#### P001 PREPARATION AND QUALITY CONTROL OF I-123 SOLUTION

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**Introduction:** Sodium Iodide-123 is well known as a radioisotope that is used for studies of the thyroid and its metastases. <sup>123</sup>I labeled compounds are used for kidney, heart and brain studies.

**Experimental:** For  $^{123}$ I production, a home made target was designed. The main part of the target contained; conical target vessel that was mounted in an aluminum cylinder of length 30 cm and diameter 8.5 cm for water cooling during bombardment. Front and rear target windows made by titanium foil and cooling system. In addition, some other part such as a cold finger, decay vessel, vacuum pump and four fingers also designed and installed on the system.  $^{123}$ I was produced from the reaction of 28 Mev protons with 99.99% enriched  $^{124}$ Xe [1] with IBA Cyclone-30. Bombarded xenon was transferred to the vessel and stored there up to about 6 hours to allow  $^{123}$ Cs decay to  $^{123}$ I with the maximum efficiency. The remaining xenon gas was then transferred into the reservoir capsule using a cryogenic technique. The produced,  $^{123}$ I is deposited on the wall of the decay vessel, was rinsed out by dissolution in 0.05  $\mu$ s water at 80°C. Nitrogen gas pressure was used to transfer the  $^{123}$ I solution to the chemical hot lab. The pH, and adjusted with sodium hydroxide.

**Results and Discussion:** A sample of production was sent to quality control. Quality control results were in good accordance with the United States Pharmacopoeia [2]. QC results and production parameters are shown in table 1 and 2 respectively. For detection of chemical impurity which might be leached from target body, produced solution was also analyzed for determination of its chemical impurity, using Atomic Absorption and ICP (table 3).

Table 1

Produced Na <sup>123</sup> I	USP 2005
Clear solution pH = 7.7 Na $^{123}$ I = 98% $^{123}$ I = 99.96%	Clear solution pH = 7.5-9 Na <sup>123</sup> I= 95% <sup>123</sup> I = 99.7%

#### Table 2

Total Activity:	210 mCi
Specific Activity:	3 mCi/ml
Production yield:	3.5 mCi/μAh
Time of bombardment:	2 h
Integrated Current:	60 μAh

Table 3

Element	Wavelength	Concentration (ppm)	RSD
Iron	259.940	0.180	1.5
Aluminium	217.023	-0.049	1.2
Nickel	232.003	-0.061	2.6
Cobalt	238.892	-0.043	2.2
Copper	324.754	-0.013	2.3

**Conclusion:** Using this system with the above specification, the production yield could be increased by increasing the current. The results show no chemical or radionuclide impurity.

**References:** [1] F.Tarkanyi, S.M. Qaim, G. Staeclclin et al. *Appl. Radiat. Isot.* 42 (1991)221. [2] U.S.P. United States Pharmacopeias, *Iodine -123*, 2005.

Keywords: Iodide-123, Production I-123, I-123 Solution, I-123 Target

# P002 EXCITATION FUNCTIONS AND YIELDS FOR Re-186g PRODUCTION BY PROTON CYCLOTRON IRRADIATION

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**Introduction:**  $^{186g}$ Re is a γ-β emitter used in metabolic radiotherapy, thanks to its nuclear properties ( $t_{1/2}$  = 90.64 h,  $E^{max}_{\beta}$  1.07, 0.93 MeV,  $E_{\gamma}$  137 keV), delivering high doses to relatively small tissue volumes, *e.g.* as [ $^{186}$ Re]-HEDP for pain palliation of bone metastases. Presently  $^{186g}$ Re is produced by neutron capture on enriched  $^{185}$ Re in thermal nuclear reactor (together with its long-lived metastable level  $^{186m}$ Re), leading to a relatively low specific activity  $A_S$  of 1-2 MBq· $\mu$ g<sup>-1</sup>, to be compared to the carrier free specific activity of 7 GBq· $\mu$ g<sup>-1</sup>, *i.e.* IDF of ~ 3 000. The possibility to use this radionuclide for therapeutic purposes in labelling of oligopeptides or specific receptor binding molecules is strictly linked to the possibility of increase the  $A_S$  as close as possible to the carrier free value.

