PRODUCTION OF ^{208,209,210}At VIA ^{209}Bi(^{3}He,xn) PROCESSES

F. Szelecsényi, Z. Szűcs, O. Solin*, J. Bergman* and S.-J. Heselius*
Institute of Nuclear Research of the Hung. Acad. of Sciences, H-4001 Debrecen, Hungary
**Abo Akademi, Accelerator Laboratory, SF-20500 Turku 50, Finland*

There is a growing interest in astatine radioisotopes from the standpoint of radiobiological and therapeutical application, particularly with respect to the use of astatine-labelled monoclonal antibodies. On the other hand, both the organic and inorganic aspects of the chemistry of astatine stimulate a continuing interest, especially in connection with the formation of complexes and stability of ionic compounds [cf.1]. Due to the very weak gamma- rays of the medically significant ^{211}At ($T_{1/2}=7.2$ h), developmental chemistry is best accomplished with other astatine isotopes (i.e. ^{208}At $T_{1/2}=1.63$ h; ^{209}At $T_{1/2}=5.41$ h; and ^{210}At $T_{1/2}=8.1$ h) which are more suitable for gamma-ray spectroscopy.

It is known that astatine is produced directly via the irradiation of bismuth with alpha particles. However, this method of production demands a high energy accelerator with $E_\alpha > 30$ MeV [cf.2]. The aim of the present study was to investigate the production possibilities of different astatine isotopes using a multiparticle compact cyclotron. Taking into account the parameters of our cyclotrons and the threshold energies of the possible charged particle induced reactions leading to At isotopes, the ^{209}Bi(^{3}He,xn) processes appeared most suitable.

Excitation functions were measured by the activation method using the stacked-foil technique. High purity bismuth prepared via evaporation of Bi on natural copper foils were used as target materials. The target stacks were irradiated in the external beam of the Turku MGC cyclotron with 28 MeV incident ^{3}He-particle energy. The average beam current on the targets was determined using a Faraday-cup and via ^{63}Cu(^{3}He,αn)^{61}Cu and ^{65}Cu(^{3}He,2n)^{66}Ga monitor reactions induced on Cu backings [3]. The activity of the irradiated samples was determined by standard Ge(Li) detector gamma-ray spectroscopy. The experimental technique and the data evaluation were similar to [4]. The total estimated errors in the cross sections are 12-17 %.

The measured excitation functions covering an energy range of 15 to 28 MeV are shown in Fig.1 together with the only previously reported values [5]. In the case of ^{209}Bi(^{3}He,3n)^{209}At reaction the present work shows good agreement with the data of Storm but for the ^{209}Bi(^{3}He,2n)^{210}At reaction, our values are significantly higher than the earlier published ones. Our presented excitation function shows a maximum of 21 mb at 24 MeV for ^{210}At production. The cross section data of Storm around 29 MeV (~165 mb) support our values measured at lower energy regions for ^{209}Bi(^{3}He,4n)^{208}At reaction. The cross section data of ^{209}Bi(^{3}He,n)^{211}At were not measured. Metallic bismuth targets of 0.25 g/cm^2 melted on Cu backings were used for thick target yield measurement. The integral thick target yields of ^{209}Bi(^{3}He,xn)^{208,209,210}At reactions calculated from the excitation functions are shown in Fig.2. The experimental thick target yields which were obtained with low beam intensities for checking the excitation functions are in good agreement with the calculated values. For separation of astatine from the bismuth target a dry-distillation method was used with 85 ± 5% overall yield [cf.6]

On the basis of our data the ^{209}Bi(^{3}He,3n)^{209}At reaction is the method of choice for the production of ^{209}At for in-house use at low energy cyclotrons.

1. Rössler K., Radiochimica Acta 47, 1 (1989)
2. Beyer G.J., Dreyer R., Oldrich H., Rösch F., Radiochem. Radioanal. Lett. 47, 63 (1981)
3. Bryant E.A., Cochran D.R.F., Knight J.D., Phys. Rev. 130, 1512 (1963)
4. Tárkányi F. Szelecsényi F., Kopecký P. Int. J. Apl. Radiat. Isotopes 42, 513 (1991)
5. Storm A., Report UCRL-9732 (1960)
6. Szűcs Z., Szelecsényi F., Izotóptech. Diagn. 33, 221 (1990)

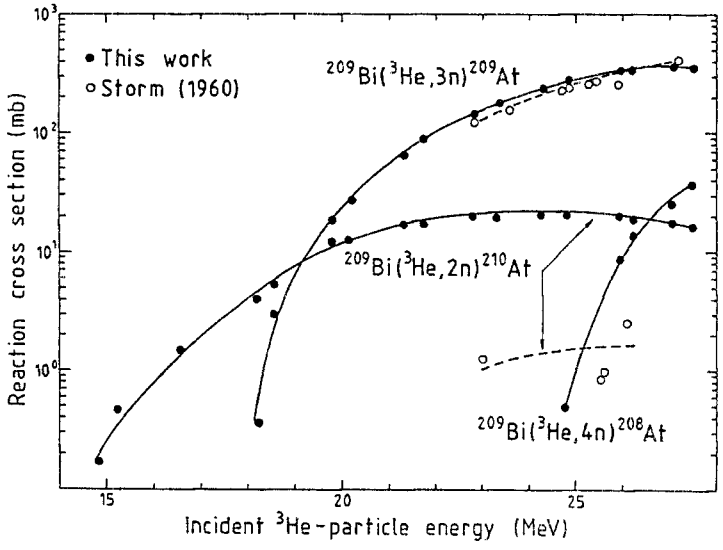


Fig. 1. Excitation functions for the formation of ^{208,209,210}At in ^{3}He-particle induced nuclear reactions on ^{209}Bi

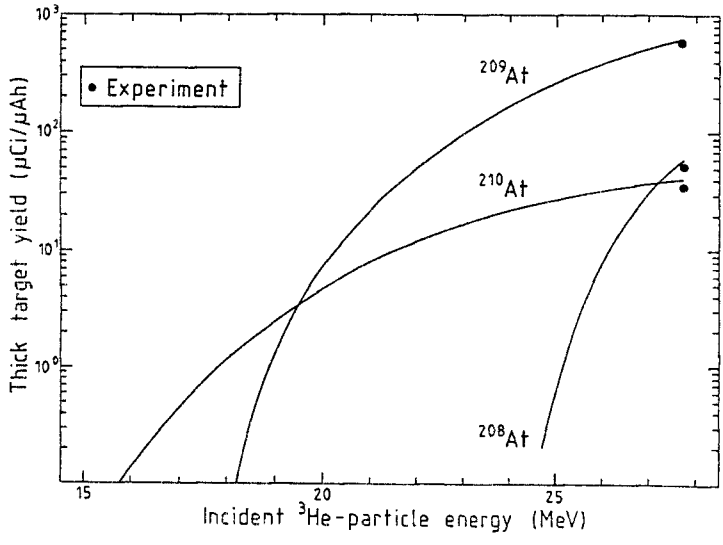


Fig. 2. Calculated thick target yields of ^{208,209,210}At in ^{3}He-particle induced reactions on natural Bi as a function of the incident ^{3}He-particle energy

This work was supported by the Hungarian Academy of Sciences and the Academy of Finland.

Paper J2

PRODUCTION OF BROMINE-75. DOSIMETRIC REHABILITATION OF THIS PET TRACER.

Cl. BRIHAYE, M. GUILLAUME, J. TRUMPER*, J.C. DEPRESSEUX, D. COMAR
Université de Liège, CRC B30, B-4000 Liège (Belgique).
*Soreq NRC, Radiopharm. Dpt. 70600 YAVNE (Israel).

Among PET tracers, Bromine-75 ($T_{½}$ 95.5 min) has had a bad press due to the fact that during its production, medium-lived Bromine-76 ($T_{½}$ 16.1 h) is also produced, which is theoretically unfavorable with regard to dosimetry. As this argument does not take into account the ^{76}Br activity administered nor the kinetics of biological excretion of the radiopharmaceutical, it is not objective and to rehabilitate Br-75 as a suitable tracer for PET, we calculated the radiation absorbed doses resulting from the administration of hypothetical meta-bromobenzyl-guanidine labelled with ^{75}Br or ^{76}Br. For that purpose, we considered the biological data of the distribution of ^{131}I-meta-iodobenzyl-guanidine (MIBG) in female dogs published in extensoby D.P. SWANSON et al(1). The MIRD method was followed, and the S factors, not available in the literature, were calculated from the decay schemes of ^{75}Br and ^{76}Br (2). The results, shown in Table 1, are compared to the radiation absorbed doses for hypothetical ^{18}F-meta-fluorobenzyl-guanidine and ^{131}I-MIBG. Even if the contribution to the absorbed dose from ^{76}Br is important, the values of the effective dose equivalents H_E suggest that, when the ^{76}Br contamination is restricted to 10-15 %, the total dosimetry for 370 MBq of ^{75}Br is of the same order of magnitude as those for other PET or single-photon medical examinations.

Br-75 can be prepared by the $^{76}Se(p,2n)$ or $^{75}As(^{3}He,3n)$ reactions (3-4). The first one requiring enriched ^{76}Se was not considered suitable for routine production. The methods for the separation of bromine from Arsenic targets have involved a dry distillation in a sophisticated set-up (4) or the distillation as Br_2 after dissolution of the As target and oxidation of the Br^- ions to Br_2 (5). The method we present here is a variation of the wet distillation of Br_2, which has the advantages of being simple and easily remotely controlled. The target is made of anhydrous Na_3AsO_4 (m.p. 1200°C), which is very soluble in water. Discs of 0.1 mm thickness are easily prepared by fritting Na_3AsO_4 under pressure in Cu backings. From the published excitation functions (4), we calculated the differential production yields for the reactions $^{75}As(^{3}He,3n)^{75}Br$ and $^{75}As(^{3}He,2n)^{76}Br$ and for ^{3}He energies ranging from 30 to 0 MeV. Under our production conditions, we deduced theoretical ^{75}Br and ^{76}Br yields of 37.4 and 2.59 MBq/µA-h respectively. Experimental yields of 22.9 and 1.78 MBq/µA-h were measured. A scheme of the set-up is shown in Figure 1. After irradiation, the Cu backing with Na_3AsO_4 is introduced into a beaker, and the arsenate is dissolved in 1 M NaOH. The solution is transferred by aspiration into a reactor, where H_2SO_4 and $KMnO_4$ are added to the solution. Br_2 is distilled by bubbling Argon through the solution and trapped by reduction with SO_2 vapour in equilibrium above a drop of saturated SO_2 solution. After distillation (15 min), the glass tube with the SO_2 solution and the radioactive bromide ions is washed with 1 ml of water. The solution is finally evaporated to dryness.

references

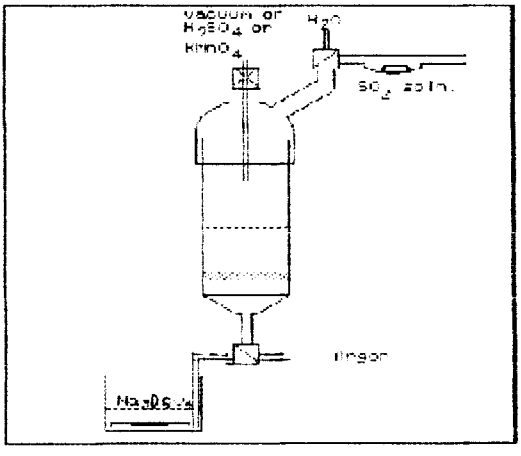
(1) D.P. SWANSON, J.E. CAREY, L.E. BROWN et al.
Third International Radiopharmaceutical Dosimetry Symposium, Oak Ridge, 1980, p. 213-224.
(2) ICRP

(3) Z. KOVACS, B. BLESSING, S.H. QUAIM et al.
Int. J. Appl. Radiat. Isot. 36 (1985) 635-642.
(4) A.M. PAANS, J. WELLEWEERD, W. VAALBURG et al.
Int. J. Appl. Radiat. Isot. 31 (1980) 267-273.
(5) C. LOC'H.
Mémoire. CNAM, Paris, 1988.

Table 1. Absorbed doses (mGy/MBq)

Organ	^{75}Br-BG	^{76}Br-BG	^{18}F-BG	^{131}I-BG
gonads	0.015	0.27	0.018	0.22
red marrow	0.011	0.13	0.013	0.054
lungs	0.027	0.34	0.035	0.17
adrenals	0.070	2.16	0.10	4.68
heart	0.027	0.38	0.030	0.15
liver	0.025	0.25	0.030	0.11
kidneys	0.032	0.31	0.041	0.14
pancreas	0.046	0.57	0.059	0.27
spleen	0.046	0.98	0.062	0.65
H_E(mSv/MBq)	0.022	0.40	0.027	0.71

Figure 1. Apparatus for distillation of ^{75}Br (^{76}Br)



Cyclotron production of high-purity Tc-94m by *in situ* sublimation

RJ Nickles, BT Christian, AD Nunn*, CK Stone. Medical Physics Department, University of Wisconsin, Madison WI and Bristol-Myers Squibb, New Brusnwick, NJ.

Positron emission tomography (PET) provides a quantitative ($\mu Ci/cm^3$) image of the *in vivo* activity distribution in transverse section isolation, with spatial and temporal resolution of $\Delta x = \Delta y = \Delta z \approx 5$ mm and $\Delta t \approx 10$ sec. On the other hand, single photon imaging is primarily based on radiopharmaceuticals labeled with ^{99m}Tc, balancing the oft-stated advantages (ideal detection energy, low patient dose, versatile chelation chemistry) against the disadvantages (tissue scatter, inability to rigorously correct attenuation, no biochemical authenticity). A new class of lipophilic technetium flow agents (teboroxime, methoxy isobutyl isonitrile,..) has spurred clinical interest in cardiac perfusion imaging, and would benefit considerably from a quantitative study of the pharmacokinetics of these agents in man. Technetium-94m, a 53-minute positron emitter (1) , can provide this service, with PET providing an accurate measurement (2) of the biodistribution and clearance of the test compound in a select group of normals and patients.

Proton irradiation of natural molybdenum at E_p=11 MeV results in thick target yields of the various technetium isotopes collected in Table I, listing the reaction, half-life, Mo natural abundance, Q-value, "signature" gamma energy for unique identification and activity per μA at end of saturated bombardment on natural Mo. In the final column, the activity remaining after a *practical* (11 MeV > E_p > 6 MeV, 40 μA, 53-minute irradiation on natural Mo; 53-minute chemistry) workup shows that a natural molybdenum metallic target is suitable for the synthesis of Tc agents for human use. Nonetheless, irradiation of an isotopically enriched Mo-94 target would considerably reduce the absorbed dose to the patient. This is borne out by serial high resolution gamma spectroscopy and measured decay curves of β^+ activity from irradiated 95% ^{94}Mo and ^{nat}Mo, respectively.

