



Evaluation of irradiation parameters of enriched ^{124}Xe target for ^{123}I production in cyclotrons

L.C.A. Sumiya*, V. Sciani

Cyclotron Department, Instituto de Pesquisas Energéticas e Nucleares—IPEN, SP-11049, Brazil

ARTICLE INFO

Article history:

Received 13 December 2006

Received in revised form

3 April 2008

Accepted 11 April 2008

Keywords:

^{123}I production process

^{124}Xe target

Cyclotron production

High yield

ABSTRACT

The production of high-purity ^{123}I that utilizes an isotopically enriched ^{124}Xe target and bombardment with 30 MeV protons, through the reactions $^{124}\text{Xe}(p, 2n)^{123}\text{Cs} \rightarrow ^{123}\text{Xe} \rightarrow ^{123}\text{I}$ and $^{124}\text{Xe}(p, pn)^{123}\text{Xe} \rightarrow ^{123}\text{I}$, is described. The aim of this work was to improve the production parameters, such as ^{124}Xe load pressure, beam current, decay time and target heating to recover ^{123}I to obtain high-production ^{123}I yield at low cost.

© 2008 Elsevier Ltd. All rights reserved.

1. Introduction

IPEN, the Energy and Nuclear Research Institute, has developed activities for the production of radioisotopes in reactors and cyclotrons in response to the increase in demand and diversification in the utilization of radioisotopes for diagnosis and therapy. The cyclotron-produced radionuclide ^{123}I has had a prominent role in nuclear medical diagnostic applications, both in labeled compounds and in monoclonal antibodies. With time ^{123}I has substituted ^{131}I (Mestnik and Mengatti, 1992; Arif et al., 1996), due to the fact that ^{123}I has the following characteristics for medical purposes (Legoux et al., 1985; Bechtold et al., 1991), such as:

- gamma-ray energy of 159 keV,
- half-life of 13.2 h and
- no beta emission.

Nowadays, the most commonly used commercial processes for production of ^{123}I are: $^{124}\text{Xe}(p, 2n)^{123}\text{Cs} \rightarrow ^{123}\text{Xe} \rightarrow ^{123}\text{I}$ and $^{124}\text{Xe}(p, pn)^{123}\text{Xe} \rightarrow ^{123}\text{I}$. (Firouzbakht et al., 1987,1992; Venikov et al., 1991), using xenon enriched 99.9% in ^{124}Xe , and irradiation with protons (Tárkányi et al., 1991). Looking for improvements in this production process, an experimental irradiation target was constructed to carry out studies of the production parameters, such as gas load pressure, beam current, decay time and target

heating to recover ^{123}I , in order to optimize our routine production activities to obtain high-production yield at low cost.

In this work, the following parameters were studied:

- integrated current used in each irradiation,
- beam current intensity,
- xenon pressure, inside the target chamber,
- decay time after end of bombardment (EOB)
- target assembly gas helium temperature and
- heating of the target chamber during ^{123}I recovering process.

2. Materials and methods

The experimental target system was made of copper coated by a thin layer of nickel. The internal chamber target has a conical geometry with a volume of 40 ml, which was cooled with de-ionized water at 18 °C. Two molybdenum foils, cooled by a compressed flux of helium gas, were set between the target and the beam line. A collimator was installed in front of the target, to delimit the irradiation area. Fig. 1 shows a schematic representation of the experimental target system.

The target system was installed at the cyclotron beam line. The cooling water and cooling helium circuits were arranged at the target position, loaded with ^{124}Xe . The cooling helium flows between the molybdenum windows of the target in a closed circuit using a compressor. The control of the equipment is made by pneumatic valves activated during each phase of the process.

* Corresponding author. Tel.: +55 11 31339937; fax: +55 11 31339928.
E-mail address: sumiya@terra.com.br (L.C.A. Sumiya).

