PARASITIC USE OF A 72 MEV PROTON BEAM FOR ONLINE PRODUCTION OF I-123

H.W. Reist, L. Rezzonico
Swiss Institute for Nuclear Research, CH-5234 Villigen, Switzerland

Summary

The motivation for the production of the valuable radio isotope I-123 by the reaction I-127(p,5n)Xe-123 using the 72 MeV injector beam parasitically lies in the production energy, production yield and in the decay rates of the products, permitting low beam intensities which can be compensated by longer beam exposures.

The target is a vacuum vessel containing pure NaI which is kept above the melting point by beam heating and external heating. A continuous flow of helium, streaming over the surface of the molten NaI, carries the produced Xe isotopes on line through a stainless steel tube in to a cold trap. The target is inserted close to the beam from the side. An electrostatic beam splitter 5m up stream from the target peals off the desired production beam from the main beam. The target is positioned horizontally and vertically using beam monitors. The beam intensity on the target is computer controlled by correcting the position of the beam splitter using the intensity and the profile of the main beam. The other parameters can be set from a console or by the computer. The control is then taken by a local loop. Presently ∿3 Ci of I-123 are produced per week in collaboration with the neighbour institute EIR.

Introduction

Radioiodine labelled physiologically specific pharmaceuticals like proteins, antibiotics, fatty acids, hormones and antibodies have become an important tool in nuclear medicine applications and in clinical research.

Due to its decay by electron capture leading to the emission of a 83% abundant 159 keV X-ray and to the minimum presence of higher energy gammas, I-123 has almost optimal characteristics for detection in medical imaging systems [1]. The high photon to electron ratio and the short half life of 13.2 h reduce the patient dose significantly compared to the more common I-125 and I-131 [2,3].

The production method via the reaction I-127 (p, 5n)Xe-123 → I-123 was developed at the Crocker Nuclear Laboratory of the University of California [4] and enables the production of high purity I-123. The nuclear reaction I-127 (p, 5n) Xe-123 has a cross section peaking at a proton energy of about 55 MeV [5-8]. The competing reaction, I-127 (p, 3n) Xe-125, leading to the undesirable by-product Xe-125 has a peak yield at about 32 MeV [5-8]. The parent daughter decay rates permit relatively long beam exposures without leading to a high I-125 contamination which allows accordingly low beam intensities. Therefore, by proper choice of the target thickness the 72 MeV injector beam is appropriate for a parasitic production of I-123 [9].

Description of the system

Target- and beam splitter arrangement

The present production facility is mounted in the transfer line from the 72 MeV injector cyclotron to the ring machine and uses ∿6 μA split from the 100-200 μA main beam. The target is placed 1.5 m after the first bending magnet and 2 m ahead of an energy analyzing slit. Protons scattered out of the target and by-passing the stopper behind it, are mostly stopped at the energy analyzing slit and do not interfere with the cyclotron operation. The horizontal and vertical beam widths measured with the beam scanners 30 cm before the target are 4 to 6 mm FWHM and 1.5 to 3 mm FWHM respectively. These rather small widths have to be taken into account in the target design.

Electrostatic beam splitter

The separation of the production beam from the main beam is made with an electrostatic beam splitter which gives a separation of 12.5 mm at the target. The splitter consists of a fixed cathode and a movable thin septum. The applied field is ∿60 kV/cm over a length of 15 cm. The beam splitter is equipped with an adjustable tilt to reduce beam losses. The movable septum is made of 15 molybdenum strips (7 mm wide and 30 μm thick) and a leading strip of tungsten (3 mm wide and 50 μm thick) because it is directly hit by the incoming protons. The strips are separately tensioned over a supporting structure. The position and flatness of the strips were carefully controlled during their tensioning. More details are given in reference [10].