**Experimental:** The irradiations were carried out with Scanditronix MC40 Cyclotron (K=38, beam current up to 60  $\mu$ A) of the JRC-Ispra (VA) of CEC, Italy.

 $^{186g}$ Re is produced by proton activation on commercially available thin natural W foils, by using the stacked foil technique at different irradiation energies: several W targets were activated together with Al catcher foils and a Ti monitor was introduced for each irradiation. The activity of the irradiated targets was measured using HPGe detectors and the peaks were analyzed with an appropriate  $\gamma$ -ray analyzing S/W.

**Results and Discussion:** The cross-section data were obtained for the  $^{nat}W(p,n)^{186(g+m)}Re$  reaction and several other radionuclides produced as well; these values were compared both with the literature ones and the model calculation EMPIRE-II of NEA-OECD, with which they appear to be in good agreement. Finally, a simple and reliable radiochemical separation of *no carrier added* Re from W target was developed, based on ion exchange chromatography on activated aluminium oxide.

**Conclusion:** The irradiation of natural tungsten target implies the production of large activities of rhenium radioisotopes with, of course, the co-production of the stable ones. For radiochemical separation purposes, in order to obtain a high specific activity, it is necessary to avoid the presence of the radioisotopes and so enriched W targets are mandatory.

**Acknowledgement:** We acknowledge the whole staff of JRC-Ispra Cyclotron, INFN for financial support (GrV-RENIO experiment) and Doctorate School of University of Study of Milano.

Keywords: Cyclotron, Rhenium-186g, Natural Tungsten, Cross-Section, Thick-Target-Yield

# P003 A FAST AND EFFECTIVE METHOD FOR THE RADIOCHEMICAL SEPARATION OF <sup>211</sup>At/<sup>211g</sup>Po FROM CYCLOTRON IRRADIATED BISMUTH TARGET

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**Introduction:** Among the radionuclides of At, the <sup>211</sup>At is the most promising for labelling compounds for the high-LET metabolic radionuclide therapy (HLRIT). This happens for the following reasons:

 $\Diamond$  the half-life of 7.214 h is long enough to label organic compounds, with methods similar to iodine ones;

 $\diamond$  the  $\alpha$  particles of the  $^{211}$ At/ $^{211g}$ Po system [energy from 5 867.7 keV to 7 450.6 keV, 100% overall abundance] have a range from 60 to 67  $\mu$ m in water, and a LET $_{\infty}$  from 100 to 130 eV ·nm $^{-1}$ , which is nearly the maximum of the RBE curve for energetic ions.

There are some problems with the production and the purification of the  $^{211}$ At from the target and/or from radionuclidic impurities. The purification is necessary to minimise the radioisotopic contamination, as  $^{210}$ Po nuclides, to negligible levels. We present a wet-chemistry method, useful in this step of production of the *no carrier added*  $^{211}$ At/ $^{211g}$ Po, obtained from  $^{209}$ Bi target. The  $\gamma$  emitter  $^{210}$ At was used as *internal spike*.

**Experimental:** The irradiations were carried out at the Scanditronix MC40 cyclotron of JRC-Ispra (Varese, Italy) of CEC at different  $\alpha$  energies. The target was wrapped with Al foil, to prevent Bi melting and At-Po evaporation. A separation, without any addition of isomorphous carrier, of the At radioisotopes from Bi target and Po impurities is mandatory to obtain the NCA  $^{211}$ At/ $^{211g}$ Po. The method is a selective radiochemical separation based on the dissolution of the target with HNO<sub>3</sub>, the dilution with HCl, the L/L extraction with DIPE and the back-extraction with NaOH at various concentrations. High resolution  $\alpha$ ,  $\gamma$  and X spectrometries have been used and liquid scintillation counting -with and without pulse shape analysis- as well.