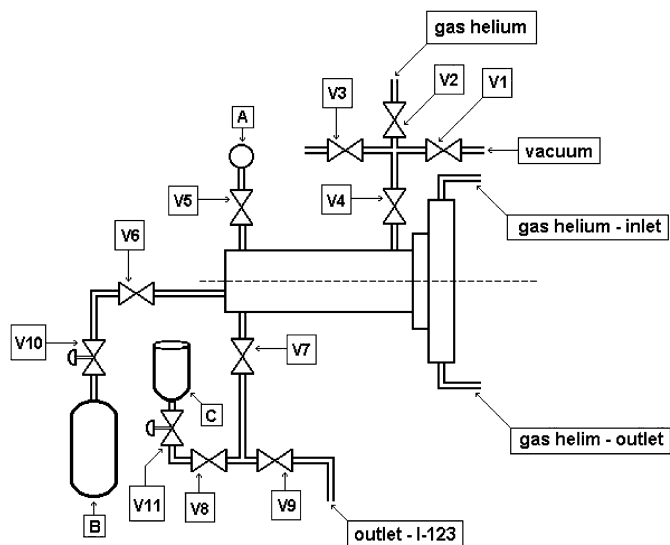


Fig. 1. Schematic representation of the experimental target system, where V1, V2, V3, V4, V5, V6, V7, V8 and V9 are pneumatic valves; V10 and V11 are manual security valves; A—manometer; B— ^{124}Xe vessel where the gas stays stored; C—vessel that is filled with H_2O , about 40 ml.

The irradiations of this work were carried out using an Ion Beam Applications (IBA; Louvain-la-Neuve, Belgium) CYCLONE-30 30 MeV cyclotron installed at IPEN (Bol et al., 1991). All irradiations were carried out at a beam current ranging from 10 to $30\ \mu\text{A}$ between 20 and 40 min, given an average integrated current at the target of $10\ \mu\text{Ah}$. After EOB, the target was allowed a cooling time for the ^{123}Cs to decay to ^{123}Xe and ^{123}I .

The recovery of high-purity ^{123}I adsorbed on the walls of the target chamber was performed by washing with water. The effects of the length of time that the water remained inside the target and the heating temperature were studied. This procedure was made in order to improve the removing of the ^{123}I from the target wall, thereby decreasing product losses.

3. Results and discussion

The obtained results in the first set of experiments are given in Table 1, where the ^{124}Xe pressure was 1.8×10^3 hPa and the decay time after EOB was fixed at 7 h. The target heating time was fixed at 10 min at $65\ ^\circ\text{C}$.

In Table 1, it can be seen that the activity and yield values are reproducible, proving that the system has a good stability in this optimization stage.

When the average yields given in the Table 1 of $87.69\ \text{MBq}/\mu\text{Ah}$ are compared with the routine productions value of $89.91\ \text{MBq}/\mu\text{Ah}$ the difference between them is approximately 2.5%, which is not significant considering routine production, in which the ^{124}Xe pressure is normally 2.0×10^3 hPa.

The experimental ^{123}I yield was determined as a function of the decay time after the EOB and is given in Fig. 2. The following parameters were fixed: gas pressure 1.6×10^3 hPa and heating time 15 min at $65\ ^\circ\text{C}$. In this figure it is observed that for time less than 13 h, a considerable amount of ^{123}I is lost but when this decay time increases to about 15 h, stability is reached. These results are important because it is possible to fix the decay time to 15 h, which increases the ^{123}I yield in 40%.

Fig. 3 shows the variation of the ^{123}I yield as a function of the temperature target chamber to remove ^{123}I from the walls. In these experiments, ^{124}Xe pressure was 1.6×10^3 hPa, decay time 7 h and heating time 15 min.

