PRODUCTION OF LONGER-LIVED POSITRON EMITTERS ^{73}Se, ^{82m}Rb AND ^{124}I

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Some new aspects of production of longer-lived β^+ emitters ^{73}Se ($T_{1/2}$ = 7.1 h), ^{82m}Rb ($T_{1/2}$ = 6.5 h) and ^{124}I ($T_{1/2}$ = 4.15 d) are described. For production of ^{73}Se via the ^{75}As(p,3n)-process (E_p = 38 → 30 MeV) a Cu_3As-alloy was irradiated in the internal target system of the injector for COSY, and ^{73}Se was separated via thermochromatography. ^{82m}Rb was produced via the ^{82}Kr(p,n) process using 90% enriched ^{82}Kr at E_p = 14.5 → 10 MeV. An efficient irradiation and target gas recovery system was developed. The ^{82m}Rb activity deposited on the inner target walls was removed by steam. For production of ^{124}I the ^{124}Te(p,n) process (E_p = 13 → 9 MeV) was investigated. A 4π water-cooled $^{124}TeO_2$ target system was used and ^{124}I was separated via dry distillation. All three radioisotopes were obtained in high purity and quantities sufficient for medical applications.

1 Introduction

In recent years the Positron Emission Tomography (PET) has been gaining considerable significance as a diagnostic tool in nuclear medicine. Large amounts of the most commonly used β^+ emitters, namely ^{11}C ($T_{1/2}$ = 20 min), ^{13}N ($T_{1/2}$ = 10 min), ^{15}O ($T_{1/2}$ = 2 min) and ^{18}F ($T_{1/2}$ = 110 min), can be easily produced[1] using dedicated small cyclotrons (E_p ~ 17 MeV; E_d ~ 10 MeV; or even proton only machines with E_p ~ 11 MeV). On the other hand, there are several less commonly used and potentially useful β^+ emitters which are of considerable importance. The longer-lived β^+ emitters are of interest for satellite PET, i.e. they can be transported from the site of production to PET centres without cyclotrons. The present work deals with some new aspects of production of three longer-lived β^+ emitters, viz. ^{73}Se, ^{82m}Rb and ^{124}I.

2 Production of ^{73}Se

The radioisotope ^{73}Se ($T_{1/2}$ = 7.1 h; E_C = 35%, β^+ = 65%; $E_{\beta+}$ = 1.32 MeV) is an interesting β^+ emitting sulphur analogue. For its production several methods have been suggested, e.g. (p,3n) and (d,4n) reactions on ^{75}As [2-5] or ^{3}He and α-particle induced reactions on Ge isotopes[2, 5-8]. The method of choice, however, is the ^{75}As(p,3n)-process over the energy range of E_p = 40 → 30 MeV[3, 7]. Several types of target materials have been described in the literature, with special emphasis on As_2O_3. We made use of a Cu_3As-alloy which was developed earlier for the production of $^{75-77}$Br [9].

The alloy was placed on a polished Cu backing sheet (1 mm thick), melted in an oven under helium flow at 900°C and then cooled down rapidly. The thickness of the alloy layer was about 160 μm, corresponding to the energy range E_p = 38 → 30 MeV while using a beam angle of 10°. The target sheet was then bent and clamped into a target head to be used whithin the internal irradiation system at the cyclotron JULIC (injector for COSY - Jülich) (see Fig. 1). The target sheet was cooled by flowing water only from one side. The H_2^+ beam (E = 76.2 MeV) fell on the target at a grazing angle of 10°. At the target surface the beam was instantly converted to 38.1 MeV protons of double intensity. Irradiations were done for 2h at beam currents of about 20 μA. Due to the very high level of radioactivity, the removal of the target from the irradiation position for subsequent transfer to the chemical laboratory was initiated after a decay time of about 2 hours after the end of the bombardment (EOB).