**Results and Discussion:** We obtained a back-extraction yield greater than 90-97% in a range from 0.75 M to 2.00 M. The wet-chemistry method is effective for the production of a very high specific activity  $^{211}$ At/ $^{211g}$ Po, characterized by a radionuclidic purity close to 100%. In agreement with the energy of the a beam, during the separation some MBq or kBq (due to a A<sub>S</sub>(CF) of 76 MBq·ng<sup>-1</sup>) of At have been separated from 0.20-0.25 g of Bi target with an overall radiochemical yield varying from 10% to 97% vs. NaOH concentration. The co-production of the  $^{210}$ Po by *direct reactions* on Bi, below the threshold (28.6 MeV) for the production of  $^{210}$ At $\rightarrow$ <sup>210</sup>Po, was investigated too.

**Acknowledgement:** We acknowledge the staff of the cyclotron of JRC-Ispra and special thanks to INFN (GrV-experiment ASTATO) and MURST for funding this project.

# P004 A WET-CHEMISTRY METHOD FOR THE SEPARATION OF *NCA* RHENIUM RADIONUCLIDES FROM CYCLOTRON IRRADIATED TUNGSTEN TARGET

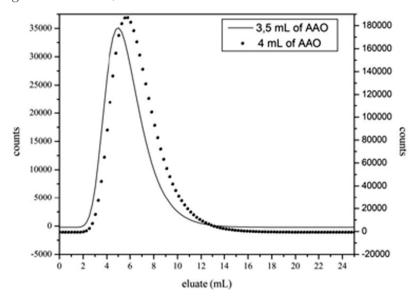
#### E. PERSICO, C. ZONA, E. QUARTAPELLE, L. CANELLA, M.L. BONARDI and F. GROPPI

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**Introduction:** In nuclear medicine radionuclides are used in the treatments of cancer and others diseases. Our attention is focussed on  $^{186g}$ Re, a  $\gamma$ - $\beta$ <sup>-</sup> emitter that, thanks to its nuclear properties, is suitable for pain palliation due to bone metastases. The goal consists in the extension of its applications for radiotherapeutic purposes. So, the first step to reach this objective consists in obtaining *no carrier added* Re and increasing its specific activity  $A_S$ . This step is dealt with the development of a simple and reliable radiochemical separation of Re from W target.

**Experimental:** The irradiations have been carried out at the variable energy Scanditronix MC40 cyclotron of JRC-Ispra. A separation, without voluntary addition of isotopic/isomorphous carrier, of the Re radioisotopes from W target is mandatory to obtain the NCA  $^{18X}$ Re. The wet-chemistry method is a selective separation based on the dissolution -under heating and stirring- of the metallic target with a mixture ( $\sim$ 3:1) of HNO<sub>3</sub> (14.5 M) and HF (24 M), the addition drop wise of pre-heated 30%  $\rm H_2O$  and the heating to remove completely the HF. The final step is the recovery of  $\rm Re^{VII}$  -probably as  $\rm ReO_4^{--}$  with a radiochromatographic method, by using an acidic aluminium oxide column (AAO-C), that retains W and elutes only the  $\rm ^{18X}Re$ . The eluate is collected in different vials, and analysed by HPGe spectrometry.

**Results and Discussion:** In the picture are presented the radiochromatograms obtained during two different separations; *i.e.* we used the  $\gamma$  line at 792.07 keV of the <sup>184m</sup>Re to make calculations. In these experiments we used two different amounts of AAO and we can see that the quantity of AAO influences slightly the position and the shape of the chromatographic peak. In fact, during the different radiochromatographies we obtained radiochemical yields greater than 99%, while less than 1% of <sup>18X</sup>Re remained onto the column. Conversely, W is retained quantitatively.



**Conclusion:** The wet–chemistry method is effective for production of very high  $A_S$  NCA  $^{18X}$ Re, characterized by a radionuclidic purity close to 100% (referred to rhenium). Further quality controls based on ET-AAS and INAA are being carried out in order to study the presence of non-isotopic impurities (*e.g.* W, Ta, Re, Al) co-eluted from AAO-C.