Table 1

Some values of EOB activity and ^{123}I yield about 7 h of decay time after EOB

Total dose (μAh)	Beam current (μA)	Time of the bombardment (min)	EOB activity (MBq)	Yield ($\text{MBq}/\mu\text{Ah}$)
10.5	23.3	27	885.04	88.43
10.0	25.0	24	920.56	87.69
10.0	25.0	24	874.68	87.32

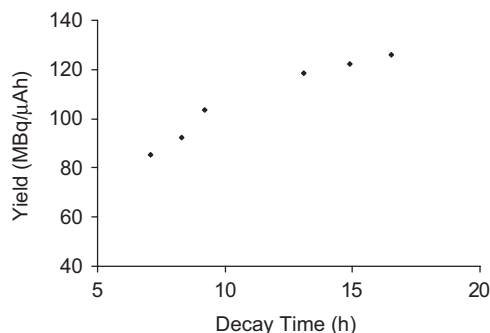


Fig. 2. Yield of ^{123}I as a function of the decay time, with gas ^{124}Xe pressure 1.6×10^3 hPa and heating time 15 min at $65\ ^\circ\text{C}$. The total dose was about $10.0\ \mu\text{Ah}$ with a middle current of $25\ \mu\text{A}$.

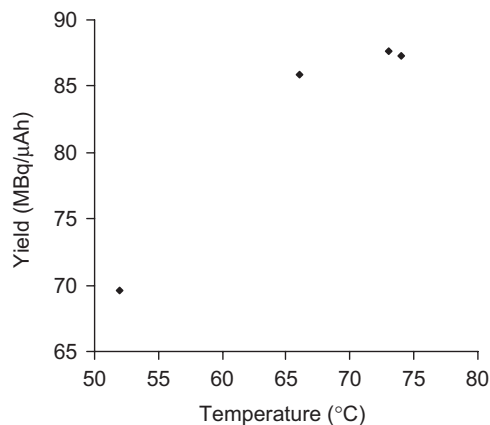


Fig. 3. ^{123}I removal yield of as a function of the target chamber temperature, with gas pressure of 1.6×10^3 hPa, decay time of 7 h and heating time of 15 min.

The temperature influence on the removal of ^{123}I adsorbed on the walls of the internal target chamber was investigated and it was observed that, for temperatures from 50 to $65\ ^\circ\text{C}$, the removal of ^{123}I is less efficient than when the temperature is above $65\ ^\circ\text{C}$. When the temperature reaches $70\ ^\circ\text{C}$ the amount of ^{123}I that can be removed becomes constant. From this result, we can consider $70\ ^\circ\text{C}$ as a working parameter.

In relation to the ^{124}Xe pressure, it was observed that this parameter clearly induces changes in the ^{123}I yield. For these experiments, the fixed parameters were:

- decay time: 15 h and
- temperature in the chamber target: $70\ ^\circ\text{C}$.

In addition, the integrated current and the irradiation time were kept constant. In Fig. 4, the variation in the ^{123}I yield as a function

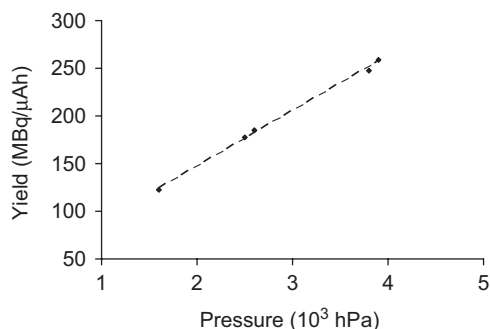


Fig. 4. Yield of ^{123}I produced as a function of the gas ^{124}Xe load pressure. The bombardment time was about 30 min with a middle current of $25\ \mu\text{A}$.