Target and target assembly

The target vacuum vessel is a stainless steel box, 17 mm wide, 48 mm high and 18 mm long in the beam direction. The wall thickness is 0.5 mm in the beam path. This vessel is half filled with pure NaI (18 g). The NaI is melted before exposure to the beam and kept liquid, if the necessary cyclotron beam intensity is not available, by external radiation heating with a tungsten filament. During a normal production run, the NaI is kept liquid by beam power dissipation (300 W). The surface temperature of the NaI should not be to far above its melting point, say 680 C^0 to avoid blockage of the He/Xe gas outlet by evaporated NaI. The target is surface cooled by radiation. The vessel outer surface is plated with 0.1 mm of copper, has cooling wings soldered to it and finally all these cooling surfaces are coated with TiO_2(by sputtering) to improve the heat transfer. The temperature of the NaI is measured by a Chromel-Alumel thermocouple, inserted through a small stainless steel tube

CH1996-3/84/0000-0403 $1.00 © 1984 IEEE

into the melt. A further thermocouple measures the gas outlet temperature. The average temperatures are ∿700 °C half way down in the melt at T1 and ∿500 °C at the gas outlet (T2) at normal production conditions. Fig. 1 shows the main features of the presently used production target.

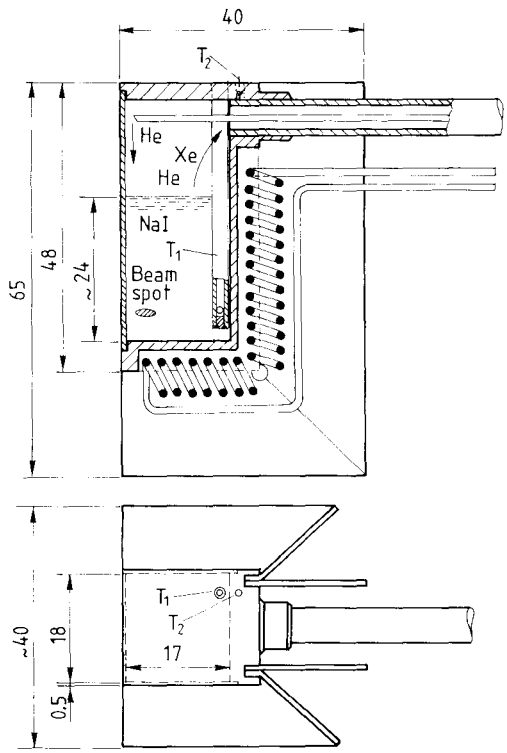

Fig. 1:
The presently used production target. The target vessel of stainless steel contains 18 g of pure NaI which is melted before the beam exposure by radiation heating with a tungsten filament. The beam is positioned in the lower left corner of the vessel towards the main beam. The helium enters through the capillary tube streams over the surface of the melt and leaves the target by the outer tube. The wings reinforce the dissipation of the beam deposited heat.

NaI is a satisfactory target material as it does not decompose by radiolyses and because the beam heating can be used to get an efficient release of the xenon. The positioning of the target in the beam proved to be essential for a good release of the xenon and for a long operational lifetime. The best position is with the beam in the lower left corner, which (i) intensifies the convection carrying the xenon to the surface by the buoyancy of the NaI in the bombarded region and reducing the temperature by dispersing the heat in the melt, and (ii) allows the target to be further away from the main beam, increasing the target safety in case of an unstable beam.

As is shown in Fig. 2 the thickness of the target is crucial with regard to the yield and to the nuclidic purity of the produced Xe-123. The effective target thickness is directly influenced by the beam intensity and the spot size on the target. At 5 to 6 μA it ranges between 3.6 and 4.2 g/cm² depending on the actual spot size of the beam. The measured I-125 activity, 36 h after completion of processing, ranges between 0.75 and 1.5 % of that for I-123.