**Acknowledgement:** We acknowledge the staff of JRC–Ispra Cyclotron, INFN for financial support (GrV–RENIO experiment), MURST and Doctorate School of the Università degli Studi di Milano.

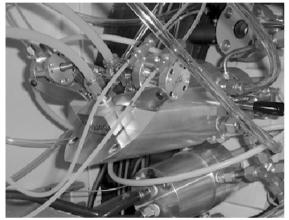
#### P005 EXPANDING THE UTILITY OF A GE PETTRACE CYCLOTRON

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**Introduction:** The NIH cyclotron facility relies heavily on a Cyclotron Corporation CS-30 four-particle and a GE PETtrace two-particle cyclotron for routine radionuclide production. In order to expand the utility of the GE PETtrace, we have built a solid plate target and cup target for production of Cu-60, Cu-64, I-124, and Tc-94m for clinical research use.

**Experimental:** The solid plate target consists of a 53 mm L x 19 mm W x 3 mm H 5N solid gold plate (with a 25 mm L x 7 mm W plated area of Ni-60, Ni-64, or Te-124) held in a water cooled aluminum block positioned 32 degrees to an incident beam of  $12.5 \pm 1.0$  MeV protons (Figure 1). The cup target consists of 200 mg of enriched Mo-94 pressed in the 15 mm diameter grid of a 38 mm diameter x 35 mm deep aluminum target cup held in a water cooled block positioned 90 degrees to an incident beam of  $12.1 \pm 0.5$  MeV protons mounted on a 36 cm L x 5.0 cm diameter beam-line tube (Figure 2). Aluminum degrader foils and Havar target foils are cooled by a flow of chilled helium.



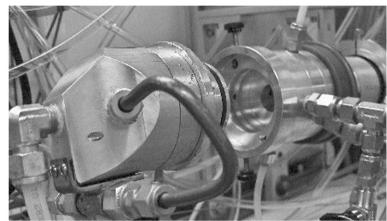


Fig. 1. GE PETtrace plate target.

Fig. 2. GE PETtrace cup target.

**Results and Discussion:** Aluminum foil degrader thicknesses were calculated using TRIM. Resulting proton beam incident energies were determined by calculating the Zn-63/Zn-65 ratio of irradiated 4N Cu foil targets. The GE PETtrace production rates for Cu-60, Cu-64, I-124, and Tc-94m are shown in Table 1 along with the production rates and energy ranges employed with the CS-30 cyclotron internal tangent target system and external cup target for comparison.

Table 1. Production rates for targetry systems

Nuclide	Ep (MeV)	PE	Ttrace	CS	CS-30		
		Targetry	mCi/μA-h (n)	Targetry	mCi/μA-h (n)		
Cu-60	12.5	plate (32°)	$15.2 \pm 3$ (6)	plate (tangent)	$49.4 \pm 9$ (8)		
Cu-64	12.5	plate (32°)	$1.5 \pm 0.2$ (6)	plate (tangent)	$10.5 \pm 3$ (28)		
I-124	12.5	plate (32°)	$0.018 \pm .005$ (4)	plate (tangent)	$0.025 \pm .005$ (8)		
Tc-94m	12.1	cup	$10.6 \pm 3$ (4)	cup	$10.1 \pm 3 (30)$		

**Conclusion:** We can routinely prepare research quantities of Cu-60, Cu-64, and Tc-94m at the NIH using the GE PETtrace cyclotron. The radionuclides are suitable for the clinical preparation of Cu-ATSM and Tc-94m Sestamibi.

Keywords: Cyclotron Targetry, GE PETtrace, Cu-64, I-124, Tc-94m

### P006 103Ru/103mRh GENERATOR

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**Introduction:** For radiotherapy of small tumors and metastases radionuclides emitting charged particles with short ranges (a few mm) are required. These criteria are met by alpha and Auger emitters. Using various selection criteria like electron energy, suitable half-life, low photon/electron ratio and availability Bernhardt and coworkers [1] found that the from known Auger emitters <sup>103m</sup>Rh best fit such criteria.