Technetium (VII) separation from metallic, natural Mo foils (≈200 mg) presently employs electrodissolution ($NH_4Cl + H_2O_2$; glassy carbon cathodic beaker, ≈3A/cm^2, 3 min), followed by MEK / 5N NaOH extraction of the TcO_4^-. This wet chemistry is not well suited for enriched ^{94}Mo, priced at ≈ \$5/mg. Early work (3,4,5) demonstrated the extraction of volatile ^{99m}Tc (VII) from low specific activity ^{99}Mo by sublimation from MoO_3 at 800°C. Repeating this separation of ^{94}Tc from proton irradiated MoO_3, we find that 95% of the Tc-activity rapidly migrates away from the melt, depositing ≈ 10 cm down the open quartz tube projecting from a 900°C muffle furnace. This activity, accompanied by ≈1 mg trace of MoO_3, was easily removed from the quartz surface in a 5N NaOH wash, followed by extraction into MEK as TcO_4^-.

Having this sublimation process take place **during** the cyclotron irradiation offers several distinct advantages:

- the ^{94m}Tc losses from handling and decay are minimized
- the $^{94}MoO_3$ target material losses are minimized, since the inventory can be permanently secured in the target chamber and
- radiation exposure to personnel is minimized as the cyclotron and sublimation target figuratively approach the ideal ^{94m}Tc generator system.

Toward this end, Fig 1 schematically shows our first developmental target, attempting to control ^{94m}Tc sublimation out of $^{94}MoO_3$ held in the molten state by the vertical beam (6) of the UW CTI 11 MeV proton cyclotron. Double focussing optics were incorporated into the design, resulting in a beam strike of 1 mm x 2 mm in the focal plane 30 cm beneath the magnet. Initial beam tuneup work, sweeping $^{10}CO_2$ out of enriched $^{10}B_2O_3$ melts have shown the need for careful target telemetry, with local temperatures on the target surface observed by both IR and visible (color) TV cameras. While the support of molten target materials is far easier in the vertical configuration, future work will be directed to develop simple horizontal targets better suited for ^{94m}Tc production at high beam currents.

Technetium-94m, coupled with the quantitative nature of PET, can offer the Tc- chemist the ability to truly follow the pharmacokinetics of new imaging agents, non-invasively, in man. This advantage, previously the bastion of orthodox PET compounds labeled with ^{11}C, ^{13}N, ^{15}O and ^{18}F, can serve a vital role in bringing new ^{99m}Tc radiopharmaceuticals into clinical use.

References

1. CM Lederer, VS Shirley, Table of Isotopes, 7th edition, John Wiley and Sons, New York, 1978.
2. RJ Nickles, AD Nunn, CK Stone, SB Perlman, RL Levine. J Nucl Med **32**,925 (1991).
3. C Perrier, E Segre. J Chem Phys **5**, 712 (1937).
4. J Vleck, V Machan, V Rusek, L Kokta, J Rohacek, Z Smejkal, J Vitkova. J Radiochem Radioanal Lett **20**, 15 (1974).
5. J Robson. US Patent 3,833,469 (Sept 1974).
6. RJ Nickles, J Nucl Med **32**, 1091 (1991).

Table I.
Technetium yields from the irradiation of ^{nat}Mo with 11 MeV protons

Reaction	$t_{1/2}$	Abund	Q	E_γ(keV)	A_{EOSB}	$A_{practical}$
$^{92}Mo(p,n)^{92}Tc$	4 m	14.8%	-8.8	1510	2 mCi/μA	20 μCi
$^{94}Mo(p,n)^{94m}Tc$	53 m	9.2%	-5.1	1522	7.2	72 mCi
$^{94}Mo(p,n)^{94}Tc$	4 hr	9.2%	-5.0	702	2.5	10 mCi
$^{95}Mo(p,n)^{95}Tc$	20 hr	15.9%	-2.4	766	10	11 mCi
$^{95}Mo(p,n)^{95m}Tc$	61 d	15.6%	-2.5	204	1 μCi	50 μCi
$^{96}Mo(p,n)^{96}Tc$	4 d	16.7%	-3.7	778	78 μCi	3 mCi

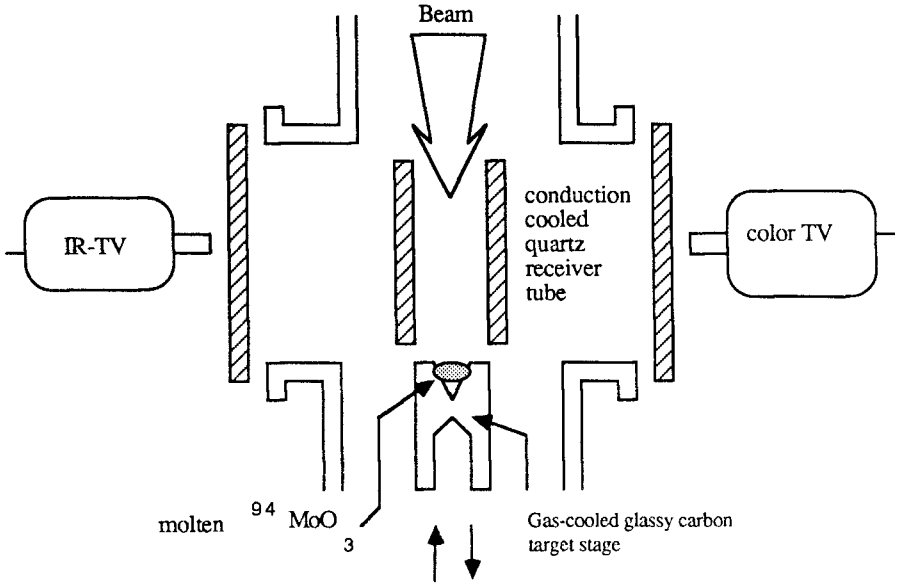


Fig. 1. Vertical beam target for the simultaneous irradiation of molten enriched $^{94}MoO_3$ (mp = 795°C) and sublimation of ^{94m}Tc. The focussed beam strike area (1 mm x 2 mm) and 11 > E > 6 MeV target thickness (200 mg/cm^2) minimize the target inventory needed.

Paper J4

NEW NUCLEAR DATA RELEVANT TO THE PRODUCTION OF THE POSITRON EMITTING RADIOISOTOPE ^{86}Y

F. Rösch*, S.M. Qaim+, G. Stöcklin+

*Zentralinstitut für Kernforschung Rossendorf, Bereich Radioaktive Isotope, Postfach 19, O-8051 Dresden, Germany

+Institut für Nuklearchemie, Forschungszentrum Jülich , 5170 Jülich, Germany

The relatively long-lived positron emitter ^{86}Y ($T_{1/2}$ = 14.7 h; β^+ = 34%; $E_{\beta}{}^+$ = 1.2 MeV) seems to be a promising radionuclide for special PET-studies since it allows to measure the uptake kinetics and individual dosimetry, preparatory to therapeutic investigations with the long-lived β^- emitter ^{90}Y. The radioisotope ^{86}Y can be produced via the $^{86}Sr(p,n)$- and $^{86}Sr(d,2n)$-processes at small cyclotrons. However, no detailed experimental data on these nuclear reactions are available in the literature. The aims of this study were (i) to prepare enriched ^{86}Sr-samples suitable for irradiations, (ii) to measure cross section data by means of the stacked-foil technique and finally (iii) to deduce optimum conditions for the production of ^{86}Y.

The most suitable chemical form for irradiation of Sr was found to be $SrCO_3$. Using a suspension of $^{86}SrCO_3$ (isotopic enrichment of ^{86}Sr = 96.3%) in acetone and collodium, samples of 10 mm diameter and of thicknesses between 12.40 and 5.93 mg/cm^2 on 25 μm thick Cu backing foils were prepared via a special sedimentation method. The samples were homogeneous, chemically stable and could be handled well. An optical emission spectroscopic analysis gave the sum of the impurities like Na, Mg, Si, Ca and Ba as <400 ppm. A diffractometric phase analysis showed that the target material was >99% $^{86}SrCO_3$.

The $^{86}SrCO_3$ thin samples and Cu monitor foils were stacked together and irradiated with 19 MeV protons at beam currents of about 50 nA at the Jülich Compact Cyclotron CV 28. The activities of the radioactive products were determined via Ge detector γ-ray spectroscopy. Excitation functions were obtained for the reactions $^{86}Sr(p,n)^{86m,g}Y$ and $^{86}Sr(p,2n)^{85m,g}Y$ from thresholds to 18.6 MeV. The excitation function of the reaction of interest, i.e. $^{86}Sr(p,n)^{86}Y$, shows a maximum at about 14 MeV.

The thick target yields of ^{86}Y calculated from the preliminary cross section data are given in Table 1.

TABLE 1 **Theoretical thick target yields of ^{86}Y calculated from the excitation function of the $^{86}Sr(p,n)^{86}Y$ reaction***

Energy range (MeV)	Thick target yield of ^{86}Y MBq (mCi)/μAh
15 → 11	426 (11.5)
11 → 6	196 (5.3)

*Assuming 100% enrichment of ^{86}Sr.

Considering the yields and impurities the optimum energy range for the production of ^{86}Y appears to be E_p = 15 → 11 MeV. However, even in the lower energy range of E_p = 11 → 6 MeV some ^{86}Y can be produced. Experimental ^{86}Y yields and contamination levels from $^{85,85m}Y$, ^{87}Y, ^{88}Y, ^{85}Sr and ^{83}Rb have been measured.

A COMPUTER-CONTROLLED AUTOMATED SYSTEM FOR THE PRODUCTION OF HIGH-PURITY ^{67}GA AND ITS CHEMICAL QUALITY CONTROL BY ANODIC STRIPPING VOLTAMMETRY.

P. Van den Winkel, L. De Vis, H. Coolsaet, C. Van den Bossche, G. Brits and F. Van Daele.

VUB-Cyclotron, Vrije Universiteit Brussel, Laarbeeklaan 103, 1090 Brussels, Belgium.

The legislation requires for the industrial production of ^{67}Ga containing diagnostics requires standardized physico-chemical procedures resulting in a bulk solution of high specific volume activity showing a well-defined radionuclidic, radiochemical and chemical quality. To enhance the reproducibility of the production process and in order to avoid any operator error, the automation of all procedures involved is therefore a must. The present paper describes a low-cost, custom-made and computer-controlled automated system used at the VUB-Cyclotron department with all radiation-sensitive devices mounted outside the hotcell for the radiochemical recovery of carrier-free ^{67}Ga from irradiated ^{68}Zn targets and its chemical quality control. The flow diagram of the radiochemistry involved is shown in Figure 1. A picture of the automated radiochemistry system is represented in Figure 2. It includes :

1. a custom-made manifold i.e. an electromechanical set-up mounted inside the hotcell, that makes possible to carry out the ^{67}Ga separation chemistry which means that physico-chemical and chemical processes such as dissolution, oxidation by gaseous reagents, ion exchange chromatography and solvent-solvent extraction can be performed automatically with an overall yield better than 95 % for ^{67}Ga;
2. custom-made interface electronics - DAC, conductivity and current threshold cards - allowing connection between the manifold and the PC that is fitted with digital I/O and optical isolated I/O subsystem cards;
3. a PC with two monitors permitting remote control of the whole separation chemistry : a VGA monitor, used in graphic mode, shows the complete flow-chart of the chemistry including the state of valves, motors and sensors of the manifold - state that can be altered by the operator through mouse activation - while the operator interacts by menu driven commands on a monochrome monitor used in text mode. Thereby the software, written in Turbo-Pascal, decides on the sequence of the production steps, registrates the state of objects (valves, motors, sensors) and keeps a logfile;
4. a custom-made sampling device making remote-controlled sampling and

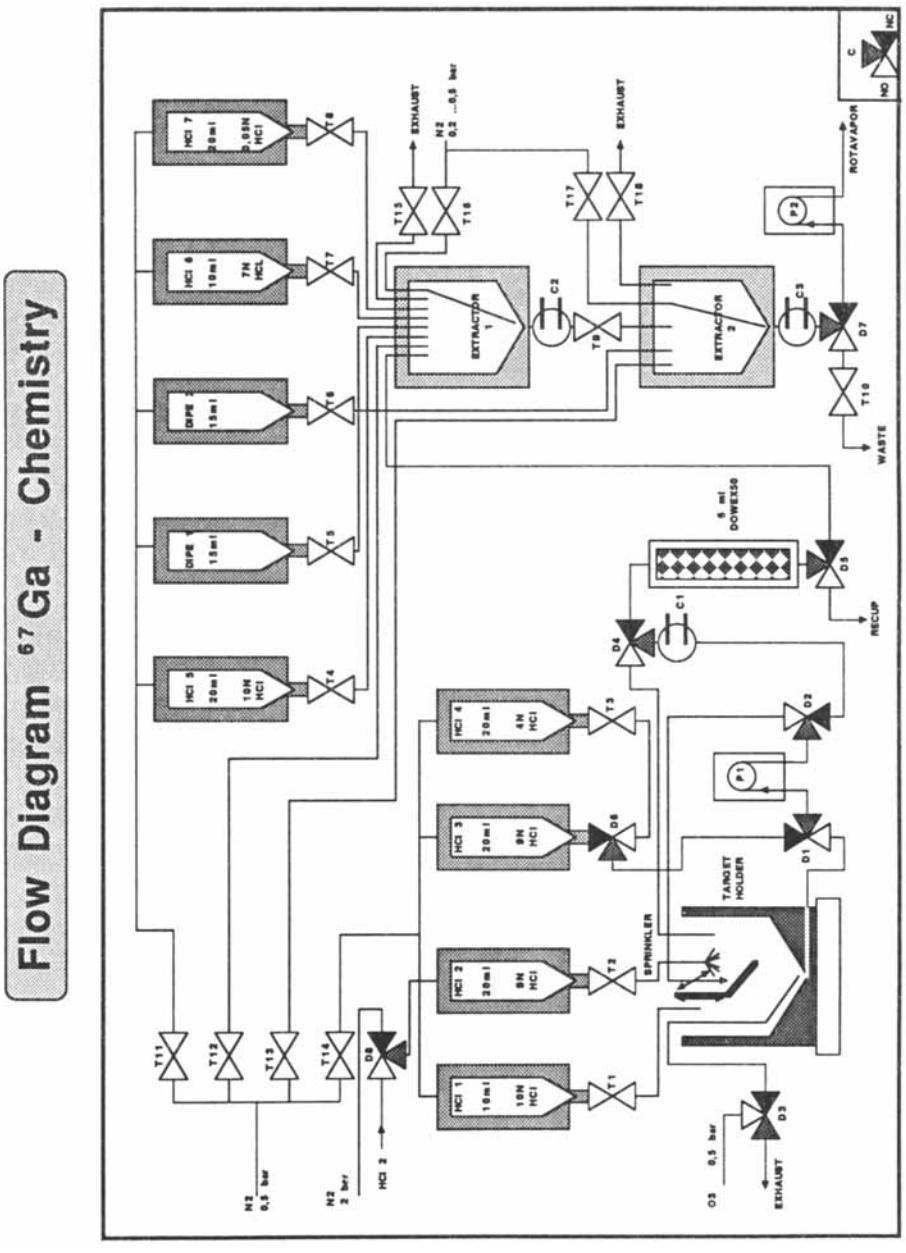

Flow Diagram 67Ga - Chemistry
HCl 7 20ml 2.05N HCl
HCl 6 10ml 7N HCl
DPE 15ml
DPE 15ml
HCl 5 20ml 10N HCl
HCl 4 20ml 4N HCl
HCl 3 20ml 8N HCl
HCl 2 20ml 8N HCl
HCl 1 10ml 10N HCl
T1
T2
T3
T4
T5
T6
T7
T8
T9
T10
T11
T12
T13
T14
T15
T16
T17
T18
EXTRACTOR 1
EXTRACTOR 2
C1
C2
C3
C
NC
NO
D1
D2
D3
D4
D5
D6
D7
D8
P1
P2
5 ml DOWEX50
TARGET HOLDER
SPRINKLER
EXHAUST
EXHAUST
N2 0,2 ...0,5 bar
ROTAVAPOR
WASTE
RECUP
N2 0,5 bar
N2 2 bar
HCl 2
O2 0,5 bar
EXHAUST

Figure 1

dilution possible prior to chemical quality control by anodic stripping voltammetry.