The separation of radioselenium from the irradiated target was effected via thermochromatography. It was observed[10] that on heating the irradiated Cu_3As-alloy at 1000°C in a stream of O_2, both arsenic and selenium are removed from the target and are deposited at approximately the same position (cf. Fig. 2(A)). However, a detailed investigation of the individual removal behaviour of the two elements as a function of oven temperature showed that a separation is possible (cf. Fig. 2(B)). We therefore developed a two step thermochromatographic separation procedure. In the first step the temperature was maintained at 660°C for 40 min whereby the [^{74}As]As_2O_3 formed was deposited about 7 cm from the end of the oven. Thereafter the quartz tube was shifted back till the [^{74}As]As_2O_3 zone was in the middle of the oven. The temperature of the oven was then raised to 750°C for about 5 min whereby [^{74}As]As_2O_3 was shifted further by 12 cm. The quartz tube was now moved to the original position and the second stage of thermochromatography started. The temperatures used successively were: 900°C (7 min), 1000°C (15 min) and 1100°C (10 min). The radioselenium was deposited about 11 cm away from the end of the oven. The quartz tube was then taken out, rinsed with 6 ml of warm 6 M HCl whereby > 98% of the radioselenium was dissolved. The overall radiochemical yield was 85%.

The thick target yield of ^{73}Se was found to be 150 MBq (~ 4 mCi)/μAh and the batch yield 6 GBq (160 mCi) at EOB. The $^{72, 75}$Se impurities amounted to < 0.05% of ^{73}Se. The procedure is presently being automated.

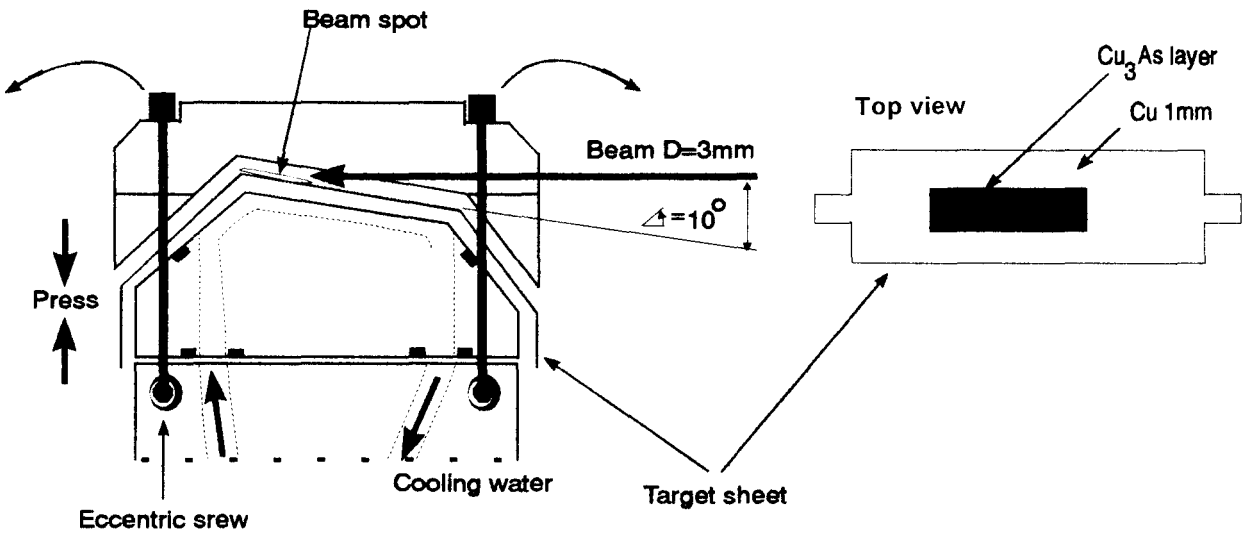
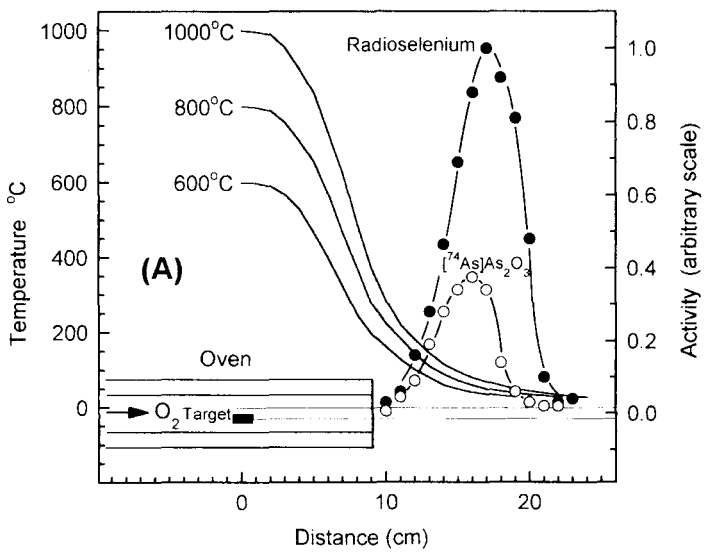


Fig. 1 Probe head loaded with the target sheet for use in the interal irradiation system at the injector of COSY.



