort for feasibility study of producing 123I with the above mentioned reactions, but using 124Xe solid target instead.

I-123 Production Reactions

The most important nuclear reactions which can be used for the production of ¹²³I, by using the cyclotron accelerator model Cyclon30 (cyclotron at NRCAM) is listed in table 1. It is clear from table 1, that in reactions using ¹²⁴Xe as a target, the purity of the produced ¹²³I is very high (>99.99%). Some famous centers around the world also selected xenon technology for producing ¹²³I. Among them the one can be mentioned is KfK

*Corresponding author:

Dr. Mahdi Sadeghi, Cyclotron Accelerator & Nuclear Medicine Dept., Nuclear Research Center for Agriculture & Medicine, P.O. Box: 31485-498, Karaj, Iran.

Fax: +98 261 4436395

E-mail: msadeghi@nrcam.org

Table 1. Nuclear reactions for Iodine-123 production, regarding NRCAM cyclotron beam outlet characteristics.

Reaction	Proton energy range (MeV)	¹²⁴ I, ¹²⁵ I impurities (%)	¹²⁸ I purity (%)
124 Xe (p,2n) 123 Cs \rightarrow 123 Xe \rightarrow 123 I	20-30	-, 0.01	> 99.99
¹²⁴ Xe (p,pn) ¹²³ Xe→ ¹²³ I	20-30	-, 0.01	> 99.99
¹²³ Te (p,n) ¹²³ I	10-15	1.7, -	98.3
¹²⁴ Te (p,2n) ¹²³ I	23-26	3.8, -	96.2
¹²⁴ Te (d,3n) ¹²³ I	8-16	0.4, -	97.5

Karlsruhe, Nordion Inc. Vancouver, TU Eindhoven (2). In Iran, at Cyclotron and Nuclear Medicine Department of NRCAM, also enriched 124Xe gas target has been selected for producing 123I and its related pharmaceuticals (3). One of the main problems in using this technology is the difficulty for keeping the very expensive enriched xenon gas inside the system (about 50000 \$ per litter). Accidentally rupturing the Titanium window foils or bad functioning of O-rings at the position of pipes junctions through the system, will lead to the immediate escape of the whole expensive xenon gas from the system. Unfortunately, this serious problem reduces the tendency for routine production of 123I from enriched 124Xe technology. In the present study it was tried to demonstrate the latest effort in order to find a solution for overcoming this problem, by replacing xenon ice instead of the gas. This type of target has the advantage of being fail-safe in the sense that the frozen xenon won't be lost in the case of foil rupture.

MATERIALS AND METHODS

Method of calculation

For designing a new system in order to replace cryogenically solidified xenon target by gas one, some necessary calculations are needed to be done, such as finding the excitation functions variation of the production reactions, thick target yield of ¹²³I production, rate of heat transfer during proton bombardment, etc. In this respect, the computer codes Alice91 ⁽⁴⁾, and SRIM ⁽⁵⁾, were used as the calculation tools.

Thick Target Yield Production

In order to calculate the production yield of ¹²³I, the following equation has been used (6):

$$Yield = 0.1017 \frac{H(1 - e^{-\lambda t})}{M} \int_{E_0}^{E_1} \frac{\sigma(E)}{\left(\frac{dE}{dx}\right)} dE$$

Yield=Thick target yield (mCi/ μ A-hr) $T_{1/2}$ = Half-Life (hour) (dE/dx) = Stopping Power (MeV/mg-

cm²)

 σ (E) = Reaction cross section (mb)

H = Isotopic enrichment (in %)

M = Molecular mass

 E_0 = Outlet energy (in this work =28 MeV)

 $E_i = \text{Inlet energy (in this work = 17 MeV)}$

In this relation, the value of the stopping power was calculated by SRIM code, and the reaction cross sections, σ (E), were extracted from the generated data in figure 1.

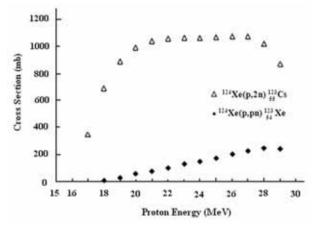


Figure 1. Cross section of the ¹²⁴Xe (p,2n) ¹²³Cs and ¹²⁴Xe (p,pn) ¹²³Xe reactions obtained via Alice code.

Heat Transfer Calculation

According to the above achieved results, the best proton energy range for producing ¹²³I via ¹²⁴Xe target bombardment was between 17-28 MeV. By using SRIM code, the best thickness of solidified xenon target was calculated to be about 2.83 mm. In order to stop the remainder penetrated 17 MeV energy proton beam, the minimal thickness of copper element, which was decided to deposit solidified xenon on it (because of its high ability for transferring heat with coefficient k equal to 401 W/m.k), was

calculated to be about 0.6 mm. Since the copper element, with 1 mm thickness, was also selected for constructing the body of the target house, there was no reason to worry about the penetration of proton beam, outside the system.