Carrier-free <sup>103m</sup>Rh is formed as a decay product of <sup>103</sup>Ru, which can be produced by the fission of uranium or by neutron irradiation of <sup>102</sup>Ru enriched target. Since <sup>103</sup>Ru has a fairly long half-life (39 days), it is possible to use it as a generator that would allow frequent withdrawal of <sup>103m</sup>Rh. The goal of the present work is elaboration of <sup>103</sup>Ru/<sup>103m</sup>Rh generator for milking therapeutic quantity of <sup>103m</sup>Rh.

**Experimental:**  $^{10\bar{3}}$ Ru, mother nuclide of  $^{103m}$ Rh, was obtained by neutron irradiation of natural ruthenium target at a neutron flux  $3x10^{14}$  n/cm<sup>2</sup>/s for 36 h. Next the target was dissolved in mixture of NaOH and KIO<sub>4</sub>. After complete dissolution of the target solution was acidified and formed RuO<sub>4</sub> was extracted to an organic (CCl<sub>4</sub>) phase. To avoid reduction of RuO<sub>4</sub> to RuO<sub>2</sub> the organic phase was contacted with a solution generating Cl<sub>2</sub> molecules (1 M HCl+KIO<sub>4</sub>). Milking of  $^{103}$ Rh was performed with contact of organic phase with 0.01 M solution of H<sub>2</sub>SO<sub>4</sub>.

The aqueous phase was purified by 3 times extraction with CCl<sub>4</sub> and finally, by passing trough an anion exchange resin.

**Results and Discussion:** The  $Cl_2$  molecules, formed in 1 M HCl+KIO<sub>4</sub> solution, distribute between the phases keeping ruthenium in the organic phase as RuO<sub>4</sub>. In the back-extraction (0.01 M H<sub>2</sub>SO<sub>4</sub>)  $^{103m}$ Rh is totally transferred to aqueous phase and RuO<sub>4</sub> has a partition coefficient of 40-50, which means that a small fraction of the ruthenium will be back-extracted with the rhodium. In the aqueous phase impurities of  $^{103}$ Ru are in the form of RuO<sub>4</sub>, but a small quantities of the ruthenium is reduced *e.g.* to RuO<sub>4</sub> or RuO<sub>2</sub> which are difficult to remove. The RuO<sub>4</sub> was removed by few times washing with a pure organic solvent. The residue of  $^{103}$ Ru in aq. phase was absorbed in the form of RuO<sub>4</sub> on anion exchange resin.

**Conclusion:** The separation process containing 3 times solvent extraction with  $CCl_4$  and adsorption on anion exchange gave separation factors between  $^{103m}Rh$  and  $^{103}Ru$  of more than  $5x10^3$ .

Acknowledgement: The research was supported by a EC grant POL-RAD-PHARM

Reference: [1] P.Bernhardt, E. Forssell-Aronsson, L. Jacobson, G. Skarnemark, Acta Oncologica 40, 602–608, 2001.

Keywords: Rh-103m Generator, Auger Emitters

### P007 PRODUCTION AND QUALITY CONTROL OF 55 Co AS A PET RADIONUCLIDE

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**Introduction:** The most commonly used cobalt radionuclide,  $^{55}$ Co, provides very good physical properties for diagnostic PET imaging [ $T_{1/2}$ =17.53h, Decay Mode:  $\beta^+$  (60%), E.C. (40%), Main gamma ray energies: 511(154%), 477.2 (20.2%), 931.3 (75%), 1316.4 (7.09%), 1408.4 (16.88%)keV].  $^{55}$ Co has been used for PET imaging studies of recurrent ischaemic strokes, primary brain tumors, vascular dementia, renal imaging, blood cell radiolabeling, monoclonal antibodies, atheroschlerotic carotid artery disease and quantitative tomocisternography. It has also been used as a therapeutic agent for implantation of  $^{55}$ Co stents.

