RADIOLOGICAL PROTECTION ASPECTS OF ^{123}I PRODUCTION

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With the Eindhoven AVF cyclotron ^{123}I for application in nuclear medicine is now routinely produced in quantities of 4 to 20 GBq per batch. Enriched telluriumdioxide on a platinum backing is irradiated with 25 MeV protons. The production reaction is $^{124}Te(p,2n)^{123}I$. Beam currents are used in the order of 20 μA.
The radiochemical separation of iodine from tellurium is carried out by heating the telluriumdioxide just above its melting point for only a few minutes. During this procedure the total amount of ^{123}I is handled as vapour in a quartz tube (1).
During the experimental stage of the project a radiological safety program was developed in close collaboration with the workers. This program was considered to be an integral part of the total production project. The radiological safety program implies normal working conditions, failure analyses and emergency procedures.

SHIELDING AT THE IRRADIATION SITE

The part of the cyclotron hall with the irradiation site is shown in fig. 1. Primal shielding is provided by a concrete wall of 1.5 m thick. The concrete roof is 0.4 m thick. Radiation levels at different locations in the cyclotron hall are permanently measured with a monitoring system for γ-rays and neutrons. The alarm levels of the detectors at the inside of outer (glass) wall correspond to a derived working limit of 2.5 μSv/h for non-supervised areas. Exceeding of the alarm levels will result in automatic interruption of irradiation (with a short delay time for adequate action by the cyclotron operators). Even with additional neutron shielding with 0.4 m paraffin around the production position, the proton beam current was restricted to less than 3 μA. Since neutrons contribute dominantly to the dose rates we paid much attention to neutron measurements. At several places under different shielding conditions neutron energy spectra were measured using a self-developed multisphere technique (2). It was shown that sky shine of neutrons passing through the roof and scattering in the labyrinth caused a major part of the dose rate outside the irradiation facility. In fig. 2 some of the measured neutron spectra are shown. In addition corresponding values of the flux density, mean energy and dose equivalent rate are given in table 1. For neutrons passing through the roof it was found that additional local shielding with 0.2 m paraffin reduced the integral neutron flux density by a factor of about seven. The mean energy of the transmitted neutrons increased with a factor of 2. A minor modification of the labyrinth lay out reduced the contribution from scattered neutrons. At present the improvements in shielding allow for beam currents of 20 μA, which corresponds to a ^{123}I production rate of about 10^{10} Bq. It is noted here that a reasonable high production rate is required not only to shorten the expensive operation time of the cyclotron but also to

Table 1. The neutron fluence rate, the mean neutron energy and the neutron dose equivalent rate at the positions A, B and C, marked in Fig. 1. Notation B_1 denotes without paraffin, B_2 denotes with paraffin. The proton beam current is 20 μA.

Position	Neutron flux density $[m^{-2}s^{-1}]$	Mean energy [MeV]	Dose equivalent rate [rem/h]	[mSv/h]
A	$3.6\ 10^{10}$	0.56	68	680
B_1	$5.6\ 10^{8}$	0.69	1.1	11
B_2	$8.6\ 10^{7}$	1.21	0.21	2.1
C	$8.6\ 10^{7}$	0.08	0.06	0.6

restrict the ^{124}I content to a low level (< 1%).
Radiation exposure during target handling after irradiation is mainly caused by γ-rays from ^{194}Au in the target support. The dose equivalent rate at 0.3 m distance from the target is about 15 mSv/h when 37 GBq of ^{123}I is produced. The irradiation set-up is at one side shielded by 0.05 m lead. The dose rate just behind the lead wall is reduced to 250 μSv/h. At working distance dose equivalent rate is less than 100 μSv/h. Operated from behind the shielding the target is removed from the irradiation position with handling tools and put into a transport container.