Details concerning the radiochemical procedure, the construction of the manifold, the electronics and the associated software and the accuracy and reproducibility of the polarographic analysis are reported.

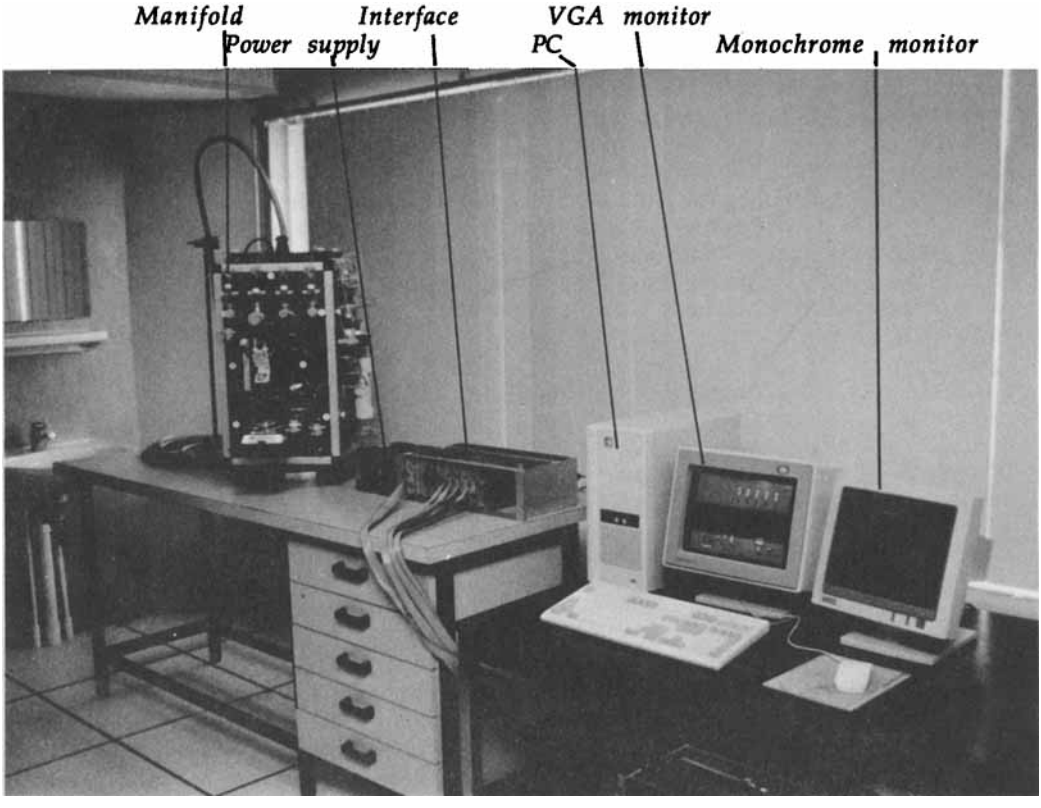


Figure 2

PRODUCTION OF NO-CARRIER-ADDED [^{15}O]CO and [^{15}O]CO_2.

R.Iwata and T.Ido.
CYRIC Tohoku University, Aramaki, Aoba-ku, Sendai 980 Japan.

Oxygen-15 labeled gases are currently the most frequently used radiopharmaceuticals for PET diagnosis. The established method for [^{15}O]O_2 and [^{15}O]CO_2 production requires the addition of O_2 or CO_2 to the nitrogen target up to 2 vol% (1) although some attempts have been made to reduce the amount of carrier involved (2, 3). The ^{15}O produced in the pure N_2 target is moderated by N_2 and eventually lost on the metallic walls of the irradiation chamber. Therefore, we expected that graphite walls inserted into the chamber would efficiently scavenge the ^{15}O to produce C^{15}O.

The pure N_2 (99.999 %) was irradiated by an 11 MeV deuteron beam (an incident energy: 6.7 MeV). It continuously flowed through the irradiation chamber which had a graphite tube inserted and was heated with a furnace as illustrated in Fig.1. The ^{15}O recovered from the target was monitored with a small radiation detector and analyzed by radio-gaschromatography. The effects of production parameters such as beam current, target gas flow rate, pressure and temperature on the production yield and radiochemical form of ^{15}O were systematically investigated.

The reaction of atomic oxygen with carbon requires a small activation energy (4-6). This explains well the correlation curve shown in Fig.3, where it can be seen that the ^{15}O production yield increases with the target temperature and then reaches a plateau over 200°C. Fig.2 demonstrates that the ^{15}O produced in the target is linearly recovered with the deuteron beam current in the temperature range above 200°C. The recovery yield of 17 mCi/min/μA with a flow rate of 300 mL/min at the target output is nearly comparable to the estimated value from the literature (7).

Although the content of O_2 present in the target is not enough to scavenge ^{15}O, it is sufficient for the radiolytic oxidation of [^{15}O]CO into [^{15}O]CO_2 (8). Thus, [^{15}O]CO_2 was found as a main product under the present production conditions. The remarkable increase in the [^{15}O]CO yield was, however, observed for the production with a higher beam current and lower target gas flow rate as seen in Fig.4. It seems likely that a radiation-induced reaction of [^{15}O]CO_2 with the graphite walls to reproduce [^{15}O]CO occurs under intense irradiation.

The content of the carrier was determined by GC with a flame ionization detector. It was usually less than 20 ppm. As [^{15}O]CO_2 can be converted into [^{15}O]CO with heated charcoal or [^{15}O]CO into [^{15}O]CO_2 with CuO without addition of the carrier, the present new production method can be adopted for routine production of no-carrier-added [^{15}O]CO and [^{15}O]CO_2.

(1) Clark J.C. and Buckingham P.D., "Short-lived radioactive gases for clinical use" Butterworths, London (1975).
(2) Votaw J.R., Satter M.R., Sunderland J.J., Martin C.C. and Nickles R.J., *J. Label. Compd. Radiopharm.* **XXIII**, 1211 (1986).
(3) Berridge M.S, Terris A.H. and Cassidy E.H., *Appl. Radiat. Isot.* **41**, 1173 (1990).
(4) Marsh H., O'Hair E., Reed R. and Wynne-Jones W.F.K., *Nature* **198**, 1195 (1963).
(5) Gleit C.E., Holland W.D. and Wrigley R.C., *Nature* **200**, 69 (1963).
(6) Otterbein M. and Bonnetain L., *C. R. Acad. Sci. Paris* **259**, 791 (1964).
(7) Sajjad M., Lambrecht R.M. and Wolf A.P., *Radiochim. Acta* **38**, 57 (1985).
(8) Christman D.R., Finn R.D., Karlstrom K.I. and Wolf A.P., *Int. J. Appl. Radiat. Isot.* **26**, 435 (1975).

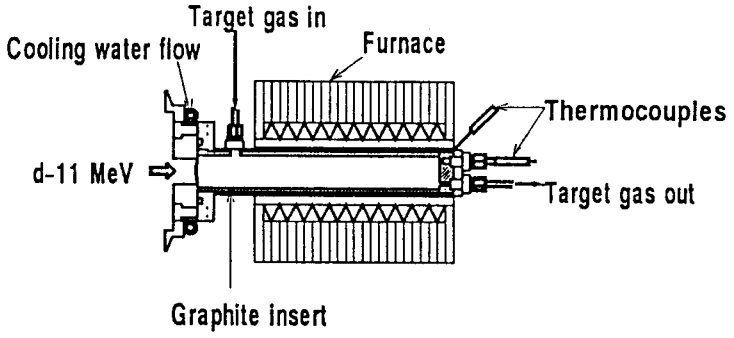


Fig. 1 A sectional side view of the irradiation chamber

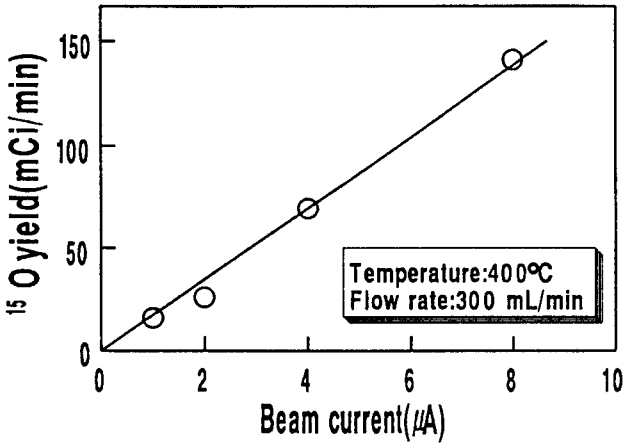


Fig. 2 A linear correlation of the ^{15}O recovery yield with the beam current

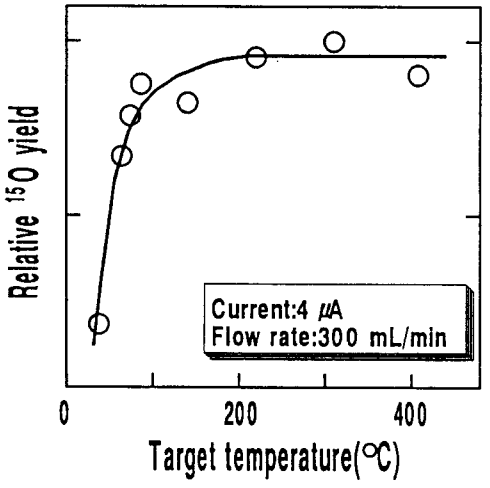


Fig. 3 Temperature dependence of the ^{15}O yield

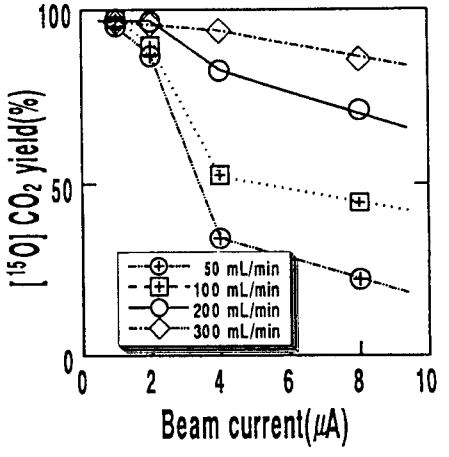


Fig.4 Radiation effects on the [^{15}O]CO_2 yield

A VERSATILE SYSTEM FOR SINGLE-BREATH INHALATION OR REBREATHING OF GASES LABELLED WITH O-15, F-18 OR C-11

R. Wagner, W. Arenz, N.Richerzhagen,K.Wienhard

Max Planck Institut für Neurologische Forschung Köln
Gleueler Str. 50, D-5000 Köln 41

Gases labelled with short lived positron emitters are widely used to measure cerebral blood flow, oxygen consumption or blood volume (for a recent review see 1) ,following two different protocols: The steady -state method with O-15 labelled gases employs a continuous inhalation of a radioactive gas stream with a fixed activity concentration .

Single-breath or short-term rebreathing is the method of choice with tracers such as methyl fluoride labelled with C-11 or F-18. In the case of O-15 carbon monoxide , the application of a small volume in the rebreathing mode can reduce toxicity problems caused by the inevitable carrier content. The method was validated by Martin et al(2), recently also the usefulness of a brief inhalation method for measurement of oxygen extraction fraction has been shown (3).

Collection and storage of the patient exhaled air is important especially in the case of O-15- O_2,because about 80 % of the delivered activity are exhaled again directly.Since the still topical reference by Clark and Buckingham (4) only marginal information about technical details of inhalation systems was available.

The system described here has been in use in our PET facility for the last five years and fulfils the following requirements:
The radioactive gas volume given to the patient is small and fixed to get reproducible specific activities.
O-15-O_2 and O-15-CO are collected and measured in the same way and remotely injected either into a single breath breathing bag (for O_2) or into a rebreathing system (for CO).
Patient exhaled air is collected directly from the face mask and stored until total decay.
The total amount of inactive carbon monoxide applied to the patient is less than 40 µmol and thus far below any toxic level.

References

1. Baron J.C.,Frackowiak R.S.J.,Herholz K., Jones T., Lammertsma A.A., Mazoyer B., Wienhard K. - J.Cereb. Bl.Flow Metab. 9: 723-742 (1989)
2. Martin W.R.W., Powers W.J., Raichle M.E., - J. Cereb. Bl. Flow Metab. 7: 421-426 (1987)
3. Altman D.I., Lich L.I., Powers W.J. - J.Nucl.Med 32: 1738 - 1741 (1991)
4. Clark J.C., Buckingham P.D., Short-Lived Radioactive Gases for Clinical Use London: Butterworths, 1975

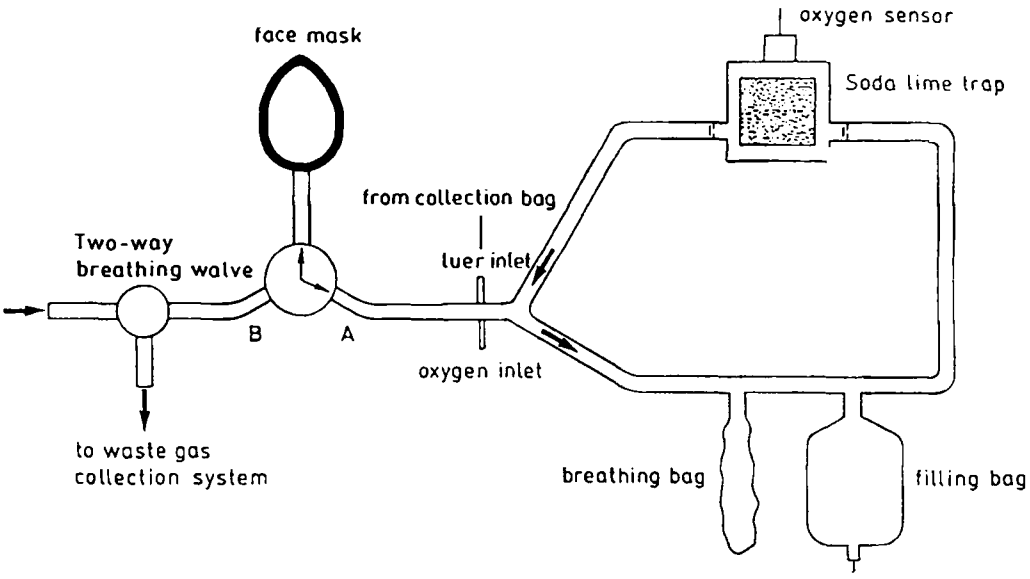


Fig. 3 Breathing system 2 for short rebreathing of carbon monoxide (or methyl fluoride).