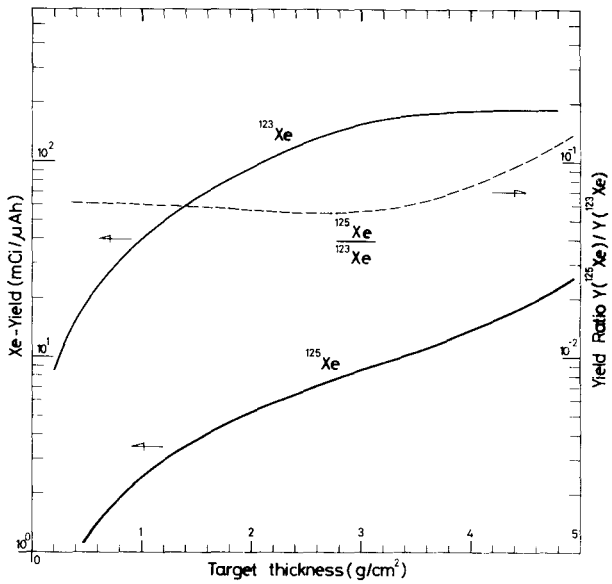

Fig. 2:
The yields of Xe-123 and Xe-125 in mCi/μAh as function of the effective target thickness in g/cm². The production cross sections are taken from reference [7] and integrated:

$$Y(t,E_{in}) = N_I \cdot I_p \frac{\ln 2}{T_{1/2}} \cdot \frac{\Delta E}{3.7 \cdot 10^7} \sum_k \frac{\sigma(E_k)}{\frac{dE}{dx}(E_k)}$$

$$t = \Delta E \sum_k \left[\frac{dE}{dx}(E_k)\right]^{-1}$$

E_{in} = 70 MeV

N_I = Number of iodine atoms in 1 g NaI

I_p = Number of protons per second in 1 μA beam = $6.242 \cdot 10^{12}$ s^{-1}

$T_{1/2}$ = Half life (h) of the produced isotope

$\sigma(E_k)$ = Production cross section (cm^2) at the proton energy E_k

$\frac{dE}{dx}(E_k)$ = Stopping power (MeV-cm^2/g) in NaI at the proton energy E_k taken from reference [11]

The ratio of the yields Y(Xe-125) / Y(Xe-123) makes clear that the target thickness should be limited at ∿4 g/cm². The I-123 yield (6.7 h after irradiation) may be obtained by multiplying the Xe-123 yield by 0.110.

The xenon is swept out of the target chamber by a continuous helium flow (16 ml /min) streaming over the surface of the melt. It is then carried through a coaxial tube to the target holder and from there, through a 40 m long stainless steel pipe of 1.4 mm inner diameter to a collecting system outside the cyclotron vault.

The target assembly consists of the target, the beam stopper and collimator, the target holder and of the supply pipes for the helium and cooling water (see Fig. 3).

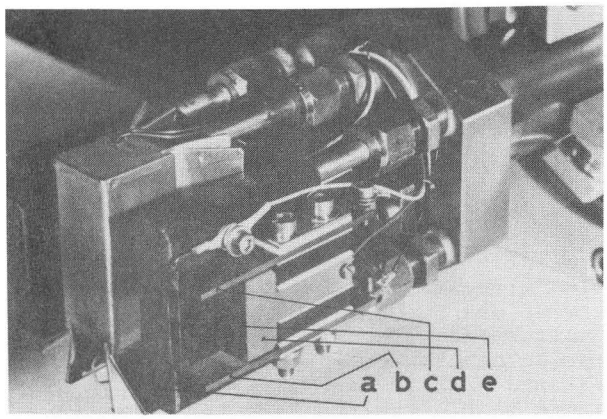

Fig. 3:
The target assembly mounted at the end of the central guide tube. The construction is compact in order to fit in a standard box for the beam scanners and because of space limitations. The beam enters from the right through a window which is limited by three tungsten fingers (a,b,c) and by a copper cheek (d) at the right. The beam is stopped right behind the target. A separate measuring finger (e) is in front of the finger (a) which protects the front wall of the target.

The target and the collimator are fastened at the target holder by Gyrolock couplings to enable precise mounting and easy and fast demounting of the target in a glove box. Three 2 mm thick tungsten fingers together with a cheek of copper on the side remote from the main beam, form a window of 12 mm x 24 mm for the production beam. Right behind the target is the beam stopper. The collimator and the beam stopper are indirectly cooled and electrically isolated. A beam current signal is derived from them, except for the finger protecting the front wall of the target which is directly cooled.