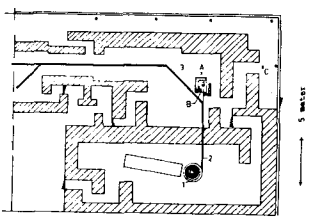

Fig. 1 Part of the cyclotron hall with the ^{123}I production site (to scale). (1) cyclotron; (2) beam transport system; (3) space for radionuclide production; (a) irradiation facility for solid targets e.g. for the production of ^{123}I, ^{77}Br and ^{87}Y; (A), (B) and (C) denote positions where the neutron spectrum and neutron dose equivalent during ^{123}I production have been determined. Position B is on the roof, 3.85 m above ground level; ● radiation detectors (alarm system).

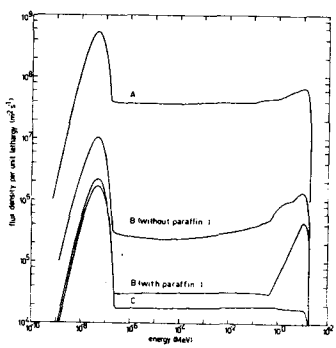

Fig. 2 Neutron energy spectra at the positions A, B and C, marked in Fig. 1. The flux density per unit lethargy as a function of the energy is given (lethargy = ln E/E_0, where E_0 stands for 10 MeV and E for the neutron energy in MeV). The proton beam current is 1 μA.

IRRADIATION SET-UP

Since the radio-iodine can be liberated from the TeO_2 target material by volatilization at temperatures above 500 °C safety precautions are necessary to minimize the risk for air contamination and consequent internal contamination of personnel. These safety measures basically are two fold: At first target temperatures must be limited and secondly containment must be provided in case radio-iodine escapes. The irradiation set-up is sketched in fig. 3. By defocussing the proton beam, hot spots on the target which may cause problems with regard to temperature control are prevented. The platinum target support is cooled by a forced water flow. The irradiation is automatically inter-

rupted when the flow rate falls below 4.5 liter per minute. The cooling system and target holder were designed for a 5 cm^2 proton beam cross section and beam current density upto 30 $\mu A/cm^2$. To minimize the consequences of accidental volatilization the TeO_2 layer is locked up in an air-tight target holder. The front side of the target holder is a 10 μm thick tantalum foil through which the proton beam passes. A second tantalum foil is placed at the end of the beam pipe to seal its vacuum. It also is a secondary barrier to prevent contamination in the beam pipe. Loss of vacuum is detected and will automatically stop the irradiation process. The entire irradiation set-up is mounted in an air-tight glovebox. The leakage rate of the box for helium gas was measured to be less than 0.1% of the total volume per hour. The air in the box is at ambient pressure and is recirculated continuously over a charcoal filterbed with a trapping efficiency for iodine of at least 80%. The recirculation rate is 1.5 liters per second which corresponds to 1/3 of the total box volume per minute.

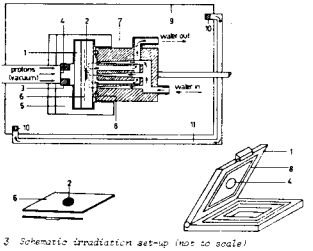

Fig. 3 Schematic irradiation set-up (not to scale) (1) target holder; (2) platinum gauze with TeO_2; (3) diaphragm; (4) tantalum foil; (5) target holder housing; (6) platinum target support; (7) target cooling; (8) O-shaped rubber sealing ring; (9) Perspex box; (10) charcoal filter; (11) recirculation system.