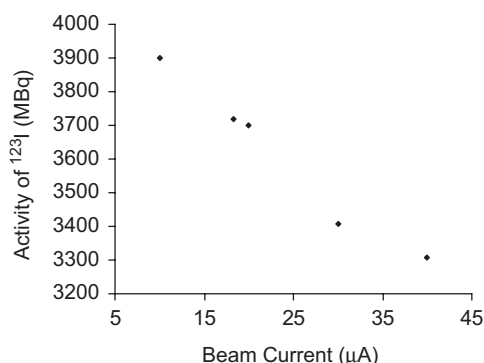


Fig. 5. ^{123}I Activity as a function of the beam current intensity. The parameters irradiation time and gas pressure were found to be about 40 min and 1.8×10^3 hPa, respectively.

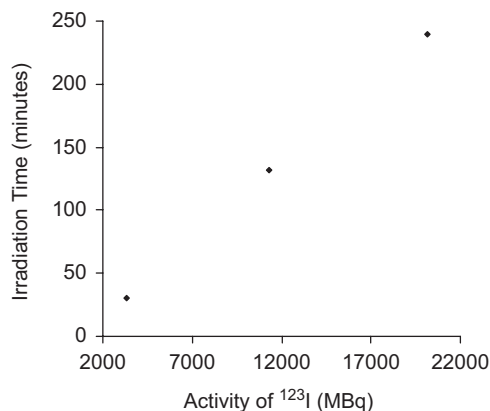


Fig. 6. ^{123}I activity as a function of the irradiation time. The gas ^{124}Xe pressure was fixed at 1.8×10^3 hPa and the beam current was about $25\ \mu\text{A}$.

of the ^{124}Xe pressure inside the target chamber is given. It is observed that the ^{123}I yield increases with the increase in gas pressure in a linear relationship. So, for 1.6×10^3 hPa, $122.1\ \text{MBq}/\mu\text{Ah}$ was obtained and for 3.9×10^3 hPa, $259\ \text{MBq}/\mu\text{Ah}$ was obtained. Unfortunately, the gas pressure inside the chamber is limited by the integrity of the chamber windows. Another parameter that can damage the windows is the temperature that can be reached during irradiation; the main factor for this effect is the beam current intensity.

In Fig. 5, the ^{123}I activity as a function of the beam current is presented. In these experiments irradiation time was fixed at about 40 min and the gas ^{124}Xe pressure to 1.8×10^3 hPa. Here it is

observed that the activity decreases when the beam current increases. This behavior was expected because at higher beam current the gas density of the irradiated volume decreases. This fact occurs because the power (energy and beam current) deposit in this volume produces gas convection and the molecules migrate to the walls of the irradiation chamber and fewer molecules interact with the proton beam (Schlyer et al., 1987).

In Fig. 6, the effect of the irradiation time on ^{123}I production is presented. Both ^{124}Xe pressure and beam current were fixed. In this figure, a linear growth of the activity as a function of the irradiation time was observed for irradiation time up to 5 h.

A close analysis of the parameters beam current, irradiation time and integrated current suggests the need for a careful balance among them to obtain optimal operational conditions for ^{123}I production. Other important factors are the ^{124}Xe pressure and beam current intensity since both are very important in the preservation of the chamber windows. In this work, these optimized parameters were found to be:

- ^{124}Xe gas pressure: 3.0×10^3 hPa and
- proton beam current intensity in the range $55\text{--}60\ \mu\text{A}$.

Additionally, some parameters were incorporated in the routine production and this modification allowed the integrated current to be decreased from 450 to $350\ \mu\text{Ah}$, a reduction of approximately 30% with an associated reduction in the irradiation time and also in the ^{123}I production cost.

4. Conclusion

For irradiation times between 10 and 60 min, the optimal decay time for ^{123}I recovery was found to be 15 h, which can reach 40% for shortened irradiation times. The ideal temperature in the target chamber wall was found to be $70\ ^\circ\text{C}$, with a heating time of 15 min.