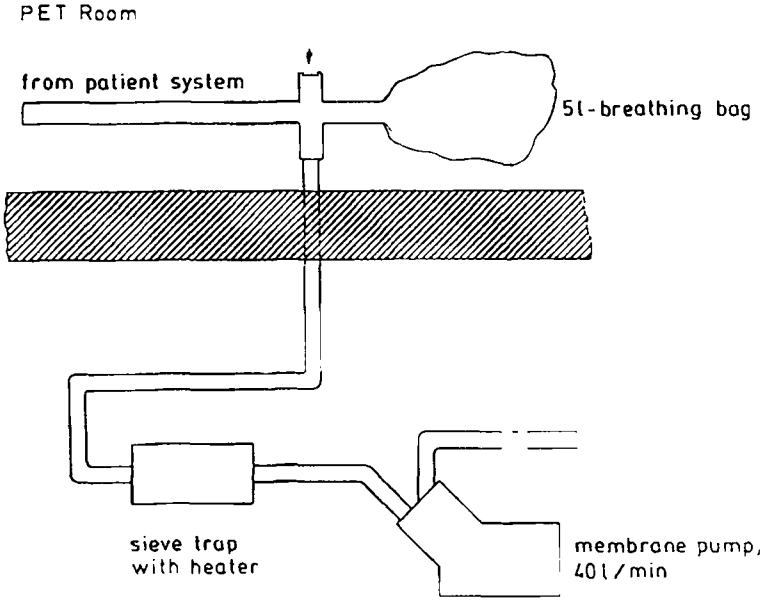


Fig. 4 Waste gas collection system
The 5 l breathing bag acts as a buffer volume, the adjacent check valve prevents excessive underpressure in the system.

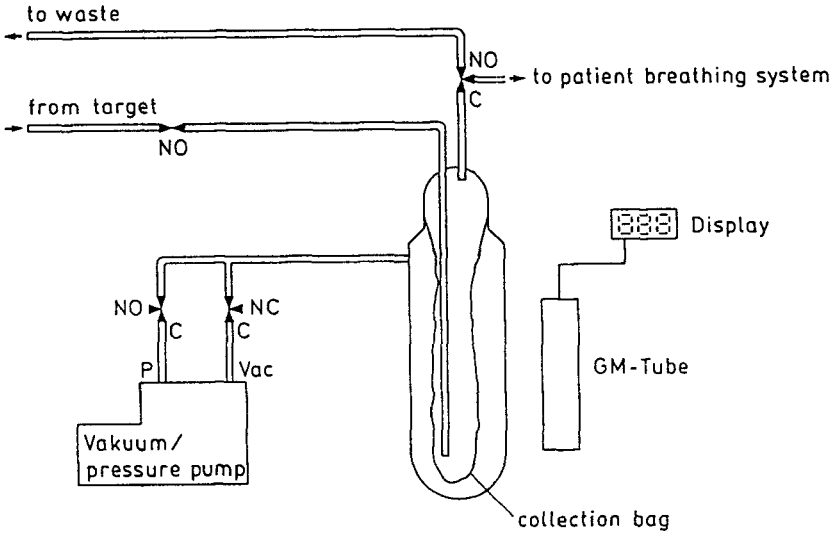


Fig. 1 Collection/Dispensing System is placed in a shielded box next to the PET.The collection bag can be either inflated(collection from target) or compressed (dispensing to breathing system 1 or 2).

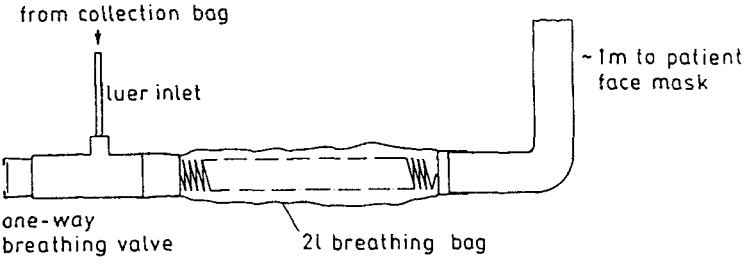


Fig. 2 Breathing system 1 for bolus inhalation of oxygen. Inhalation tube is connected to point A (see Fig.3) at the face mask. After a deep single breath, the three-way stopcock is switched to room air breathing and exhalation to the waste gas system.

AN ADVANCED SYSTEM FOR THE ADMINISTRATION OF ^{15}O-WATER

K.-H. Matzke, G.-J. Meyer, H. Hundeshagen
Abteilung Nuklearmedizin und spezielle Biophysik
Medizinische Hochschule Hannover, D-3000 Hannover 61, Germany

^{15}O labelled water is used for the investigation of blood flow in various organs, mainly the brain and the heart. It is also been applied to measure the perfusion status of tumors and the amount of extravascular water in the lungs. Depending upon the clinical question, it may be administered by different protocols, such as a high activity bolus injection (max. 4 GBq), or an infusion with constant, increasing or decreasing specific activity in the range of 0.1 to 1 GBq for up to 30 min.
Advanced PET- study protocols for quantitative analysis of flow, require repeatable and striktly defined input functions. These can be achieved best by a widely automated production and infusion system. For this reason we improved our previous constant infusion system (Meyer et al (1)).

^{15}O is produced by the ^{14}N(d,n)^{15}O reaction in a nitrogen gas target containing 0.5 % oxygen as carrier. The target is operated at a pressure between 7 and 14 bar and a beam current of 10 to 30 µA under constant flow conditions.
The ^{15}O-water system (see fig 1) is in a distance of about 50 m from the cyclotron, right beside the PET camera. It is shielded by a lead box (20x30x20 cm, wall thickness 5 cm) mounted on a laboratory trolley. Inside the lead box the target gas is controlled by a flowmeter, than it is mixed with hydrogen,and passed through a tube oven, filled with appr. 0,2g Pd (1%) catalyst on a charcoal support (4 - 8 mesh). Reaction temperature is 150^{o}C.
The ^{15}O-water vapour is trapped in 4 necked flask, filled with isotonic saline solution. The level of the saline in the flask is monitored continuously by a set of 10 light barriers. This level control unit commands a gear pump outside the lead box, which withdraws physiol. NaCl solution from an infusion bottle. With a second small gear pump, the ^{15}O-water is pumped out, via a calibration detector and a sterilization filter. In case of constant or ramp infusion, this follows the instructions of the level control unit. In case of a bolus injection, the labelled water is pumped as fast as possible into a pneumatic driven syringe, placed in a dose calibrator, from where it is injected by remote control when the desired activity level is reached.

1) G.-J. Meyer et al. - J. labelled Comp. Radiopharm.23: 1209-1210 (1986)

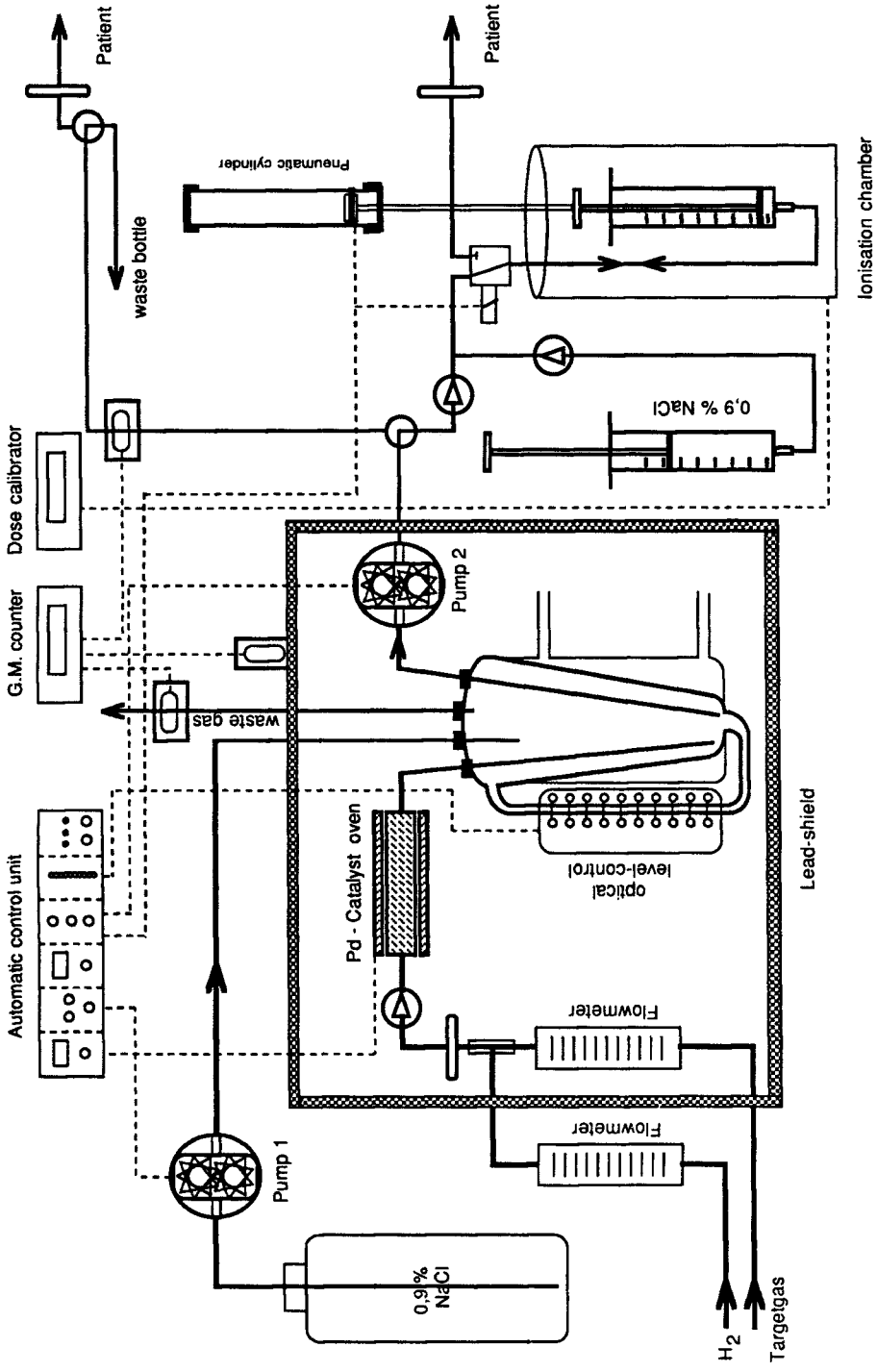


Fig. 1: ^{15}O-Water production and infusion system

Paper J9

Proton Irradiation of Dilute Aqueous Ethanol for In-Target Production of [^{13}N]Ammonia: Studies on the Fate of Ethanol.

Richard A. Ferrieri*, Keith MacDonald, David J. Schlyer and Alfred P. Wolf

Brookhaven National Laboratory, Department of Chemistry, Upton, New York, USA 11973.

[^{13}N]Ammonia is used in PET as a radiotracer of myocardial blood flow (Schelbert, *et al.*, 1986), and as a precursor for synthesis of various L-[^{13}N]amino acids (Barrio, *et al.*, 1983). Most facilities have relied on the chemical reduction of [^{13}N]nitrate and [^{13}N]nitrite formed as secondary products during the proton irradiation of pure water (Parks and Krohn, 1978; Tilbury and Dahl, 1979) as a result of radiolytic oxidation of [^{13}N]ammonia. Even so, increased interest is seen in a method where the ammonia state can be maintained during irradiation using dilute aqueous ethanol (Wieland, *et al.*, 1991). Ethanol is presumed to scavenge hydroxyl radicals formed during bombardment that would otherwise cause this oxidation. At 5 mmolar concentration, which seems to be the minimum level needed to suppress this undesirable process, all of the ethanol is consumed. This is disconcerting in view of the fact that ethanol may undergo oxidative cleavage to a known carcinogen such as formaldehyde. We carried out a study to determine what ethanol decomposes to under production conditions. [^{11}C]Ethanol, labelled in the C_1 position, was prepared by conventional methods through [^{11}C]CO_2 reaction with methyl magnesium bromide (Langstrom, *et al.*, 1986). Purified [^{11}C]ethanol was mixed with authentic sample to yield concentrations of 5 and 50 mmolar. Target studies were carried out with these solutions on the BNL 60-inch cyclotron using a 0.6 mL unpressurized Cu target with a 1 miL thick Ti window. Irradiations were of 2 minute duration using a 20 μA beam of 18 MeV protons. After bombardment, target waters were subjected to radio-GC and radio-HPLC to analyze for [^{11}C]organic compounds. Target volatiles were also processed through a series of traps suitable for collecting carbon oxides. Results showed that 90% of the [^{11}C]ethanol decomposed under the above irradiation conditions, regardless of the concentration. It is suspected that the remaining alcohol may be a feature of the target dead volume. Of that which did decompose, 97% of the activity was recovered as [^{11}C]CO_2, and the remainder as [^{11}C]CO. No labelled organic products were detected. Additional studies are underway to determine what happens to ethanol when the label is on the C_2 position. These results will also be discussed. This research was carried out under contract DE-ACO2-76CH00016 with USDOE and supported by its OHER.

References

Barrio, J.R., Braumgartner, F.J., Henze, E., Stauber, M.S., Egbert, J.E., MacDonald, N.S., Schelbert, H.R., Phelps, M.E. and Liu, F.-T. (1983) *J. Nucl. Med.* **24**, 937.

Langstrom, B. Antoni, G., Gullberg, P., Halldin, C., Nagren, K., Rimland, A. and Svard, H. (1986) *Appl. Radiat. Isot.* **37**, 1141.

Parks, N.J. and Krohn, K.A. (1978) *J. Appl. Radiat. Isot.* **29**, 754.

Schelbert, H.R. and Schwaiger, M., in Phelps, M.E., Mazziotta, J.C. and Schelbert, H.R. (eds.), (1986) *Positron Emission Tomography and Autoradiography*, Raven Press, NY, Chapter 12.

Tilbury, R.S. and Dahl, J.R. (1979) *Radiat. Res.* **79**, 22.

