

Production and Chemical Separation of “No Carrier Added” Iodine-124 from a Reusable Enriched Tellurium-124 Dioxide / Aluminum Oxide Solid Solution Target

著者	Yamazaki H., Funaki Y., Horiuchi Y., Ishii K., Kanai Y., Kikuchi Y., Matsuyama S.
journal or publication title	CYRIC annual report
volume	2006
page range	90-94
year	2006
URL	http://hdl.handle.net/10097/50367

VI. 2. Production and Chemical Separation of “No Carrier Added” Iodine-124 from a Reusable Enriched Tellurium-124 Dioxide / Aluminum Oxide Solid Solution Target

*Yamazaki H.¹, Funaki Y.¹, Horiuchi Y.¹, Ishii K.², Kanai Y.²,
Kikuchi Y.², and Matsuyama S.²*

¹*Cyclotron and Radioisotope Center, Tohoku University*

²*Graduate School of Engineering, Tohoku University*

Introduction

The development of PET from a predominantly neuroscience tool to a routine oncological investigation climaxed about 6 years ago, when PET/CT imaging became clinically available. However, this development had gathered momentum only slowly, as most of radiopharmaceuticals were labeled with short-lived nuclides and could not be distributed. Therefore experience with newly developed radiopharmaceuticals was acquired locally, and the spread of new tracers was hampered.

Iodine-124 is one of alternative long-lived PET nuclides attracting increasing interest. For example, the half-life of 4.2 days allows ¹²⁴I to be used in long-term PET studies of small animals. Further, ¹²⁴I can be used in PET scanners that are located distant from the producing cyclotron facility. Established methods in the chemistry of radioiodinations can be easily translated to ¹²⁴I. In general, ¹²³I and ¹³¹I have been used in conjunction with a gamma camera. The former is short lived but yields reasonable scintigrams with low radiation exposure, while the latter has a longer half-life but yields poor images and entails comparatively high radiation exposure. As regards image quality and radiation exposure, ¹²⁴I is close to optimal. Although there are some limitations such as a low positron abundance (22-25%) and a rather complex decay scheme,¹⁻²⁾ ¹²⁴I is still useful for medical PET imaging³⁻⁴⁾. Some applications of ¹²⁴I in oncology have been reviewed recently⁵⁾. The preferred production route for ¹²⁴I is the ¹²⁴Te(*p,n*)¹²⁴I process⁶⁾. This nuclear reaction leads to good target yields with a ¹²⁴I product of the highest radionuclidic purity⁷⁾. Iodine-125 (*T*_{1/2}=60 d) is the only significant impurity after the decay of ¹²³I (*T*_{1/2}=13 h). The radioactivity level of ¹²⁵I depends on the quality of enriched [¹²⁴Te]TeO₂ target material.

This study details the essential elements for the complete production and separation by thermal processing of iodine-124 in high radionuclidic purity from the low energy, proton irradiation of an enriched tellurium-124 dioxide/aluminum oxide glassy solid solution matrix target.

Material and methods

Chemicals and solvents used were obtained from Aldrich and Wako Pure Chemicals. The enriched ^{124}Te was purchased as tellurium dioxide in 98.5% purity from Trace Sciences International. Radionuclide purity was measured using closed-end coaxial Ge-detector (EURISYS MEASURES, EGPC50-195-R) with PC-based multichannel analyzer and Aptec software. The system was calibrated using ^{152}Eu standard source (Amersham International Limited, QCR.Q8435).

Target design and irradiation conditions

The existing aluminum target holder was modified in order to accommodate a platinum disk with an outer diameter of 20 mm. The disk has a cavity of 10 mm diameter and 1.5 mm depth. The target itself consisted of an admixture of 0.280 grams of tellurium-124 dioxide and 6% by weight of aluminum dioxide. The glassy target matrix was formed at 720°C by allowing the admixture to be melted within the platinum cavity, the base of which had been roughened by random scratches in order to improve the adhesion of the target material. The physical thickness of the target was 350 mg/cm^2 . The disk was covered with an aluminum foil (40 μm thickness) held down by a clamping plate. The target was manually assembled into aluminum target holder, which was compatible with the automated transfer system at the beam line.