**Experimental:** The radionuclide  $^{55}$ Co was produced via the  $^{nat}$ Ni(p,x) $^{55}$ Co process [1]. The high current  $^{nat}$ Ni target was electroplated on a gold-plated Cu backing [2]. A 30μm layer of Ni was enough due to the target angle. The target was irradiated by a 150μA current of 15MeV protons (15-8MeV on the target). The irradiated target was dissolved in refluxing warm 7M HNO<sub>3</sub> (20ml) for 30 min. It was heated almost to dryness and dried again after the addition of 20ml distilled water. The residue was reconstituted by 9M HCl (20ml) and passed through an anion exchange resin (AG1X8, Cl<sup>-</sup> form, 100-200 mesh, h: 8cm, Ø: 1cm) preconditioned with 25ml of 9M HCl. The column was washed by passage of 50ml of 9M HCl to remove Ni ions. Co and Cu ions were recovered by passage of 20ml of 4M HCl through the column. The eluent was evaporated to dryness and dissolved in 25ml of 0.3M HCl-94% ethanol and was then injected to another anion exchange resin (AG1X8, Cl<sup>-</sup> form, 100-200 mesh, h: 5cm, Ø: 1cm) pre-equilibrated with 25ml of 0.3M HCl-82% ethanol. More than 95% of Co ions were recovered as  $^{55}$ Co-CoCl<sub>2</sub> by passage of 25ml of 0.3M HCl-72% ethanol. Quality control was performed in two steps:

*Radionuclide purity Control:* Radionuclide purity of the product was controlled by gamma spectroscopy of the final sample using an HPGe detector coupled with a Canberra<sup>TM</sup> MCA. The peaks were observed for 1 h.

*Chemical purity Control*: The presence of Ni cation was detected using visible colorimetric assays. DMG reacts with Ni ions in a neutral or ammoniacal medium to form a pink, flocculent precipitate.

The amount of Au cation was controlled in the final solution using color formation with acidic rhodamine B reagent. **Results and Discussion:** The production yield was 270.2  $\mu$ Ci/ $\mu$ Ah at E.O.B. The radiochemical separation yield was 95%. The radionuclide purity was >99.3% (<0.7%  $^{57}$ Co). No nickel ions were detected in the sample with a detection limit of 2 ppm. Gold concentration was less than 0.9 ppm.

**Conclusion:** The method used in this study for the production and radiochemical separation of <sup>55</sup>Co using natural nickel targets was simple and cost effective, with high production yields and low levels of contamination.

**References:** [1] P. Reimer and S.M. Qaim, Radiochimica Acta, 80 (1998), 113-120. [2] S. Spellerberg, P. Reimer, G. Blessing, H.H. Coenen, S.M. Qaim, Appl. Radiat. Isot., 49(1998), 1519-1522.

Keywords: 55Co, production, Targetry, Radiochemical separation, Quality control

# P008 ENHANCED PROSPECTS OF <sup>99m</sup>Tc GEL GENERATOR USAGE WITH THE AID OF POST-ELUTION CONCENTRATION OF PERTECHNETATE

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**Introduction:** The diagnostic imaging in nuclear medicine continues to be centred on  $^{99m}$ Tc with its use accounting for  $\sim$ 80% of the 25 million studies per annum. Assuring reliable, continued, world-wide availability of  $^{99m}$ Tc is hence of paramount importance. There are emerging trends such as preference to avail products from centralised radiopharmacy services, need-based growth of small-scale production facilities in developing countries, development of post-elution concentration techniques for pertechnetate solution, advances in automation of radiochemical processing/synthesis and a need to address the consequences of phasing out of highly enriched uranium (HEU) from commercial use (as HEU targets are employed for production of fission  $^{99}$ Mo). This paper makes out a case for meeting limited demands of  $^{99m}$ Tc by enhancing usage of gel generator after coupling with on-line post-elution concentration option.