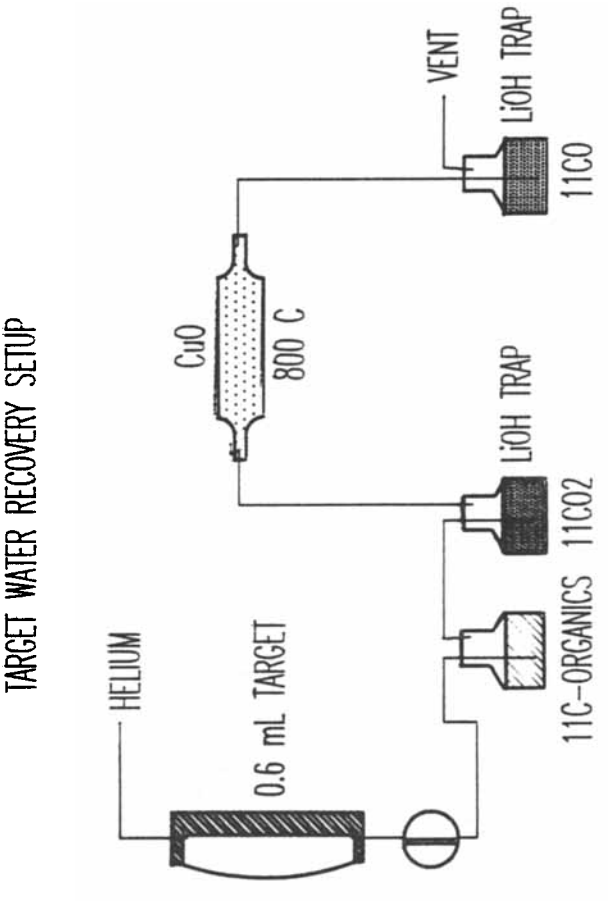
Wieland, B.W., Bida, W., Padgett, H., Hendry, G., Zippi, E. Kalbalka, G., Morelle, J.-L., Verbruggen, R. and Ghyoot, M. (1991) *Appl. Radiat. Isot.*, in press.

RESULTS FROM BNL ^{11}C-EtOH EXPERIMENTS

Pre-irradiation Conditions[a]		Post-irradiation Act. Distribution (mCi)[b]		
EtOH Molarity (mMol)	^{11}C-EtOH Act. (mCi)	^{11}C-EtOH	$^{11}CO_2$	^{11}CO
5	3.34	0.36 (10)	2.95 (89)	0.03 (1)
50	4.58	0.61 (12)	4.06 (84)	0.18 (4)

a. All experiments carried out on the BNL 60-inch cyclotron using an 0.6 mL volume Cu target with a 1 miL thick Ti window and 10 mm collimator. Irradiations were carried out using a 20 μA, 18 MeV H^+ beam for 2 minutes.

b. Numbers in parentheses represent percent distribution.


TARGET WATER RECOVERY SETUP
HELIUM
0.6 mL TARGET
CuO
800 C
LiOH TRAP
11C-ORGANICS 11CO2
VENT
LiOH TRAP
11CO

Paper J10

PRODUCTION OF ^{73}Se BY PROTON IRRADIATION OF Cu_3As TARGET ALLOY

Deborah de F. Santos and Regin Weinreich
Institute of Medical Radiobiology, University of Zürich
c/o Paul Scherrer Institute, CH-5232 Villigen, Switzerland

^{73}Se ($T_{1/2}$=7.1hr, β^+=65%, EC=35%) is a positron emitter and has similar chemical properties as sulphur. It is assumed that it can be used as substitute for sulphur in tumor-seeking radiopharmaceuticals, the distribution of such compounds in-vivo can be studied by PET.

During the last years, some papers have been published about ^{73}Se excitation functions using different charged particles and targets. Looking for the experimental data, $^{75}As(p,xn)^{73}Se$ proved to be the best reaction, as far as a cyclotron with medium proton energy is avaiable [1,2]. The choice of target material is dependent on the beam current of the the cyclotron and the efficiency of the cooling system.

In our laboratory, Cu_3As alloy was chosen as a target instead of elemental As or As_2O_3, that have been used as target material for ^{73}Se production in earlier work [2].

The yield of reaction was measured for ^{73}Se 1.22GBq/uAh (33±7mCi/uAh), ^{72}Se(0.34%), ^{75}Se(0.007%) at EOB.

The chemical separation consisted of two parts:

-Separation of copper (and radionuclides produced from $^{nat}Cu(p,x)$) from As target material (and radionuclides produced from $^{75}As(p,x)$) using cation exchange chromatography. The targed is dissolved in HNO_3 conc.+ H_2O_2, this solution is percolated on a DOWEX 50WX8, H^+ form, 100-200 mesh preconditioned with HCl 0.1N/acetone (4:6 v/v). As(and accompanying radionuclides) eluted first with HCl 0.1N/acetone, Cu(and accompanying radionuclides) is eluted with HCl 8M.The separation yield for this step is 98%.

-As and Se are separated using anion exchange chromatography [2].

1.Nozaki T. et al.,-Appl. Radiat. Isot. 30:595(1979)
2.Mustaq A. et al.,-Appl. Radiat. Isot. 39:1085(1988)

Paper J11

PREPARATION OF ^{90}Sr-^{90}Y GENERATOR

Jin Xiaohai, Yu Haibin, Zhang Jingming, Zhang Peixin, Lin Qiongfang
Isotope Department of China Institute of Atomic Energy,
P.O.Box 275(58),Beijing 102413, P.R. China

In recent years, ^{90}Y has been considered as one of the best radionuclides for tumor radioimmunotherapy when chelated to tumor-associated antibodies.This evaluation is based on the superior properties of this radionuclide (suitable half-life,pure β-ray emitter of intermediate energy, stable daughter, and suitabe chemical properties) and because it is available as a radionuclide generator product by decay of its 28 years parent Sr.In this paper experimental conditions of the ^{90}Sr-^{90}Y generator are described. The influence factors of ^{90}Y seperation from ^{90}Sr parent were investigated. The effect of EDTA concentration on ^{90}Sr and ^{90}Y absorption is shown in Fig.1. The absorption of ^{90}Sr does not depend on the concentration of EDTA. The effect of pH values on ^{90}Sr and ^{90}Y absorption is shown in Fig.2. The pH values of ^{90}Sr absorption was selected between 3-5.5. The effect of the various complex agents on ^{90}Y decontamination factors are given in Table 1. The results indicated that the EDTA complex agent seem to be better than others. The elution efficieny of $^{90}Sr-^{90}Y$ generattor reaches 98%. One of the most important problems is the ^{90}Sr contamination breakthrough from the generator.

The level of ^{90}Sr contamination must be controlled to reach the clinical standard . The cation exchange resin 732 (100-150 mesh) was successfully used for the separation of ^{90}Y from ^{90}Sr. Our method provides a ^{90}Y-HAc solution which is very simple and safe for administration to the patients. ^{90}Y was separated from ^{90}Sr almost completely, the level of the ^{90}Sr contamination per 740MBq ^{90}Y product was only 0.74kBq. However the toxicity of ^{90}Sr is extremely high, the human life-time permissible dose is 74kBq, then 740MBq of ^{90}Y is allowed to be administrated to a patient for 50-100 times.

Table 1. Effect of the various comples agents on ^{90}Y decontamination factors

No.	717 resin(made in China) 100-150 mesh			
	complex agents	mol/L	pH	decontamination factors $\times 10^6$
1	^{90}Y-EDTA	0.003	5.5	2.3
2	^{90}Y-HAc	0.003	5.0	0.3
3	^{90}Y-Citrate	0.5	5.0	0.2

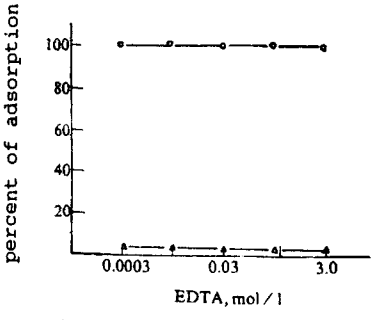


Fig.1 Effect of EDTA concentration on ^{90}Sr and ^{90}Y adsorption

○—^{90}Sr; ▲—^{90}Y.

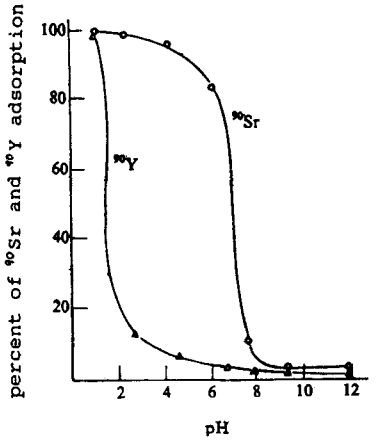


Fig.2 Effect of pH solution on ^{90}Sr and ^{90}Y adsorption

Paper J12

$^{113}Sn/^{113m}In$ GENERATORS - A STUDY OF THEIR PERFORMANCE AND QUALITY

F RAKIÁS, M. BODOR, J. GERSE
National Institute of Pharmacy, Hungary
H-1372 Budapest, P.O.Box 450

Three commercial (A,B,C) $^{113}Sn/^{113m}In$ generators obtained from different manufacturers were studied in our Institute to test the quality of their eluates. The following parameters have been investigated:
Elution efficiency, radionuclidic purity, radiochemical purity Zr, Zn and Ni contents, labelling characteristics, practical aspects.
Dose calibrator was used in order to determine the elution efficiency (CAPINTEC CRC-120). We have measured the radionuclidic purity by high purity Ge semiconductor detector (CANBERA and ICA-80 analyzer). The radiochemical purity was checked by paper-chromatography (USP XX and paperchromatography by Merlin) and thin-layer chromatography (ITLC-SG,Gelman). The Zr, Zn, and Ni levels were determined by colorimetric test strips. The labeling characteristic was investigated by DTPA kits (produced by A and B manufacturers) The measured values were compared with the declared data of manufacturers and the USP XX.
Conclusions:
- Although generator A is bilt difficult to handle (3 vials are needed for one elution) and generator B also has some handelig problems too (ex.: its vials are overloded in their elution volumes), the elution efficiency of all generators proved to be good as declared.
- Generators B and C showed Sn-113 breakthrough higher than allowed.
- Generator B gave several times not allowed Zr and Zn contents and was overloded with Sn-113.
- As far as radiochemical purity methods were concerned Merlin's methods was the best one.
- Both the eluates, produced by any of the generators, and the the radiopharmaceuticals labelled by them were of good quality.
- As for their practical properties, because of above mentioned problems, only Generator C was suitable.

Paper J13

XENON-127 PRODUCTION IN A REACTOR FOR MEDICAL APPLICATIONS.

Vereshagin Yu.I., Prusakov V.N. and A.F. Yashin

I.V. Kurchatov Institute of Atomic Energy,
Moscow, USSR.

Xenon-127 hase been used as a pulmonic diagnostic radionuclide. This application of Xe-127 is based on the nuclear-physical properties of this isotope which is a pure γ-irradiator with the energy of the principal γ-line of 202.8 keV (a 67.7 % yield).

Another radionuclide being used for these purposes,Xenon-133, irradiates 90 keV γ-quanta (a 36.3 % yield) and β-particles with the maximum energy of 346 keV.

The comparison of the properties of these radionuclides indicates that the Xe-133 detection is considerably more complicated and the radiation load on the internal organs of a patient is three times higher with Xe-133 then with Xe-127.

The report suggests a procedure of the optimal Xe-127 production in a nuclear reactor; physical principles of the $^{126}Xe(n,\gamma)^{127}Xe$ reaction are laid down followed by the analysis of accompaning additions of radionuclides depending on the isotop seperation of Xe-127. Experimental results are presented.

Paper J14

THE Sr-82 CYCLOTRON PRODUCTION FOR MEDICAL PURPOSES. METHOD. EXPERIMENTAL RESULTS.

Vereshagin Yu.I., Zagryadskiy V.A.,Prusakov V.N.
I.V. Kurchatov Institute of Atomic Energy,
Moscow, USSR.

Rubidium - 82 is one of the perspective positron emission radionuclides, which can be used at the positron tomography method. The short half-life of the ^{82}Rb (75 sec) allow significantly to reduce radiation dose to patients and hospital personnel.Rubidium - 82 is obtained from a generator system through its 25-day half-life parent ^{82}Sr.

This report is devoted to problem of the ^{82}Sr cyclotron production for the ^{82}Sr-^{82}Rb generator. The ^{82}Sr was received into RbCl (99%^{85}Rb) target with help of the $^{85}Rb(p,4n)$ reaction, using 70 MeV protons at a current approximately 75 μA. The target was irradiated at the inner cyclotron bunch. Chemical processing included chromotographic isolation of Sr with help of ion exchange column.

The experimental methods and results are presented in this report.

Paper J15

USE OF VERTICAL PROTON BEAM TARGET FOR SHORT-LIVED RADIONUCLIDES PRODUCTION

V.I.Tchuev, E.P.Derevyanko, I.V.Ekaeva, V.G.Isotov, T.A.Katunina, M.N.Klebanov, L.S.Larionov, N.S.Marchenkov

(Kurchatov Atomic Energy Institute, Moscow)

The preparation of ^{11}C, ^{13}N and ^{18}F on electrostatic tandem accelerator and following syntheses of radiopharmaceuticals are investigated. The main feature of target being used is its location on vertical proton beam channel of the accelerator.

The target is designed as a test-tube (150 cm^3; quartz, stainless steel or plastic) closed with 30 μm aluminium foil on its top side faced to the beam. The branch-pipes have been used for filling the target with gases and liquids (the maximum pressure 0.2 MPa) and extraction of irradiated materials.

For [^{11}C]CO_2 and [^{11}C]CO preparation the proton irradiation of boron oxide has been chosen. During irradiation boron oxide melts at the test-tube bottom and liberates just formed molecules of [^{11}C]CO_2, which escape from B_2O_3 matrix to gaseous phase. There is no problem with vertical target to keep melted boron oxide at the beam axis. Triple washing of the target vessel with helium is quite enough for 95% activity extraction.

The same target test-tube has been used for [^{18}F]fluoride preparation by proton irradiation of water with natural content of ^{18}O. Due to low beam parameters (5.0 MeV, 5 μA) there is no need of heat removal and water condensation. Up to 0.5 MBq ^{18}F without ^{13}N have been produced after 2 hours irradiation.

Analogous vertical target test-tube has been applied for gaseous target substance ([^{13}C]CH_4). In this case a mixture of N-13 labeled products was obtained.