We used 14 MeV proton beams of 10 mm spot size. It was calculated using range / energy data (SRIM-2003.26 version) that after passing the aluminum foil the incident beam was 13.7 MeV on the target and the energy decreased 4.0 MeV in the tellurium dioxide of 350 mg/cm^2 thickness. This energy range was also confirmed as being an optimum from our preliminary measurements using natural TeO_2 target of the same surface density. The target was irradiated at 1-4 μA beam current depending on the aimed production yield of ^{124}I .

Dry distillation of radioiodine-124

The distillation apparatus was similar to the one reported by Glaser *et al.*⁸⁾ with slight modification (Fig. 1). Briefly, the target holding part named as the “spoon” was stabilized and served as an inlet for the carrier O₂ gas. The male ground glass joint of the spoon had a frit attached to the main quartz tube. The other end of main tube was reduced to 0.25in tube for connecting to the trap containing 0.5 mL of 0.1 M NaOH. When attached to the furnace, the flat plate at the end of the spoon was positioned at the center of the furnace. The irradiated target was placed on the plate during the dry distillation procedure.

An irradiated target was placed in the distillation apparatus. The flow of O₂ gas was adjusted 12.5 mL/min, and the oven was turned on. Initially the temperature was set at 650°C. Once the oven set point was reached, the temperature was increased to the maximum of 710°C and the maximum temperature was kept for 30 min. The total distillation time was about 75 min. The radioiodine started releasing at around 550°C. The activity in the NaOH trap was monitored by NaI scintillation detector. Once the activity reached a flat level, after 10 min the distillation was stopped by turning off the oven. The gas flow was continued while the oven was cooling below 400°C, and then the trap was transferred to another hot cell for chemical preparation. The distillation efficiencies were 94-97% and the recovery yields of radio distilled were 82-88% (n=4) for irradiated natural TeO₂.

Quality assurance

The iodine-124 production cycle was repeated using tellurium-124 oxide target (admixture of 6 wt% Al₂O₃) of 350 mg/cm² density and 14 MeV proton beam of 2-4 μA. Following the recovery of the iodine-124 solution, an aliquot of the solution was assayed for gross nuclide composition. Figure 2 shows the high resolution gamma spectrum at 95 hours after end of bombardment (EOB). Due to the high isotopic enrichment of the tellurium-124 oxide used for the target preparation, the isotopic composition calculated to 95 hours after EOB was found to be ¹²⁴I at 98% and concurrently produced ¹²³I was 1%. Trace amount of ¹²⁴Sb, which may be produced by a subsequent (*n, p*) reaction of tellurium-124 by neutrons produced in the target matrix, contaminated the recovered iodine-124 solution due to the sublimation at the heating of the production cycle. As the energies of main gamma rays from ¹²⁴Sb are almost same as those from ¹²⁴I, the ¹²⁴Sb content in the recovered iodine-124 solution was determined by checking the decay rate of the radioactivity in the recovered solution. The radionuclide content of ¹²⁴Sb was calculated to be 0.6% at 95 hours after EOB. The gamma spectrum of dry-distilled target after

cooling of 635 hours showed the production of ^{126}I impurity ($T_{1/2}=13\text{ d}$). The radionuclide content of ^{126}I was calculated to be around 0.3% at 95 hours after EOB.

Typical batch yields for iodine-124 were estimated at 380-420 MBq at EOB based on 85% recovery for the dry distillation process. Calculated yields in this target matrix for the production of iodine-124 are in excellent agreement with the literature value of 20 MBq/ μAhr on a thick target of a glassy tellurium-124 oxide target (admixture of 6 wt% Al_2O_3)⁸⁾.