**Results and Discussion:** The processing of irradiated molybdenum trioxide into a free-flowing zirconium molybdate - <sup>99</sup>Mo gel powder has been reported, while details of engineering features of the shielded handling facility are less well-known (1,2). While the original concept came from ANSTO, many other groups have sustained interest in this method. In China nearly 25% of <sup>99m</sup>Tc needs are met by gel generators, while two batches (370GBq lots) of gel generator per month are produced in India. In Kazakhstan gel generator is prepared twice a month for centralised distribution of separated <sup>99m</sup>Tc. Brazil has set up an automated processing facility and plans production after reactor power upgrade to obtain <sup>99</sup>Mo of required specifc activity. A new facility is under construction in Egypt based on Chinese technology.

The capacity of gel generator is 10-35 GBq depending upon the initial specific activity of <sup>99</sup>Mo. Adding the provision of post-elution concentration to gel generators will enhance its capacity and duration of utility, as one can use larger gel beds after reckoning with the additional shielding requirements. The dual-purpose use of alumina trap column for purification from <sup>99</sup>Mo traces and concentration of pertechnetate eluate in deionised water (3) and automation of radiochemical processing feasible nowadays, enable 20-100 GBq gel generators in regular practice.

**Conclusion:** This work under a CRP would diversify supply options, especially for countries with limited resources and strengthen availability of <sup>99m</sup>Tc through enhancing utilisation of medium flux research reactors.

**References:** [1] IAEA-TECDOC-852 (1995). [2] Saraswathy P, et al. in IAEA-TECDOC-1029 (1998) 385-397. [3] Sarkar SK, et al. Nucl Med Commun 25 (2004) 609-614.

Keywords: Tc-99m Gel Generator, Post-Elution Concentration, Mo-99 Specific Activity, Zirconium Molybdate Gel, Alumina Trap Column

# P009 MEASUREMENT OF THE THICK TARGET YIELDS FOR $^{nat}$ S( $\alpha$ ,x) $^{34m}$ CI REACTION AND ESTIMATION OF ITS EXCITATION FUNCTION UP TO 70 MeV

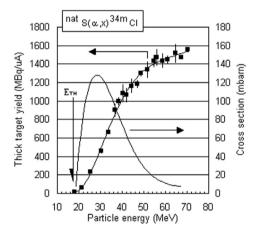
### K. NAGATSU<sup>1</sup>, T. FUKUMURA<sup>1</sup>, F. SZELECSÉNYI<sup>2</sup>, Z. KOVÁCS<sup>2</sup> and K. SUZUKI<sup>1</sup>

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**Introduction:** Chlorine-34m ( $T_{1/2}$ =32.0min,  $\beta^+$ =55.4%, IT=44.6%) has been recognized as an important PET radionuclide. Several methods have been reported for the production of <sup>34m</sup>Cl. Among them the <sup>32</sup>S( $\alpha$ ,x)<sup>34m</sup>Cl process seems to be preferable since it has relatively high yield at a low production cost; however, no information on its excitation function is available. To optimize the production energy window, we tried to estimate the cross sections, by measuring in detail the thick target yield as a function of energy up to 70 MeV.

**Experimental:** Three grams of natural sulfur crystal ( $^{32}$ S = 95atom%, purity 99.999%) placed into an aluminum target cell (Ø  $16 \times 10$ mm) was melted to prepare a uniform target. This target was thick enough to stop all beam energies up to 70 MeV. Before irradiations, the target cell was sealed with a 20  $\mu$ m Al foil. Irradiations were carried out with 40, 50, 65 and 74 MeV alpha particles at 10 nA for 3 min at the NIRS AVF-930 cyclotron. Using Al energy absorber, the incident energy on target was varied between 14.4 to 70.3 MeV.