Paper J16

The $^{194}Os/^{194}Ir$ Generator--Production and Purification of ^{194}Os

S. Mirzadeh, D. E. Rice, A. P. Callahan and F. F. Knapp, Jr., Nuclear Medicine Group, Health and Safety Research Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6022

Iridium-194 ($t_{1/2}$ = 19.15 h) decays by β-particle emission (E_β^{max} = 2.236 MeV) and is a potential candidate for radioimmunotherapy [1,2]. An important characteristic is availability of carrier-free ^{194}Ir from β-decay of ^{194}Os ($t_{1/2}$ = 6 y). The ^{194}Os parent nuclei is produced in a fission nuclear reactor with double neutron capture on ^{192}Os (41.0% natural abundance).

$$^{192}Os[n,\gamma] \xrightarrow{\sigma=2.0\pm0.1\ b} {}^{193}Os[n,\gamma] \xrightarrow{\sigma=240\pm40\ b} {}^{194}Os$$

$$^{193}Os \xrightarrow{\beta^-,\ 30.0\ h} {}^{193}Ir \qquad {}^{194}Os \xrightarrow{\beta^-,\ 6.0\ y} {}^{194}Ir\ (\beta^-,\ 19.15\ h)$$

We report here the preliminary nuclear data for production of ^{194}Os and discuss the feasibility of producing sufficient quantities of ^{194}Os required for the fabrication of the large-scale generators for further studies. In addition, we describe a novel gas-thermochromatographic method (GTC) for the one step conversion of metallic Os to OsO_4 and subsequent separation and purification of OsO_4.

Production of ^{194}Os

Relative to the 1.28 mb cross-section for $^{58}Fe[n,\gamma]^{59}Fe$ flux monitoring reaction, a cross-section of 240±40 b was calculated for the $^{193}Os[n,\gamma]^{194}Os$ reaction. The irradiation was performed in the HFBR where 27.1 mg of 99.3% enriched ^{192}Os was irradiated for one cycle (24 d) at position "Modified V-16". After about 6 months cooling period, the induced radioactivities in the ampules were measured directly without chemical separation. A summary of the reported cross-section for this reaction is given in Table 1. Based on our data, 12 mCi of ^{194}Os can be produced by irradiating 25 mg of enriched ^{192}Os for 60 d at the HFIR (ϕ_n=2x10^{15} n.s^{-1}.cm^{-2}) and the production of ^{194}Os at this facility is currently being explored.

Table 1. Thermal Neutron Capture Cross-section of ^{193}Os

Reactor/ Irradiation position	ϕ_n(2200 ms^{-1}) n.s^{-1}cm^{-2}	Target mass, g as Os metal (Enrichment, %)	Irradiation Period	Effective Cross-section, b	References
HFBR-BNL	4.5x10^{14} *	27.1 (99.3)	24 d	240±40#	Mirzadeh[2]
ORNL-Graphite	~1x10^{12}	-	-	200	Lindner[3]
MTR	3x10^{14}	50	150 d	8	Williams[4]
-	-	-	-	1540	Mughabghab[5]§
ILL/GAMS1-3	8x10^{14}	84 (99.1)	on-line	38±10	Casten[6]

*Measured §No experimental details were found for this measurement
#Relative to the cross-section of 1.28 mb for $^{58}Fe[n,\gamma]^{59}Fe$ flux monitoring reaction

Gas-Thermochromatographic Separation of Osmium

The irradiated ^{192}Os (50 mg or less) target was transferred to a quartz boat which was then placed in the chromatography tube assembly as shown in Figure 1. While maintaining an air flow rate of 10-20 mL/min the furnace was turned on. As OsO_4 formed, it was carried by the air stream and trapped in 2 mL of 0.1 M KOH placed down stream (Figure 1). The first evidence of conversion of metallic Os to OsO_4 occurs at ~300°C within the first 30 minutes of distillation, as indicated by a slight yellowish color in the first trap. Distillation was continued for the next 2 hours while the furnace temperature was increased to ~500°C. At this point, conversion was considered complete, and the furnace was turned off. The air flow was continued for an additional 3 hours to insure quantitative transport and recovery of the OsO_4. The quantitative conversion of 50 mg of Os to OsO_4 within 2.5 hours under the above conditions was demonstrated by analysis of the mass of the boat before and after distillation in several cold runs employing natural Os metal. Consistently, a light-weight black residue (~1% of the Os mass) remained in the boat after distillation. By spectrographic analysis, the residue was found to be primarily graphite.

Consistent with the previous observation, the reaction between Os and O_2 occurs rapidly at ~300 °C. Below this critical temperature, however, the reaction rate is quite slow with no measurable reaction at room temperature within 24 hours. The Teflon tubing which was used as the delivery tube, connecting the chromatography column to the KOH trap, showed no affinity for adsorption of OsO_4 at room temperature. The mean transfer time for 100 mg of OsO_4 in a 50-cm long Teflon tubing (0.6-mm I.D.) was found to be ~2 h when volume flow rate of the carrier gas (air) was 15 mL/min. The recovery of Os was found to be essentially quantitative. Greater than 99% of the activity was recovered in the first trap, with less than 1% of the activity in the 2nd trap, and no activity on the activated carbon filter. The radioisotopic purity of distilled ^{194}Os in the described thermochromatograph was 100% with a separation factor of $\geq 10^6$ from ^{192}Ir, the predominant radionuclidic impurity.

1. Mirzadeh S., Callahan A. P., and Knapp, F. F., Jr. (1991) Iridium-194 – A New Candidate for Radioimmunotherapy (RAIT) from an Osmium-194/Iridium-194 Generator System. In: Proceedings of the 38th Annual Meeting of the Society of Nuclear Medicine, Cincinnati, Ohio, June 1991, *J. Nucl. Med.*, 32, 1089.

2. Mirzadeh S., Knapp F. F. Jr. and Callahan A. P. (1991) Production of Tungsten-188 and Osmium-194 in a Nuclear Reactor for New Clinical Generators. In: *Proceedings of the International Conference on Nuclear Data for Science and Technology*, May 1991, Julich, FRG.

3. Linder M., (1951) *Phys. Rev.*, **84**, 240.

4. Williams D. C. and Naumann R. A., (1964) *Phys. Rev.*, **134**, B289.

5. Report BNL-325 3rd ed. (Mughabgab S. F. and Carber D. J.) Vol. 1, U.S. Government Printing Office, Washington D.C. (1973).

6. Casten R. F., Namenson A. I., Davidson W. F., Warner P. D. and Borner H. G., (1978) *Physics Letters*, **76B**, 280.

Acknowledgements: Research supported by the Office of Health and Environmental Research, U.S. Department of Energy, contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

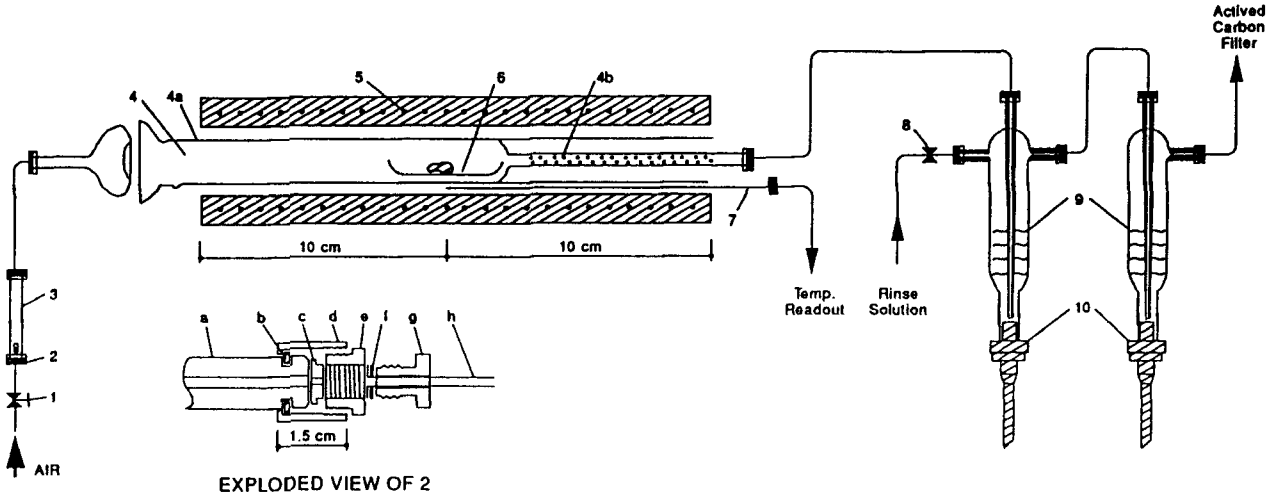


Figure 1. Schematic of the Gas Thermochromatograph for Separation of Neutron Irradiated Enriched Osmium-192.

1. Metering valve, 2. CHEMINERT® glass/Teflon connector (see the exploded view), 3. Air volume flow meter, 4. Chromatographic tube assembly; 4a. Quartz tube (2.8 cm O.D., 2.4 cm I.D. and 20 cm long), 4b. Chromatographic tube (quartz, 6 mm O.D., 12 cm long, containing 60-mesh quartz powder held in the tube by quartz wool, 5. Split furnace, 6. Quartz boat containing Os target, 7. Thermocouple, 8. Sliding Teflon valve, 9. 15-ml extraction funnel containing 2 ml of 0.1 M KOH, 10. Teflon stopcock.

Exploded view of 2. a. Glass connector (2 mm I.D., 6 mm O.D.), b. Split ring (PVC), c. Bed support (Teflon), d. Nut (PVC), e. Bushing (PVC), f. Washer (SS), g. Tube end fitting (PVC), h. Teflon tubing (0.6 mm I.D.) with flanged end.

Paper J17

Spontaneous Electrochemical Separation of Carrier-free ^{64}Cu and ^{67}Cu from Zn Targets.

S. Mirzadeh, D. E. Rice and F. F. Knapp, Jr. Nuclear Medicine Group, Health and Safety Research Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831-6022

Electrochemical processes are attractive techniques for the separation of electropositive metal ions. We report the first application of the spontaneous electrodeposition technique for separation of carrier-free radioisotopes of copper from proton- or neutron- irradiated zinc targets. There continues to be considerable interest in exploring the potential use of ^{67}Cu for therapeutic applications and ^{64}Cu for diagnostic applications, and these radioisotopes of copper are routinely produced at several institutes. We have previously described the application of electrochemical technique for the separation of no-carrier-added ^{67}Cu from a highly radioactive zinc target following irradiation [1]. A similar electrochemical technique has been used at the Los Alamos National Laboratory for routine production of ^{67}Cu [2]. The first application of electrolytic separation of ^{64}Cu under an external electromotive-force (EMF) was reported by Steigman [3] and the application of this technique for the production of no-carrier-added ^{67}Cu was reported later by several investigators [1,2,4].

We have now extended our investigation to spontaneous electrodeposition of carrier-free ^{67}Cu on a platinum electrode. In this process, electrodeposition occurs without an external EMF. The absence of measurable current in this process eliminates the hydrogen overvoltage and preserves the nobility of the Pt electrode. Consequently, the highest degree of separation of $^{67}Cu^{+2}$ from interfering metal ions (i.e., ions of Co, Fe, Ni, etc.) could be expected. The process is intrinsically simple with a minimum of manipulations and, therefore, amenable to automation and remote processing. In addition, methods described herein define a useful and simple methodology for separation of all electropositive metal ions. The results of this investigation also offer further understanding of the chemistry of elements at extreme dilutions and provide a basis for future studies.

The electrolysis apparatus consisted of a U-shape cell (right and left half cells), a Zn electrode (1.5 mm O.D. rod) immersed in a saturated solution of $ZnSO_4$ in 1 M H_2SO_4 (left half cell), and a Pt electrode (1.5 mm O.D. Pt rod spot welded to a 2.5 cm Dia. Pt mesh circle) immersed in a solution of 0.1 M in $ZnSO_4$ and 1 M in H_2SO_4 containing ^{67}Cu (right half cell). The two electrolytes were separated by a porous barrier (10-20 micron fritted glass disc, 2 mm thick, ~2 cm^2 surface area), which allows electrical contact but prevents excessive mixing of the solutions by interdiffusion. To study the effect of low concentration of Zn^{2+} ($\leq 10^{-3}$ M), however, it was necessary to further reduce the mixing of the two electrolytes, and this was achieved by incorporating two fritted glass discs and a 5-ml chamber filled with 1 M H_2SO_4 between the two half cells as shown in Figure 1. The external connection of the two half cells is simply achieved by connecting the Pt and Zn electrodes with a suitable conductor. During the electrolysis, the right electrolyte was mixed magnetically, and the volume of each electrolyte was 25 mL. In a typical study, 10 µL aliquots of the right electrolyte were taken at various time points and analyzed for ^{67}Cu content. At the end of electrolysis, the Pt electrode was removed from the electrolysis apparatus, and the deposited ^{67}Cu dissolved by immersing

the Pt electrode in concentrated HNO_3 for 1-2 minutes. To study the effect of temperature, the right half cell was fitted with a water jacket, and its temperature was thermostatically controlled within ±0.5°C by use of a circulating water bath.

The effect of the duration of electrolysis is shown in Figure 2. At room temperature, the fraction of electroplated ^{67}Cu increased exponentially with time and reached ~95% completion within 30 minutes. The process was found to be independent of copper ion concentration, ranging from $\leq 1x10^{-8}$ M to $1x10^{-3}$ M, and the deposition was found to be rather insensitive to the concentrations of $ZnSO_4$. Quantitative plating can be achieved within 30 minutes when the concentration of zinc was <1.0 M. When the concentration of zinc was ≥1 M, quantitative plating was achieved by extending the duration of electrolysis to 60 minutes. No significant variation was observed in the plating efficiency when the concentration of H_2SO_4 increased by 3 orders of magnitude, from $1.0x10^{-3}$ M to 1.0 M. When the Zn^{+2}/Zn half-cell was replaced with half-cells of Fe^{+2}/Fe, Co^{+2}/Co, Ni^{+2}/Ni, Sn^{+2}/Sn, Fe^{+3}/Fe, Sn^{+4}/Sn, and $PbO_2/PbSO_4/H^+$, quantitative plating of carrier-free ^{67}Cu was achieved within 30 minutes in all cases with the exception of the $PbO_2/PbSO_4/H^+$ half-cell, where no copper was plated. Since the standard half-cell potential (E^0) of $PbO_2/PbSO_4/H^+$ is more positive than Cu^{+2}/Cu by ~1.3 volts, no plating of copper would be expected.

Evaluation of the kinetics of electrodeposition of carrier-free ^{67}Cu yielded a deposition velocity of $(1.50\pm0.27)x10^{-1}$ min^{-1}, corresponding to a deposition constant of ~0.77 $cm.min^{-1}$. Under our experimental conditions, the deposition process was found to be largely irreversible. From the temperature dependence of the deposition velocity constants, an activation energy of 0.18±0.03 eV was calculated. Furthermore, it appears that the thermodynamic spontaneity is satisfied at all concentrations of copper ions. The experimental approach to test the above criteria is severely limited, however, a rigorous test of the above condition could be made by use of carrier-free 9.7-m ^{62}Cu or 5.1-m ^{66}Cu (with theoretical specific activity of $3.1x10^2$ and $1.6x10^3$ Ci/mg) available from the $^{62}Zn/^{62}Cu$ and $^{66}Ni/^{66}Cu$ generator systems, respectively.