^{124}Xe pressure has been shown to be an important parameter for the ^{123}I yield, but care has to be taken together with the beam current intensity to avoid windows damage. Concerning the decay time, it was concluded that for short irradiations (20–40 min) this parameter should be approximately 15 h, avoiding a loss up to 40% in the ^{123}I yield obtained. The heating temperature parameter for the removal of the ^{123}I is not frequently mentioned in the literature. Nevertheless, in this work the ^{123}I yield was influenced by this parameter and depending on the range of the temperatures chosen, a loss of about 25% occurs. The optimal ^{123}I removal temperature was $70\ ^\circ\text{C}$.

The ^{124}Xe load pressure has shown to be a very important factor because it affects the ^{123}I yield and because it also has an influence on the target window integrity. Therefore, care should be taken to guarantee the operational safety of the system.

To optimize the production of ^{123}I , there must be a balance between these operational parameters to obtain good ^{123}I yield with a reduction in production cost.

References

- Arif, M., Zaidi, J.H., Qureshi, I.H., Iqbal, M.Z., 1996. Fission neutron spectrum averaged cross sections of some threshold reactions on tellurium: estimation of radioactive impurities in (n, γ)-produced ^{131}Te – ^{131}I . *Radiochim. Acta* 73, 45–48.
- Bechtold, V., Kernert, N., Schweickert, H., 1991. Modern gas target technology for the production of high quality radiopharmaceuticals. In: *Matin, B., Ziegler, K. (Eds.), 12th International Conference on Cyclotrons and Their Applications, May 8–12, 1989, pp. 519–522.*
- Bol, J.L., Chevaleyre, A., Conard, E., Jongen, Y., Ladeuze, M., Lannoye, G., Ledocte, T., Lacroix, M., Niname, A., Rijckewaert, G., Zarembo, S., 1991. High intensity H^- cyclotrons for radioisotope production. In: *Matin, B., Ziegler, K. (Eds.), 12th*

- International Conference on Cyclotrons and their Applications, May 8–12, 1989, pp. 149–151.
- Firouzbakht, L.M., Teng, R.R., Schlyer, D.J., Wolf, A.P., 1987. Production of high purity iodine-123 from xenon-124 at energies between 15 and 24 MeV. *Radiochim. Acta* 41, 1–4.
- Firouzbakht, L.M., Teng, R.R., Schlyer, D.J., Wolf, A.P., 1992. Production of iodine-123 from xenon-124: cross-section and yields. *Radiochim. Acta* 56, 167–171.
- Legoux, Y., Cieur, M., Goutheraud, R., Drouet, J., Crouzel, C., Syrota, A., 1985. Production of high specific activity ^{123}I for protein iodination for medical use. *Appl. Radiat. Isot.* 36, 63–67.
- Mestnik, S.A.C., Mengatti, J., 1992. Studies for production of iodine-123 at CV-28 cyclotron of IPEN-CNEN/SP. In: Fourth International Workshop on Targetry and Target Chemistry, September 9–12, 1991, Switzerland. Regin Weinreich, Switzerland, pp. 71–72.
- Schlyer, D.J., Heselius, S.J., Wolf, A.P., 1987. Temperature distribution in a gas target during irradiation. In: Proceedings of the workshop on Targetry and Target Chemistry, September 22–25, 1987, Heidelberg.
- Tárkányi, F., Qaim, S.M., Stöcklin, G., Sajjad, M., Lambrecht, R.M., Schweickert, H., 1991. Excitation functions of (p, 2n) and (p, pn) reactions and differential and integral yields of ^{123}I in proton induced nuclear reactions on highly enriched ^{124}Xe . *Appl. Radiat. Isot.* 42, 221–228.
- Venikov, N.I., Novikov, V.I., Sebyakin, A.A., Fomichev, D.I., Shabrov, V.A., Kurchatov, I.V., 1991. Production of high-purity ^{123}I on IAE cyclotron. In: Matin, B., Ziegler, K. (Eds.), 12th International Conference on Cyclotrons and their Applications, May 8–12, 1989, pp. 519–522.