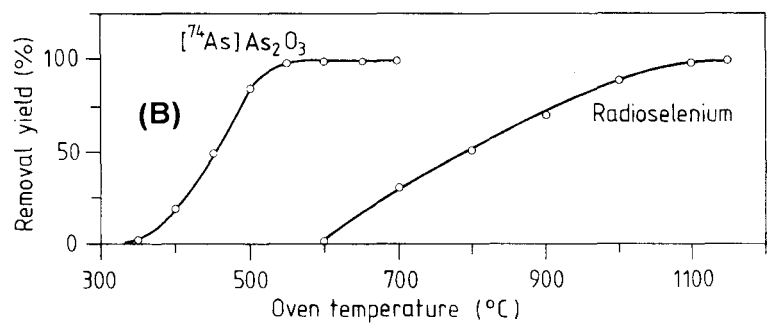


Fig. 2
(A) Deposition zone of $[^{74}As]As_2O_3$ and radioselenium in an O_2 atmosphere for an oven temperature of > 1000°C.
(B) Removal of $[^{74}As]As_2O_3$ and radioselenium from the Cu_3As-target in an O_2 stream as a function of oven temperature.

3 Production of ^{82m}Rb

For myocardial blood flow studies using PET four competing tracers, namely [^{13}N]NH_4^+, $H_2{}^{15}O$, ^{38}K and ^{82}Rb, are commonly used. The first three agents are produced at the site of application. In PET centres without a cyclotron ^{82}Rb ($T_{½}$ = 1.2 min), obtained from a ^{82}Sr - ^{82}Rb generator system, is commonly used. Due to the relatively high cost of the generator, we proposed the longer-lived β^+ emitting ^{82m}Rb ($T_{½}$ = 6.5 h; EC = 74%, β^+ = 26%; E_{β}+ = 0.80 MeV) as a possible substitute[11]. Cross section measurements on highly enriched ^{82}Kr showed that the $^{82}Kr(p,n)^{82m}Rb$ process over the energy range E_p = 14.5 → 10 MeV is ideally suited for the production of ^{82m}Rb.

We now developed a target system at the compact cyclotron CV 28 which allows high current irradiations and an efficient recovery of the 90% enriched ^{82}Kr target gas. A schematic representation of the system is given in Fig. 3. Some important components like the vacuum system and the Kr safety circuit are only indicated in the diagram. More details are given on the target body used for irradiations, together with the accessories for recovery of the target gas and removal of the radioactive products from the target walls. The target body consists of a conical cylinder made of Ni (12 cm long; front Ø = 1.5 cm; beam stop Ø = 3.0 cm) with two Ni foils (each 30 μm thick) as a double window, through which He cooling gas flows. The total volume of the target is 52 ml. The target is filled to a pressure of 3 bars and irradiation is done with 14.8 MeV protons at 10 μA. After the irradiation the enriched target gas is recovered by cryopumping. Thereafter steam is led into the target chamber, allowing an efficient removal of the ^{82m}Rb activity deposited on the inner walls of the target. Finally, the activity collected in a small volume of water is mixed with saline solution and, after sterile filtration, is ready for application. The thick target yield of ^{82m}Rb amounts to 370 MBq (10 mCi)/μAh and the only radionuclidic impurity detected is ^{83}Rb (0.04%).