In conclusion, we have shown the first application of the spontaneous electrodeposition technique for separation of carrier-free ^{64}Cu and ^{67}Cu from proton- or neutron-irradiated Zn targets. This method has many advantages over the solvent extraction techniques and is simpler to conduct.

1. Mirzadeh S., Mausner L. F. and Srivastava S. C. (1986) Production of no-carrier-added ^{67}Cu. *Int. J. Radiat. Appl. Instrum. Part A, Appl. Radiat. Isot.* 37, 29.
2. Bentley G. and Taylor W. (1984) Electrolytic isolation of Cu-67 from proton-irradiated zinc oxide. *Fifth Int. Symposium Radiopharm. Chem.*, Tokyo, Japan, p.287.
3. Steigman J. (1938) Concentration of radioactive copper by use of a high speed rotating cathode. *J. Phys. Rev.* 53, 771.
4. Maziere B., Stulzaft O., Verret, J. M., Comar D. and Syrota A. (1983) ^{55}Co- and ^{64}Cu-DTPA: New Radiopharmaceuticals for Quantitative Tomocisternography. *Int. J. Appl. Radiat. Isot.* 34, 595.

Acknowledgements: Research supported by the Office of Health and Environmental Research, U.S. Department of Energy, contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

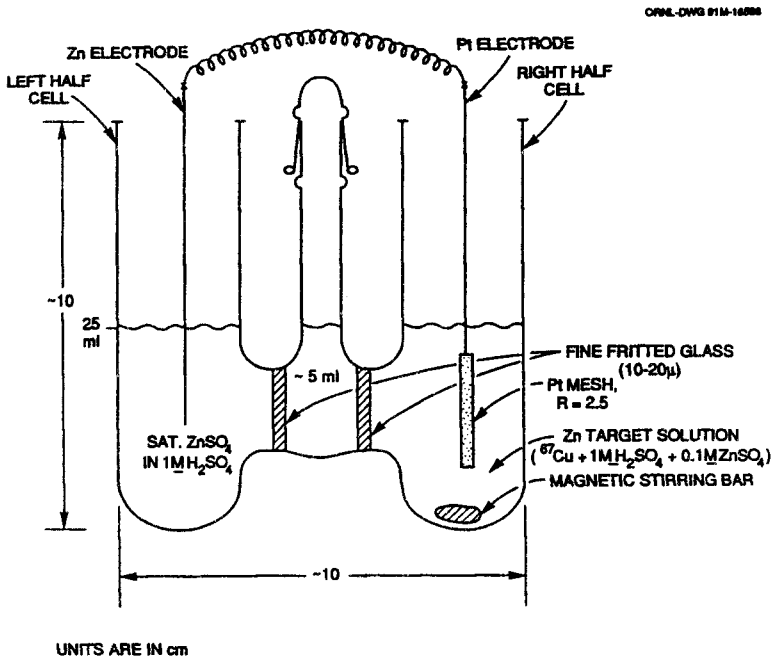


Figure 1. Apparatus for Spontaneous Electrodeposition of Carrier-free ^{67}Cu.

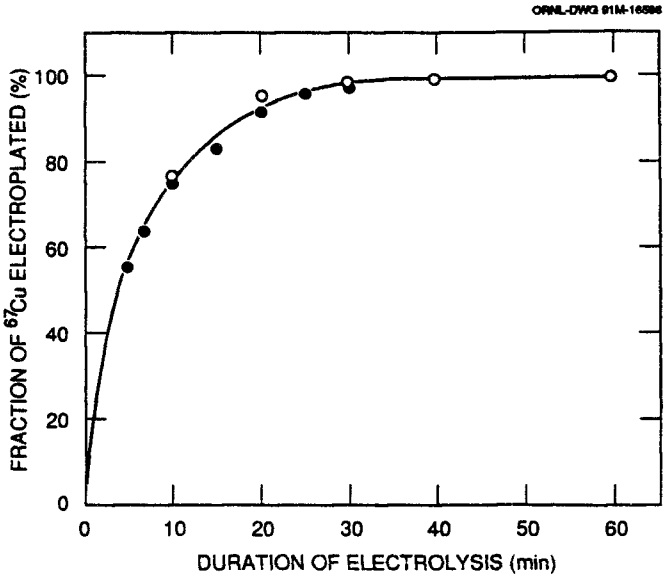


Figure 2. Effect of the duration of Electrolysis.

Composition of Electrolyte:
Left: Saturate solution of $ZnSO_4$ in 1 M H_2SO_4
Right: 1 M H_2SO_4, $ZnSO_4$; $\leq 1x10^{-3}$ (●) or 0.1 M (○)

Paper J18

PRODUCTION and RECOVERY of NON CARRIER ADDED ^{76}Br,^{77}Br SUITABLE for LABELLING of MONOCLONAL ANTIBODIES.

J. Mertens[1], A. Hermanne[1], D. Terriere[1], F. Lambert[2]
[1]VUB-Cyclotron, Laarbeeklaan 103, B-1090 Brussels, [2]Institute of Pharmacy, RUG, B-9000 Gent.

Up to now for several types of monoclonal antibodies electrophilic NCA radiobromination is the only way to obtain a potential PET tracer for tumoral affections. This requires radiobromide solutions of high radiochemical purity. This paper describes a fast and simple recovery chemistry, based on ion-exchange, rather than irradiation optimization.
As the VUB cyclotron permits use of high energy α-beams (up to 43MeV), nuclear reactions on ^{75}As (100 % isotopic abundance) are the obvious choice. Two types of targetmaterial were tried : As powder (purity > 99.9%) and As_2O_3, both sandwiched as a pellet between two aluminium foils.
For the ^{76}Br production a thin pellet(< 100μm) is irradiated with 43MeV α particles using the $^{75}As(\alpha,3n)^{76}Br$ reaction and with a ^{77}Br contamination of less than 2%.
Use of thick pellets (up to 400 μm) and α particles with an energy limited to 30 MeV yield nearly pure ^{77}Br resulting from the ^{75}As (α, 2n)^{77}Br reaction.
Arsenic is dissolved in 3 ml of a 10 % H_2O_2 solution followed by destruction of the H_2O_2 by 350 μl of a 80 % hydrazine hydrate solution. This bulk is made up to 10 ml with H_2O.
As_2O_3 easily dissolves in boiling water without any additive. In case of ^{76}Br production with 43 MeV α particles an important ^{18}F activity is generated making this material less attractive.
The obtained arsenic solution is passed through a mini-column of pretreated Dowex 1-X8 resin (200-400 mesh) in OH^- form (30 mg of resin contained within layers of white sand in a 4 mm diameter seringe) at a flow of 0.5 ml/min. The column is consecutively washed with 3 ml of 2 N NaOH and 10 ml of H_2O. The NCA radiobromide is recovered in ± 500 μl of 1 N $NaHSO_4$ (flow-rate : 0.07 ml/min) with a yield of ± 85 %.
Recovery experiments with ^{76}As-As_2O_3 have shown that > 99.9 % of the amount of arsenic is eluted in the initial and NaOH fractions and that less than 10^{-3} % is present in the $NaHSO_4$ aliquot of interest.

This research is supported by FGWO.grant 3.0099.89

Paper J19

PRODUCTION OF ^{38}K AT A LOW ENERGY CYCLOTRON

S.M. Qaim+, F. Tárkányi*, Z. Kovács*, G. Stöcklin+

+Institut für Nuklearchemie, Forschungszentrum Jülich , 5170 Jülich, Germany

*Institute for Nuclear Research, Hungarian Academy of Sciences,
H-4001 Debrecen, Hungary

The radioisotope $^{38}K(T_{1/2}$ = 7.6 min; β^+ = 100%; E_{β^+} = 2.7 MeV) is a useful tracer for myocardial blood flow studies using positron emission tomography (PET). Among the several suggested methods for its production (for recent reviews cf. Refs. 1, 2) only the $^{35}Cl(\alpha,n)^{38}K$ process using a NaCl target at a medium-sized cyclotron has found routine application (1-3). For production at a low energy medical cyclotron the $^{38}Ar(p,n)^{38}K$ process appeared promising. Some yield values using 30% enriched ^{38}Ar were reported (1). In order to assure the required quality and quantity of ^{38}K, use of highly enriched target gas is mandatory. We measured cross sections and production yields using 95.7% enriched ^{38}Ar.

Stainless steel cells (vol. 8 ml) having thin Al windows were filled with ^{38}Ar to a pressure of 0.5 bar and Cu monitor foils placed in front and at the back of each cell. Due to the short half-life of ^{38}K only one cell was irradiated at a time for 10 min at a beam current of 200 nA. Each irradiated gas cell was counted directly. The absolute activity of ^{38}K was determined via Ge detector γ-ray spectroscopy using the characteristic 2167 keV γ-ray (99.99%). The total error in each cross section value was estimated to be between 12 and 18%.

The measured excitation function of the $^{38}Ar(p,n)^{38}K$ reaction is shown in Fig. 1. In principle the whole excitation function could be used for production purposes. However, in view of the high cost of the target gas the optimum energy range appears to be E_p = 16 → 12 MeV; the calculated saturation thick target yield of ^{38}K for this energy range amounts to 21 mCi (777 MBq)/μA.

The yield of ^{38}K was also determined experimentally under high current production conditions. A small amount of enriched gas was transferred to a conically shaped production target (4) and irradiated for 30 min at 10 μA. A few minutes after EOB the target was evacuated and rinsed with 10 ml water. The ^{38}K activity in the solution was determined. The production yield of ^{38}K was about half of the calculated yield. The level of radionuclidic impurities was < 10^{-5}%.

Our measurements show that the (p,n) reaction on highly enriched ^{38}Ar can easily produce about 60 mCi (2220 MBq) of ^{38}K using a 2 MeV thick target at a low energy cyclotron. The yield via this reaction is by an order of magnitude higher than that via the presently used routine process $^{35}Cl(\alpha,n)^{38}K$ at a medium-sized cyclotron. The cost of the target gas would be about 10000 $ but it can be used repeatedly without much losses.

1. Guillaume M., DeLandsheere C., Rigo P., Czichosz R. - Appl. Radiat. Isotopes 39: 97 (1988)
2. Qaim S.M., Sutisna M.S., Ollig H. - Appl. Radiat. Isotopes 39: 479 (1988)
3. Blessing G. and Qaim S.M. - Appl. Radiat. Isotopes 41: 1229 (1990)
4. Tárkányi F., Qaim S.M., Stöcklin G. - Appl. Radiat. Isotopes 39: 135 (1988)

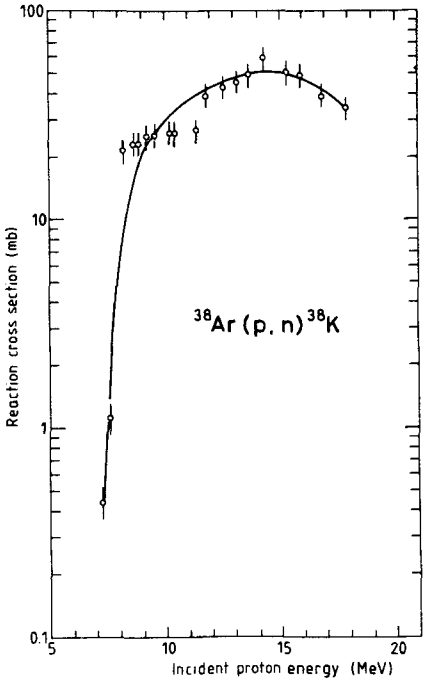


Fig. 1 Excitation function of $^{38}Ar(p,n)^{38}K$ process

Paper J20

Cerium-134/Lanthanum-134, A New Radionuclide Generator System For Positron Emission Tomography

J. Zweit[1], R. Goodall[2], J.W. Babich[1], H.L. Sharma[3] and R.J. Ott[1]
[1]Joint Department of Physics, Royal Marsden Hospital, Sutton, Surrey SM2 5PT, [2]MRC Cyclotron Unit, Clatterbridge Hospital, Bebington, Merseyside L63 4JY and [3]Department of Medical Biophysics, University of Manchester, Manchester M13 9PT, U.K.

Access to generator produced positron emitting radionuclides could expand the use of positron emission tomography (PET) in centres that do not have a local cyclotron. The Ce-134/La-134 is a new radionuclide generator system that could provide a useful source of PET radiopharmaceuticals. This generator produces the positron emitting (62%) La-134 radionuclide which has a short half-life of 6.7 minutes. This is short enough for repeat imaging yet is sufficiently long to allow chemical incorporation into compounds. The half-life (76h) of the parent radionuclide Ce-134 gives the generator a shelf-life comparable to that of the Mo-99/Tc-99m system. The electron capture decay of Ce-134 leads only to x-ray emissions at 33 and 38 keV making the generator easy to shield. Accordingly, we have investigated the development of this new system which includes production and radiochemical processing of the parent radionuclide and development of the generator itself.

For the production of Ce-134, the La-139(p,6n)Ce-134 reaction was investigated. Anhydrous La_2O_3 targets were pressed under vacuum into pellets which were irradiated using the 62 MeV external proton beam from the MRC Cyclotron at Clatterbridge Hospital, U.K. The targets withstood beam intensities up to 10 μA for more than one hour.

The production yields of Ce-134 and other radionuclide impurities obtained from the proton bombardment are shown in table 1. The optimum target thickness is from 61-50 MeV which results in a high Ce-134 yield whilst minimising the level of radionuclide impurities. Short-lived impurities could be further reduced by allowing a reasonable time for the target to cool before chemical processing begins.

The radiochemical processing consisted of target dissolution in 6M HNO_3 and separation by anion exchange chromatography which retained the Ce-134 activity while La was eluted with 0.075M $HBrO_3$/6M HNO_3 mixture. The Ce-134 was then eluted with 0.5M HNO_3. The elution profiles of Ce-134 and La-135 are shown in figure 1. Over 90% of the La-135 was eluted in the first 15 mL and a total eluent volume of 60 mL was used to ensure complete removal of La. Subsequently the Ce-134 was eluted with 0.5M HNO_3 in >90% yield (10 mL).

For the development of the generator system itself, the distribution coefficients of Ce(IV) and La(III) between MnO_2 support and dilute HNO_3 were measured. The results are presented in figure 2 which shows several orders of magnitude difference between the Ce(IV) and La(III) values in the 0.1-0.5M range. Based on these results, we have constructed a generator system in which the Ce-134/0.5M HNO_3 was loaded onto MnO_2 column from which the La-134 daughter was eluted with 0.5M HNO_3. The generator performance was evaluated over a 24-120 hour period after

loading. Following elution of the system void volume, >85% of the available La-134 was obtained in 3-4 mL volume. The Ce-134 breakthrough was <0.001% of eluted La-134 activity.