The target is positioned close to the main beam using the current signal on a separate measuring finger, mounted in front of the finger protecting the front wall, then the beam splitter peals off the production beam. Afterwards the target is positioned vertically using the current signal on the lower tungsten finger.

Target unit
The target unit consists of the target assembly which is fixed at the end of a central guide tube, a drive mechanism with high precision spindles and stepping motors. The whole assembly is mounted in a chassis. Fig. 4 shows the target unit attached to a standard box in the beam line on a support. The target is moved vertically by adjusting the support and horizontally by a driven spindle parallel to the central guide tube. The vacuum connection is made by a bellows around the central guide tube. Linear potentiometers and switches marking the end positions are used for control of the movement of the target.

The target is moved into the beam region in two steps: Fast, without position control from the park position to the beginning of the linear potentiometer, then slowly to the wanted position. In case of a fault the target can be moved back quickly into the park position and a valve in front of the box shut, separating the target from the vacuum system of the beam line.

The target unit is characterized by its precision and its freedom from frequent maintenance work. Three target units are used in rotation, in order to keep the dose rate at a tolerable level. A target lifetime of more than 2000 µAh has been achieved.

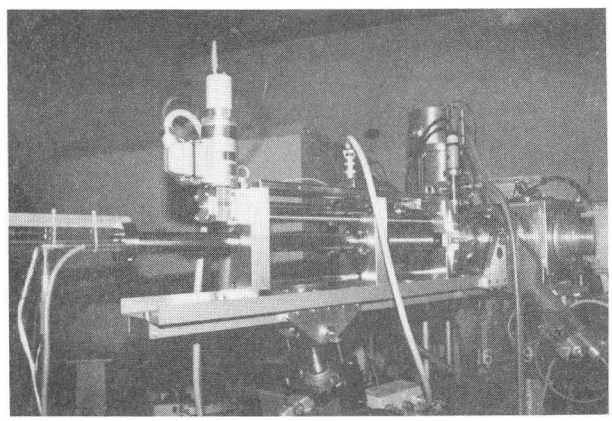

Fig. 4:
The target unit assembled on its chassis and attached to a standard box in the beam line. Between the box and the park position of the target is a valve. The target is vertically positioned by tilting the support around an axis at the location of the bellows between the box and the chassis. The water- and the power supply for the heating filament and the signal cables are fed through the central guide tube to the target holder.

Control system
The different parameters of the production station can be set from a console in the control room or by the cyclotron control computer. The principle of the control system is given in Fig. 5.

Individual items (e.g. power supply for the target heating, drive motors, etc.) are fitted with local control loops. The desired value is written in a 12 bit-DAC and the local loop then takes control (e.g. changes the current in the filament heater or moves the probe, etc.).

The beam intensity on the target is directly controlled by a computer program written in FORTRAN. The current measured at the beam stopper behind the target is converted by a logarithmic current to voltage converter and read by the computer. Taking into account the temperature of the melt T1, the intensity of the main proton beam and its profile the computer calculates and corrects the position of the beam splitter. The program is started by an interrupt generated by:

- a timer every 15 s
- a comparator, if the target current is

CH1996-3/84/0000-0405 $1.00 © 1984 IEEE

higher than the desired value set in a DAC.

- a second comparator, if the target temperature T1 is higher than the maximum allowable one (TMAX), also set in a DAC.

The program also integrates the target current over a production run to give a check on the production efficiency (mCi/μAh).