4 Production of ^{124}I

The radioisotope ^{124}I ($T_{½}$ = 4.15 d; EC = 75%, β^+ = 25%; E_{β}+ = 2.13 MeV) is the only longer-lived β^+ emitting radioisotope of iodine and has found both diagnostic[12] and therapeutic[13] applications. In particular for the latter, it has been used for labelling some biomolecules, specific for tumour research with PET. For its production, the $^{124}Te(d,2n)^{124}I$ process over the energy range of E_d = 16 → 9 MeV is commonly used.[14, 15] We performed detailed measurements on the excitation function of the $^{124}Te(p,n)^{124}I$ reaction[16] which had not been investigated well below 12 MeV[17]. Our data showed an energy shift of about 2.5 MeV compared to the literature values[17], thereby suggesting that the $^{124}Te(p,n)^{124}I$ reaction should be suitable for production of ^{124}I at a small cyclotron. A thick $^{124}TeO_2$ target (99.9% enriched; wt. = 0.18 g/cm^2), similar to the one used earlier for ^{123}I production,[18] was irradiated in a 4π water-cooled system with 15 MeV protons at nominal currents of 10 μA. The on-target proton energy was E_p = 13 → 9 MeV. After irradiation the radioiodine

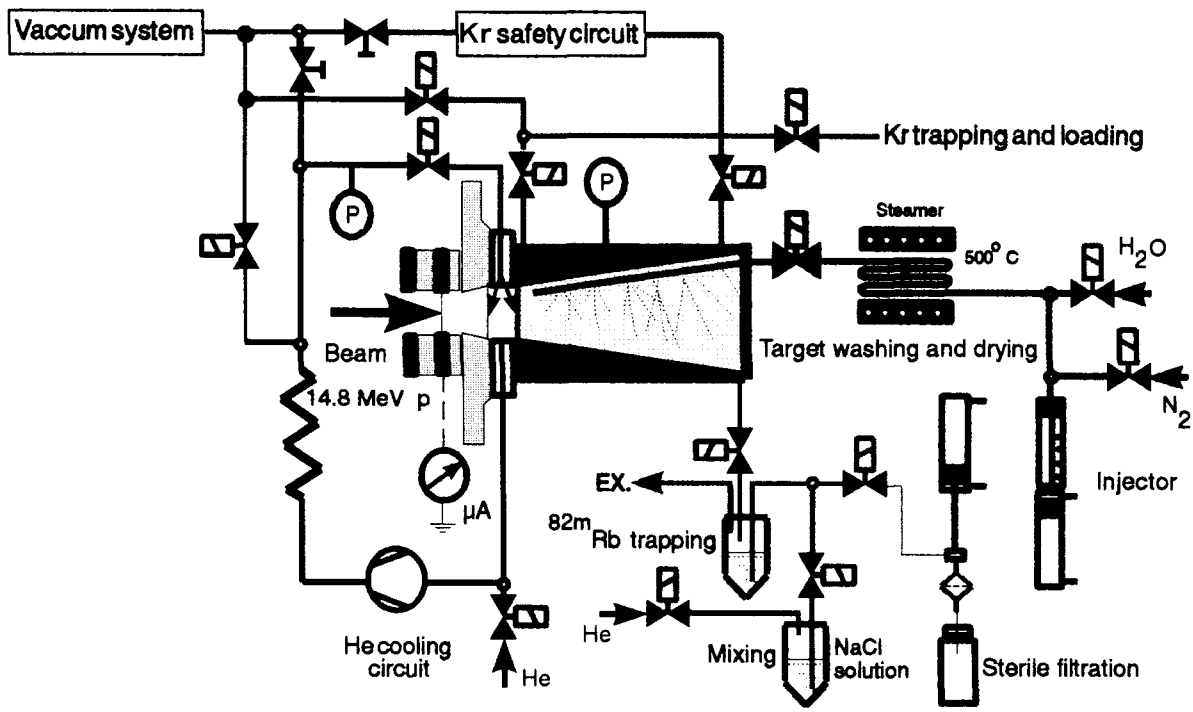


Fig. 3 Schematic representation of the target system developed for irradiation and safe recovery of highly enriched ^{82}Kr gas, and efficient removal of the ^{82m}Rb activity by steam.

was separated by the standard dry distillation method[18]. The thick target yield of ^{124}I amounted to 12 MBq (0.3 mCi)/μAh. This corresponds to about 50% of the theoretical value. A 3 hour irradiation at 10 μA led to a batch yield of 370 MBq (10 mCi) ^{124}I at EOB. The major impurity was ^{123}I but it was not a serious problem. Due to its shorter half-life it decayed out within about two days. Presently efforts are underway to increase the batch yield of ^{124}I.

Conclusion

The results reported here on the production of three radioisotopes should demonstrate that constant efforts are necessary to develop new production routes for potentially useful longer-lived β^+ emitters. Several types of accelerators and interdisciplinary work are involved.

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