One of the initial applications envisaged for La-134 would be to label human serum albumin (HSA) through a bifunctional chelate to covalently link the La-134 to the protein. The labelled HSA would then be used as a blood pool imaging agent in a similar way to Cu-62 labelled HSA (1,2).

We have demonstrated the development of a new PET generator that could potentially complement other PET generators currently in use such as the Ge-68/Ga-68 and Sr-82/Rb-82 systems. Experiments are underway to further evaluate and improve the system for potential clinical use. Work on the developments of compounds labelled with La-134 has also been initiated.

Support for this work was provided by the Cancer Research Campaign, the Institute of Cancer Research and the Royal Marsden Hospital.

1) Mathias, C.J.; Welch, M.J.; Green, M.A.; Diril, H.; Meares, C.F.; Gropler, R.J. and Bergmann, S.R. J.Nucl. Med., 32, 475 (1991).
2) Subramanian, K.M. J.Nucl. Med., 32, 480 (1991).

Table 1. Production Yields at EOB of Cerium Radionuclides from Proton bombardment of natural La_2O_3 Targets.

Energy Window (MeV)	Yield (mCi/μAh)				
	Ce-134 (76h)	Ce-135 (17h)	Ce-137g (9h)	Ce-137m (34h)	Ce-139 (138d)
61-45	3.95	14.56	3.90	1.49	0.005
61-50	3.24	7.18	1.67	0.52	0.004
61-55	2.43	3.51	0.82	0.31	0.005

ELUTION PROFILES OF La-135 AND Ce-134 FROM AG1-X8(NITRATE) ADSORBANT

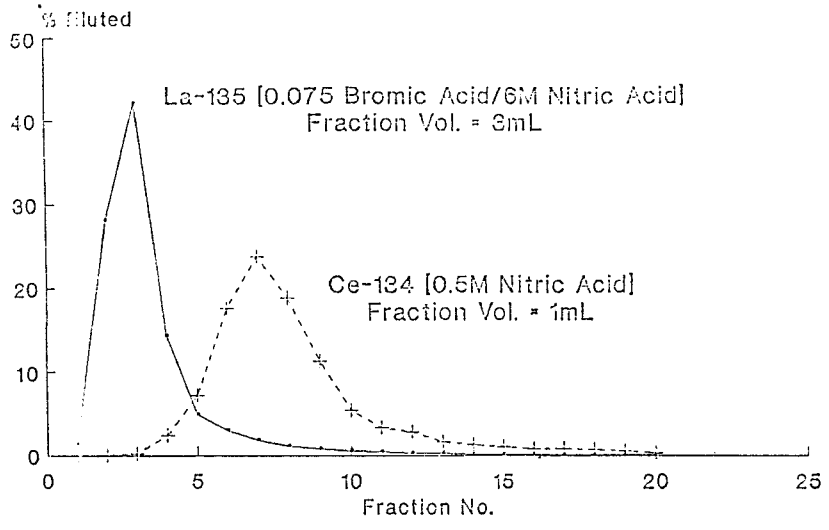


Figure 1

DISTRIBUTION COEFFICIENTS OF Ce(IV) AND La(III) BETWEEM MnO_2 AND DILUTE HNO_3

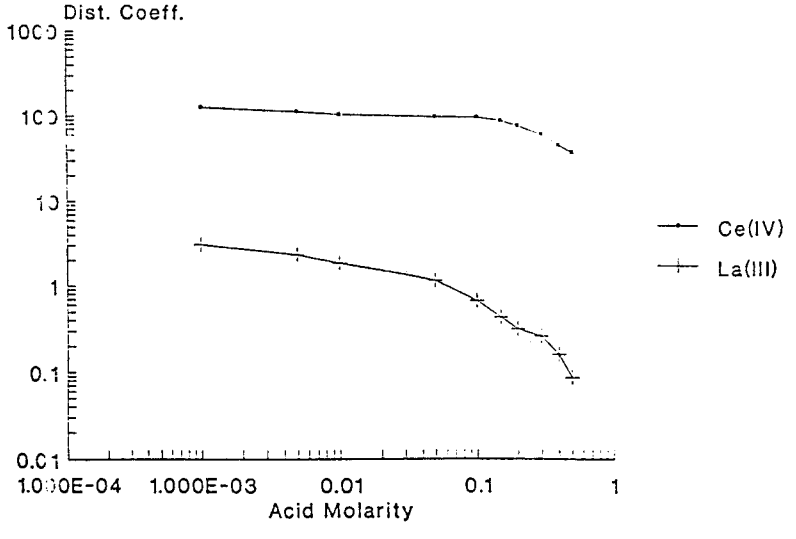


Figure 2

Paper J21

GOLD-199: A POTENTIAL RADIONUCLIDE FOR DIAGNOSTIC PURPOSES

Mauro Bonardi#, Mario Gallorini$, Flavia Groppi# and Gianluca Sanvito#,
#Accelerators and Applied Superconductivity Laboratory, LASA, University and INFN Milano, via F. Cervi 201, 20090 Segrate (MI), $Centre for Radiochemistry and Activation Analysis, CRAA, CNR, Italy.

The gold-198 ($t_{1/2}$=2.70 d; 412 keV) was used since a long time as a radiopharmaceutical for i.v. injection in colloidal form both for diagnostic and radiotherapeutical purposes [1]. The gold-199 ($t_{1/2}$=3.13 d; 158, 208 keV) shows better gamma emissions to be used as a radiodiagnostics and can be obtained in very high specific activity forms. A radiochemical separation has been developed to produce the NCA radiotracer gold-199 via neutron irradiation on platinum target of natural isotopic composition (7.2 % in ^{198}Pt). Dosimetric evaluations for this radionuclide based on the MIRD protocol are in progress.

Gold-199 production via neutron irradiation on platinum:

At present, we are developing a radiochemical procedure to separate NCA gold-199 from Pt targets via ^{198}Pt(n,γ)$^{199m+g}$Pt --> ^{199}Au reactions. The total neutron cross-section for thermal neutrons on platinum-198 is 3.727 barn [2], leading to the theoretical yield for gold-199 of 43 mCi/g of Pt after a 1 day irradiadion with a flux of 10^{13} n/cm^{2}s. This means that it should be possible to obtain about 300 mCi of ^{199}Au after a 1 day irradiation on 500 mg of 100 % enriched ^{198}Pt. The Pt powder can be neutron irradiated in amounts varying up to about 500 mg. The results discussed here refers to a 75 mg amount of platinum (99.9999 %, J&M, UK) irradiated for 1 day with a flux of 9 10^{12} n/cm^{2}s in the central thimble facility of the TRIGA Mark II (thermal + epithermal) research reactor of the University of Pavia, obtaining about 0.1 mCi (3.7 MBq) of gold-199 at the EOP (3 days after the EOB). The present discrepancy between our yield and the theoretical one is under investigation.
The target is processed via two main steps (see also Fig.1):
a. an ion exchange quantitative separation (on tin dioxide, TDO, 30-50 Mesh, Carlo Erba, Italy) of NCA gold-199 from both platinum target and iridium-192g, that is produced by neutron activation on iridium impurities always present in platinum. The ^{199}Au is eluted from TDO by 12 N HCl with a radiochemical yield greater than 70 %. A non tolerable amount of inactive tin is also leached from TDO at this step;
b. purification of the NCA ^{199}Au from Sn impurities by extraction of the gold chlorocomplexes in either diethy- or isoprophylether with a radiochemical yield greater than 98%.

Purity and Specific Activity:

The radionuclidic purity of ^{199}Au is very high and greater than 99.99 % (see Figs.2). The only radionuclidic contaminant present is an almost undetectable amount (about 10^{-3} %) of ^{198}Au that is obtained by neutron activation of stable gold impurities present in the target material (σ = 98.8 barn)[2]. The gold carrier was determined in the final solution by NAA via ^{198}Au, showing a specific activity of about 1 mCi/μg (37 MBq/μg) under the irradiation conditions adopted in this work. The amount of inactive Sn present in the final solution was determined by NAA via ^{125m}Sn and is of the order of 2 μg in total. Work is in progress in order to improve the decontamination factor from tin.

References

1. Simon S. - Medical Atlas of radionuclides used in medicine, biology, industry and agriculture, Report EUR 4606 f,e, CEC, Bruxelles, Belgium, 1972, p. 255.
2. Hughes D.J. and Harvey J.A. - Neutron Cross sections, Report BNL 325, Upton, NY, 1955.

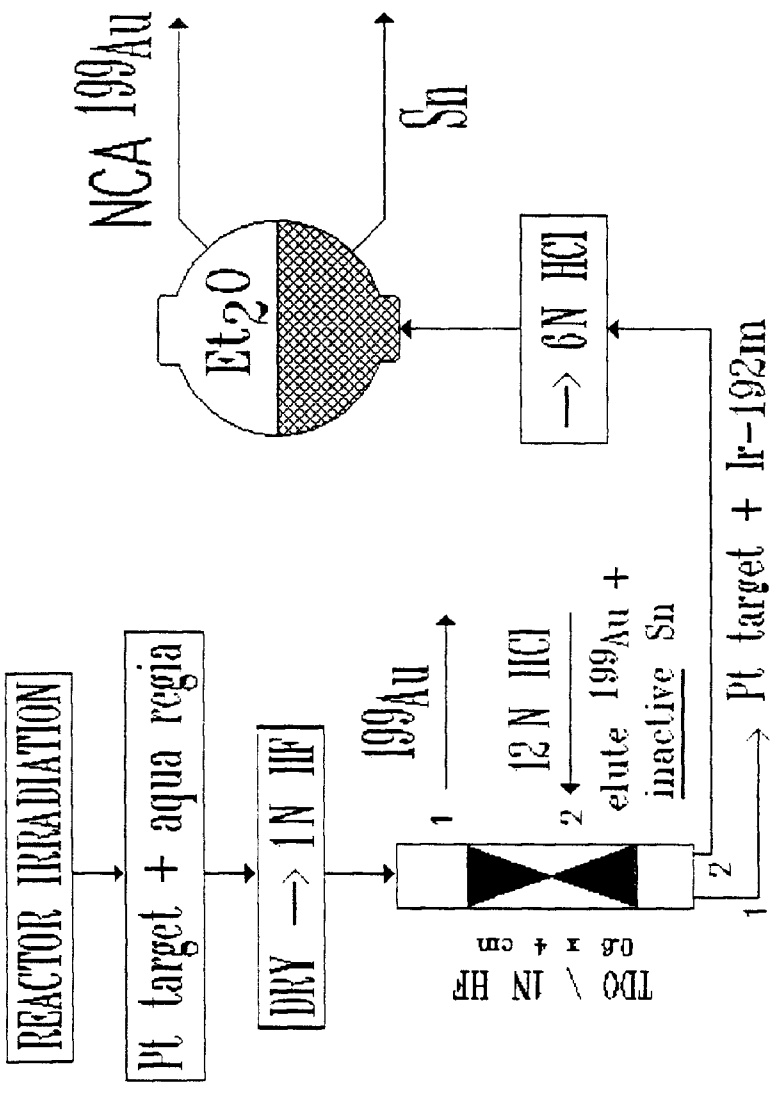
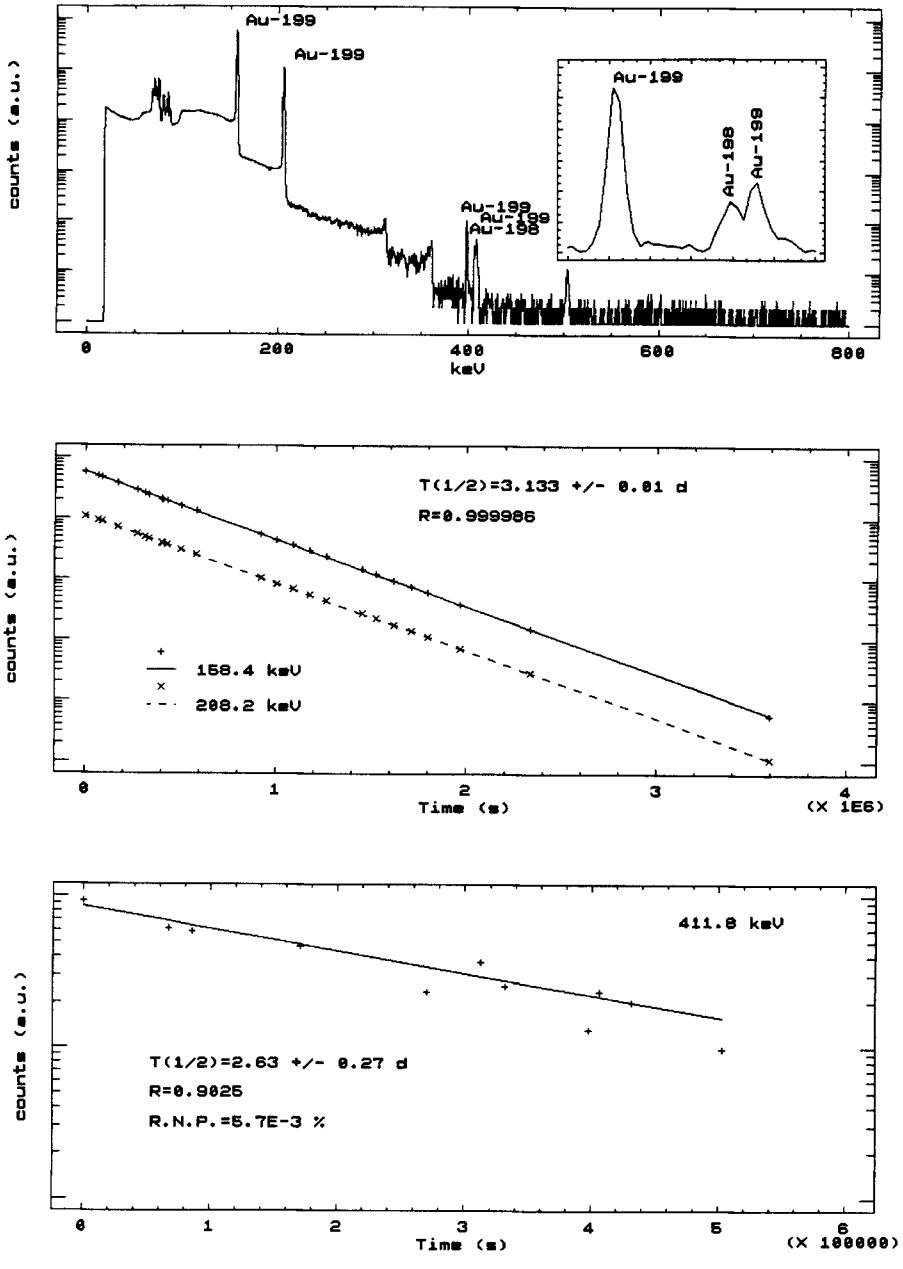


Fig. 1 Simplified scheme of the radiochemical separation of NCA ^{199}Au from Pt target and ^{192g}Ir.



Figs 2 **Decay fittings of high purity ^{199}Au and its radionuclidic impurity ^{198}Au**