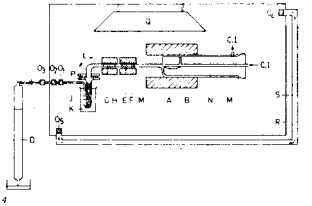

Fig. 4
Apparatus for the separation of radioactive iodine from the target. (A) Platinum support with TeO_2; (B) Tubular oven (740° C); (C) Air flow (60 ml/min); (D) Hydrostatic water column; (E+F) Aluminium-oxide absorber; (F) Tubular oven (210° C); (H) Tubular oven (700° C or more than 820° C); (I) Nitrogen gas flow (40 ml/min); (J) Recipient with glass beads; (K) Liquid nitrogen (not used for routine production); (L) Heating ribbon; (M) Outer quartz tube (fixed); (N) Inner quartz tube (movable); (O_1) Charcoal filters; (P) Magnetic flanges; (Q) Water cooling system (for emergencies air cooling can be used) (R) Perspex box; (S) Recirculation system;

THE RADIOCHEMICAL SEPARATION SET-UP

To reduce external radiation exposure the separation process is operated from behind a 0.05 m thick lead shielding. Handling tools were constructed for all types of manipulations including the input of the target from the container into the apparatus, the displacement of ovens, as well as the filling of the-ready-for-transport glass capsules. The dose equivalent rate just behind the lead shielding ranges from 20-50 μSv/h. The separation technique is described in detail elsewhere (1). The apparatus is shown schematically in fig. 4.
During operation the apparatus is kept at reduced pressure which is maintained by the hydrostatic water column (maximum pressure reduction 0.6 m water). Before the start of each separation procedure the entire apparatus is tested for leakage. During separation the iodine liberated from the target due to heating, is forced to flow to the recipient containing glass beads (J). The trapping here is about 75%. The rest of the iodine will be trapped in the charcoal filters (O_1, O_2). The third filter (O_3) is permanently monitored for radio-iodine content. Radioactivity in here has never been detected. The carrier gas is not released but collected in the upper part of the hydrostatic water column. In analogon with the irradiation set-up a perspex glovebox is provided

for reasons of containment in case of iodine escaping out of the separation apparatus. In this box the air is recirculated via filters (O_4, O_5) at a rate of 1.5 liter per second which corresponds to 1/6 of the total box volume per minute. Here also the trapping efficiency is about 80% per passage.
Since heating in three different ovens is essential in the chemical separation process there is a risk for the perspex box to melt, with consequent risk for loss of containment. As safety measure a water cooling system is provided above the ovens. As an additional safety precaution it is possible to turn over to air cooling in case of system failure.

AIR MONITORING

For continuous measurement of the radio-iodine in air outside the glovebox an air monitoring system was developed. Via a maximum number of 6 inlets air is sampled with a flow rate of $2.8\ 10^{-3}$ m^3/s from "critical" positions just outside the glovebox. A GM-end-window tube type 18546/01 is used as detector just above the coal filter assembly (trapping efficiency > 95%). The detection efficiency is $3\ 10^{-2}$ cps/Bq for ^{123}I. Based on the respective DAC values being $9\ 10^4$ Bq/m^3 for ^{123}I and $1\ 10^3$ Bq/m^3 for ^{124}I and considering that the ^{124}I content is about 0.8% at EOB, the weighted value for the effective DAC was taken to be equivalent to $5\ 10^4$ Bq/m^3 of ^{123}I.
The time derivative of the count rate is simply proportional to the radio-iodine concentration in the sampled air (expressed in units DAC). Since the maximal count rate corresponds to the total accumulated activity it is a measure for the upper level of the potentially inhaled radioactivity. The count rate is recorded during the entire separation process. Adjustment of the air flow rate resulted in a simple relation between the count rate R and the upper level for intake $I = 10^{-8}R \times ALI$. It was shown that concentration at 0.1 DAC level can be detected within 25 seconds.
In three years of production only two times a small air contamination was measured at a level of 2 × DAC. Both cases were due to improper handling of the capsules with the glass beads.

FINAL REMARKS

Just in case of extreme system failure: coincident loss of containment and breakage of separation quartz tube an emergency procedure is developed as a result of which the accidental intake by personnel reasonably can be expected to be less than 0.1 × ALI.

The results of personal dosimetry measurements show that for the group of 5 workers the accumulated collective dose equivalent was less than 5 mSv for a production of 300 GBq in a period of three years.

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