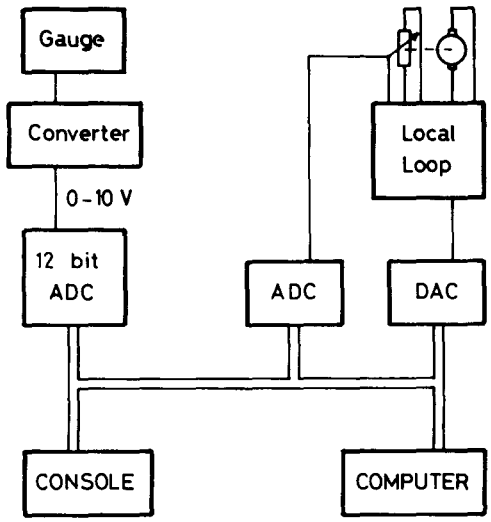

Fig. 5:
The principle of the control system. The signal of a gauge is converted to a voltage (0 to 10 V) then digitised by a 12 bit-ADC. The digital information is read through a parallel data-way (ROAD) by the cyclotron control computer and by the console where it is also displayed.

If either the beam splitter or the target leaves its park position the control system is connected to the operation interlock of the cyclotron. The interlock system is broken when the current of the beam stopper or of a collimator finger increases above a preset value, if the high voltage on the beam splitter drops below a preset value or in case of a break down of the vacuum, one of the power supplies or of the cooling circuit.

The helium flow is controlled by a mass flow meter. If it falls below 10 ml/min an acoustic alarm is given.

Xenon collecting system

The collecting system for the xenon is installed in a shielded box which is at a controlled under pressure. It is attended to the neighbour institute EIR. Coming from the production target, the helium-xenon gas mixture flows through a $Ca(OH)_2$-drier then through liquid nitrogen traps. The xenon is collected in one cold trap for 6 hours which is then bye passed. The maximum I-123 activity is reached after a 4 hour decay period during which time the xenon is collected in the other cold trap. More than 95% of the xenon is frozen out in a single cold trap and a second is provided for safety reasons. After the decay interval the cold trap is warmed up and the remaining xenon is flushed out with hydrogen and also to reduce any iodate to iodide. The iodide is dissolved in 1 to 5 ml of 0.1n-NaOH afterwards.

The helium flows through the trap into a buffer volume where the rest of the radioxenon decays.

The current product rate of I-123 is ~3 Ci/week.

Acknowledgment

A reliable production of I-123 is not imaginable without the excellent contributions of M. Graf and H. Oehninger to the design and construction of the production station. We express our thanks also, to all the groups of SIN and EIR for their valuable help and also thanks to F. Atchison for his criticism and remarks while preparing the paper. Special thanks belongs to H. Willax for his effort in the development of the method.

References

[1] M.C. Lagunas-Solar, H.H. Hines "Proceedings of the Int. Symposium on the Developing Role of short-lived Radionucleides in Nuclear Medicine Practice", May 3-5, 1982, Washington D.C.

[2] W.G. Myers, H.O. Anger, J. of nucl. Med. 3, 183 (1962).

[3] A.W.G. Goolden, H.I. Glass, D.J. Silvester, Br. J. of Radiol. 41, 20 (1967).

[4] M.C. Lagunas-Solar, J.A. Jungerman, N.F. Peek, F.E. Little Proceedings of the 27th Conf. on Rem. Systems Technol. CRSTBJ 27, 1-432, 295 (1979).

[5] A.M.J. Paans, W. Vaalburg, G. van Herk, M.G. Wolding, Int. J. of Appl. Rad. and Isotopes 27, 465, (1976).

[6] M. Diksic, L. Yaffe, J. of Inorg. nucl. Chem. 39, 1299 (1976).

[7] D.B. Syme, E. Wood, I.M. Blair, S. Kew, M. Perry, P. Cooper, Int. J. of Appl. Rad. and Isotopes 29, 29 (1978).

[8] H. Lundqvist, P. Malmborg, B. Langstroem, S.N. Chiengmai, Int. J.of Appl. Rad. and Isotopes 30, 39 (1979).

[9] F. Hegedues, H. Oehninger, L. Rezzonico, H. Willax, 15th Int. Europ. Cyclotron Progr. Meeting, Berlin, 1978.

[10] M. Olivo, U. Rohrer, E. Steiner, IEEE Transaction on Nuclear Science, NS-28, 3094 (1981).

[11] C. Serre, "Evaluation de la perte d'energie unitaire et du parcours de particules chargees traversant un absorbant quelconque", CERN 67-5 (1967)