RESULTS AND DISCUSSION

Excitation functions calculation

Cross section variation determination (excitation functions) for two (p, 2n) and (p, pn) reactions were the most important subject for research in this field. For reaching the highest ¹²³I production efficiency, it was necessary to find the best range of proton energy which in this energy range, the cross sections of these reactions had the maximum accessible value. In this regard the Alice91 code was used and the excitation functions of ¹²⁴Xe (p, 2n) ¹²³Cs-¹²³Xe-¹²³I and ¹²⁴Xe (p, pn) ¹²³Xe-¹²³I is shown in figure 1, the best energy range of incident proton beam was between 17-28 MeV.

Comparisons of the achieved results with experimental data for the above two reactions are shown in figures 2 and 3. As it was shown in previous sections, the calculated excitation functions for the ¹²⁴Xe (p, 2n) ¹²³Cs and ¹²⁴Xe (p, pn) ¹²³Xe reactions, was in agreement with the results obtained at other laboratories around the world ^(7, 8).

The thick target yield of ¹²³I production, as a function of the proton energy was calculated after 6.6 hours bombardment (the waiting time at the end of bombardment

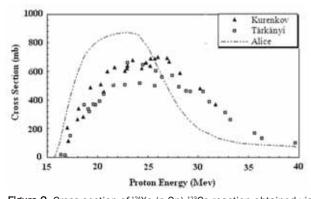


Figure 2. Cross section of 124 Xe (p,2n) 123 Cs reaction obtained via Alice code and experimental data.

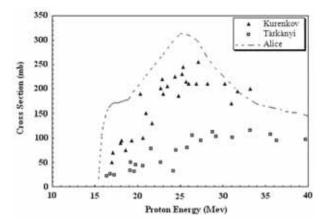


Figure 3. Cross section of ¹²⁴Xe (p,pn) ¹²³Xe reaction obtained via Alice code and experimental data.

needed to maximize the activity of the produced ¹²³I) are shown in figures 4 and 5 for the above two reactions. It is clear from these curves that the production yields of ¹²³I through (p, pn) reaction has been negligible in comparison with (p, 2n) reaction.

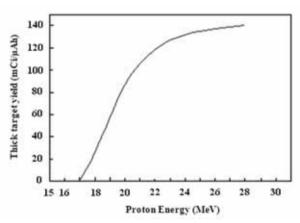


Figure 4. Thick target yield of ¹²³I for ¹²⁴Xe(p,2n) ¹²³Cs reaction as a function of the proton energy for solid xenon target.

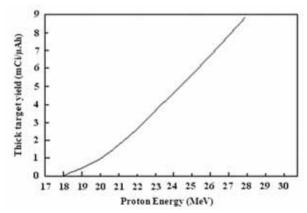


Figure 5. Thick target yield of ¹²³I for ¹²⁴Xe(p,pn) ¹²³Xe reaction as a function of the proton energy for solid xenon target.

Figure 6 shows the variation of the thick target yield versus the proton energy for ¹²³I production via xenon gas target, which was reported by Firouzbakht *et al.* ⁽⁹⁾. Comparison of the production yield of ¹²³I through solid xenon target (figures 5 and 6), which was calculated at 6.6 hours after the end of bombardment, is much higher than that of the gas xenon target ⁽⁹⁾.

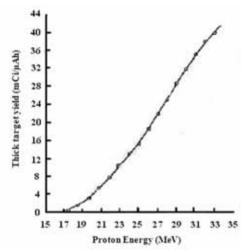


Figure 6. Integral thick target yield of ¹²³I versus proton energy for xenon gas target technology reported by Firouzbakht *et al.* ⁽⁹⁾.

However, because of lower stopping power for solid xenon target than the gas one, the thick target yield of ¹²³I has been much higher for the former than the latter (about 140 mCi/µAh and 35 mCi/µAh, at proton energy range of 17-28 MeV).

Because of intense heat characteristic of solid xenon material, its maximum thickness in solid form was not more than 0.5 mm (10). So to let the proton beam penetrate from inside the formed xenon ice body up to 2.83 mm, and to decrease its energy to 17 MeV, it was necessary to make the target house conically. Calculation showed that the best summit angle for this conical shape target house should be 12°. At this angle, the real thickness of xenon ice layer was about 0.2 mm, which was less than critical 0.05 mm thickness. The melting point of solid xenon was about 161°K (-112°C) and the temperature of liquid nitrogen was about 133°K (-140°C). In the worst condition of heat transferring between these two medium, the maximum proton beam current permitted to enter the target house was about 1.46 µA.

Design of target vessel

The schematic diagram of the suggested conical irradiation copper vessel is shown in figure 7. The thickness of the cone is 1 mm with about 5 cm height and 1cm iterance diameter (in front of proton beam). The summit angle of this cone is 12°. The calculation showed that the weight of xenon gas which was sent inside the conical trap and cooled by liquid nitrogen, should have been about 0.7 g (~120 cm³) in order to deposit on the inside surface of it as a solid target material.