References

- 1) Herzog H.T.L., Qaim S. M., Spellerberg S., et al., Appl. Radiat. Isot. **56** (2002) 673.
- 2) Qaim S.M., Hohn A., Bastian T., et al., Appl. Radiat. Isot. **58** (2003) 69.
- 3) Pentlow K.S., Graham M.C., Lambrecht R.M., et al., Med. Phys. **18** (1991) 357.
- 4) Pentlow K.S., Graham M.C., Lambrecht R.M., et al., J. Nucl. Med. **37** (1996) 1557.
- 5) Glaser M., Luthra S.K., Brady F., Int. J. Oncology **22** (2003) 253.
- 6) Scholten B., Kovacs Z., Tarkanyi F., et al., Appl. Radiat. Isot. **46** (1995) 255.
- 7) Bastian T., Coenen H.H., Qaim S. M., Appl. Radiat. Isot. **55** (2001) 303.
- 8) Glaser M., Mackay D.B., Ranicar A.S.O., Radiochim. Acta. **92** (2004) 951.

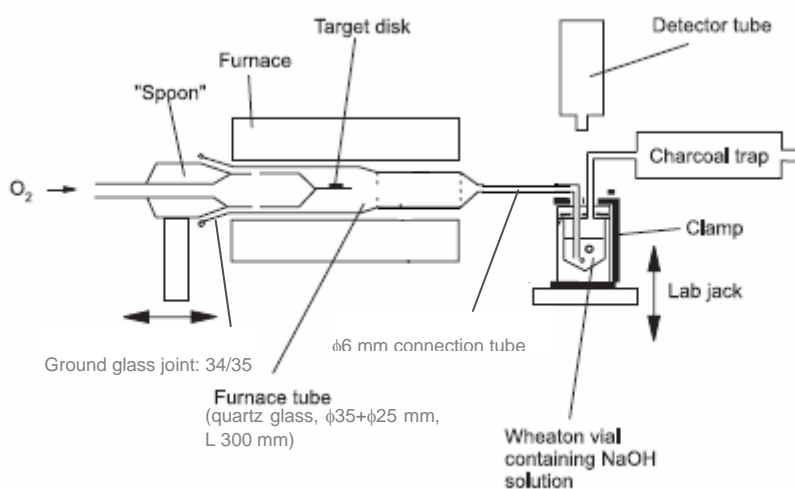


Figure 1. General scheme showing a cross-section of ^{124}I distillation apparatus.

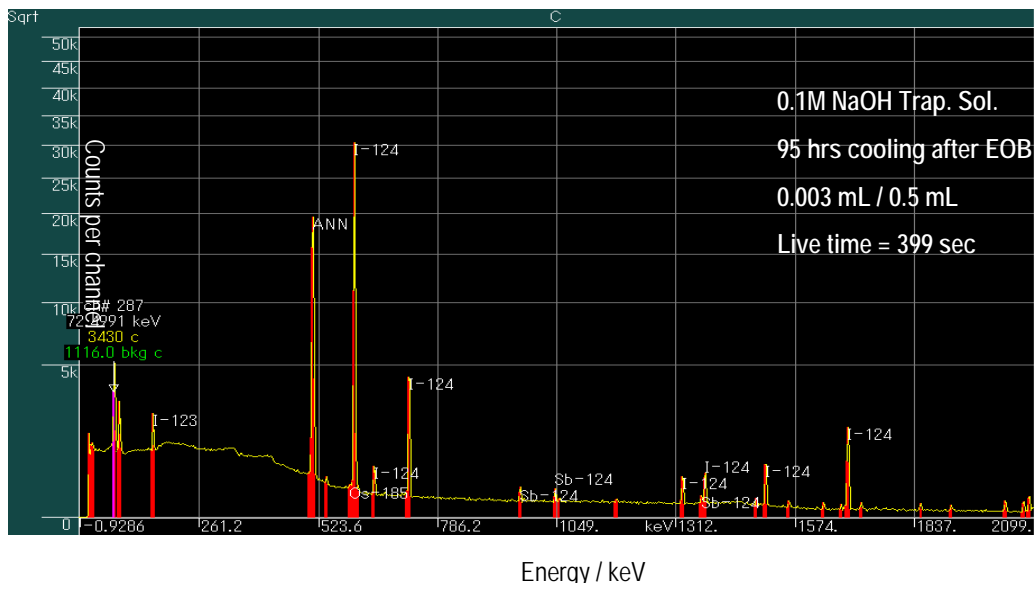


Figure 2. Gamma spectrum of the recovery of iodine-124 in a 0.1 M NaOH trap.