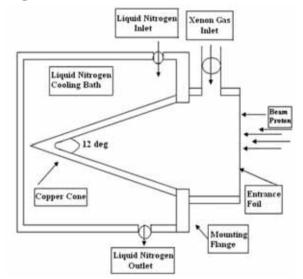


Figure 7. Schematic Diagram of our suggested target vessel for the proton bombardment of solid xenon.

Firouzbakht et al. (11), have suggested a similar conical target vessel design with 15 degrees at summit of the cone. It is believed that by increasing the angle from 12 degree, the perpendicular thickness of the solid xenon, inside surface of the cone should increase in some way which eventually causes that xenon, like insulator, prevents the good heat removing from the system, during proton bombardment. Furthermore, in the present design the shape of summit was curved at inside edge. This form will be helpful for the reduction of the temperature gradient at the inside edge of summit, which eventually prevents the xenon gas change to ice shape, at this place, more than the other parts of target house.

Other advantages of this technique in comparison with the previous one (with xenon gas target), can be summarized as follows:

- ➤ Prevention of sudden loss of the whole expensive enriched ¹²⁴Xe gas from the system, because of window foil failure or bad functioning of some O-rings at the position of pipes junctions.
- ➤ In solid xenon target process, the initial pressure inside the system should be 1 bar (atmosphere pressure). However, in xenon gas target process, the initial pressure inside the target vessel should be at least 5 bar.
- ➤ The amount of enriched ¹²⁴Xe which is needed in each bombardment, for solid target process has been about 0.7 g (120 cc at STP); whereas for gas, it should be more than 4 grams. This way, the required inventory of expensive ¹²⁴Xe for preparing the target will reduce effectively.

According to the suggested design, a conical shaped irradiation copper vessel with 1mm thickness, 1cm outlet diameter, 5cm length and 12° angle at summit was fixed inside a liquid nitrogen housing chamber. The 124Xe gas was sent to the inside of the very cold conical trap and eventually deposited on its surface in solid form. The calculation showed that during bombardment with 17-28 MeV proton energy, the thickness of solidified xenon layer will remain around 0.28mm. Likewise. thermodynamical calculation showed that in order to prevent the evaporation of solidified xenon, the maximum permissible proton beam current for the system should have been less than 1.4 µA. According to these working conditions, the production yield of ¹²³I could be predicted to be around 150 mCi/µAh.

REFERENCES

- Lindner L, Brinkman G, Suer THG, Schimmel A, Veenboer CJ, Karten FHS, Visser J, Leurs CJ (1973) Radiopharmaceuticals and Labeled Compounds. IAEA, 1: 308-316, Vienna, Austria.
- Firouzbakht ML, Schyler DJ, Wolf AP (1992) Effect of foil material on the apparent yield of the 124Xe (p,x) 123I reaction. Appl Radiat Isotopes, 43: 741-745.
- Kamali Moghaddam K, Fateh B, Mirzaii M, Jalilian AR (2005) Production of I-123 radiopharmaceutical via xenon gas target, using NRCAM cyclotron, International Symposium on Trends in Radiopharmaceuticals (ISTR-2005), IAEA, Vienna, Austria.
- Blann M and Bislinghoff J (1991) Code Alice-Livermore 9, Lawrence Livermore national Laboratory, UCID-19614, RUSSIA.
- Ziener JF, Biersack JP, Littmark U (2006) The code of SRIM, the Stopping & Range of Ions in Matter, USA.
- Fateh B (2000) Investigation of hydro dynamical behavior of gaseous material which put in front of the high proton beam and designing gas target for producing ¹²³I radioisotope, Ph.D. thesis, Amir-Kabir University, Tehran, Iran.
- Tárkányi F, Qaim SM, Stöcklin G, Sajjad M, Lambrecht RM, Schweickert H, (1991) Excitation functions of (p,2n) and (p,pn) reactions and differential and integral yield of ¹²³I in proton induced nuclear reactions on highly enriched 124Xe. Applied Radiation and Isotopes, 42: 221-228.
- Kurenkov NV, Malinin AB, Sebyakin AA, Venikov NI (1989) Excitation functions of proton induced nuclear reactions on Xe-124: Production of ¹²³I. J Radioanal Nucl Chem Letters, 135: 39-50.
- 9. Firouzbakht ML, Schyler DJ, Wolf AP (1993) Production of high purity iodine-123 from xenon -124 at energies between 15 and 34 MeV. Applied Radiation and Isotopes, 38: 421-425.
- Fukugaki K, Inoue T, Shimoura A, Miyamoto S, Amano S, Mochizuki T (2003) Rotating cryogenic drum for continuous supply of solid Xe target, 2nd Int. EUVL Symposium Antwerp, Belgium.
- 11. Firouzbakht ML, Schyler JD, Wolf AP (1993) A solid xenon ice target for the production of I-123. *Journal of labelled compounds and radiopharmaceuticals*, **32**: 243-244.