**Results and Discussion:** The thick target yield of the  $^{\rm nat}S(\alpha,x)^{34\rm m}Cl$  reached 1.63 GBq/ $\mu$ A at 70 MeV (Fig.1). To evaluate the optimum production energy window, cross sections were also calculated from the fitted curve of thick target yield. The excitation function curve showed a maximum of 128 mbarn at 28 MeV (Fig.1).



**Conclusion:** According to our study, more than 1.3 GBq/ $\mu$ A (35 mCi/ $\mu$ A) <sup>34m</sup>Cl can be produced in the E $\alpha$ =50 $\rightarrow$ 22 MeV energy region, that seems to be enough for practical PET studies.

Keywords: Chlorine-34m, natS(a,x)34mCl, Cross Section, Thick Target Yield

# P010 A PROOF OF PRINCIPLE ION SOURCE FOR INCREASING THE SPECIFIC ACTIVITY OF REACTOR PRODUCED Re-186

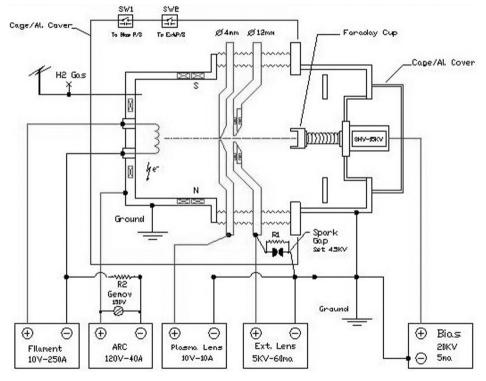
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**Introduction:**  $^{186}$ Re ( $^{6}$ ,  $^{6}$ ,  $^{1}$ ,  $^{1}$  = 3.7 d) is a promising radionuclide for targeted radiotherapy. Several groups have already reported success with  $^{186}$ Re -labeled antibodies, both in animal and human models.  $^{186}$ Re is commonly produced in reactors by a neutron capture reaction on  $^{185}$ Re. This results in a product with low specific activity which is not optimal for site-specific targeting. This work focuses on the development of an ion source for investigating the feasibility of an ionization and mass separation approach to producing  $^{186}$ Re in high specific activity.

**Experimental:** A cusp-type ion source device for this purpose has been constructed at TRIUMF. This type of ion source utilizes a magnetically contained plasma for the ionization of gas phase species.  $^{186}$ Re as HReO<sub>4</sub> is dried on a quartz boat and placed on a tantalum stand in the center of the plasma chamber. A plasma is created by the addition of hydrogen gas, which is ionized by a heated tantalum filament. The plasma heats the quartz boat and causes the HReO<sub>4</sub> to enter the gas phase. Negative rhenium oxide species created in the plasma are accelerated toward the faraday cup, which is maintained at 15 kV positive to ground. The ions are implanted in the faraday cup, which can be removed for assessment of ionization efficiency.

**Results and Discussion:** This ion source device has been successfully commissioned with accelerated H<sup>-</sup> ion beams. At present we are conducting tests with generator produced <sup>188</sup>Re. The shorter half-life of this isotope allows for a shorter turnaround time between experiments. Preliminary tests indicate that up to 20% of the rhenium in the gas phase may form a negative ion oxide species that can be extracted by our system.



**Conclusion:** A proof of principle ion source to investigate the feasibility of an ionization and mass separation approach to improving the specific activity of reactor-produced <sup>186</sup>Re has been constructed and successfully commissioned. If this system performs as expected, we plan to construct a magnetic mass separation device to be coupled to this ion source for further experiments.

**Acknowledgement:** The authors would like to thank Mike McDonald, Ray Dube, Ken Buckley and Wayne Sievers at TRIUMF for valuble advice and technical support. This work is funded by NSERC I2I and TRIUMF Life Sciences.

Keywords: 186Re, Ionization, Mass Separation, Specific Activity

# P011 INVESTIGATION OF NEW ROUTES FOR PRODUCTION OF THE THERAPEUTIC RADIONUCLIDES 169 Yb AND 165 Er

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**Introduction:** The radionuclides  $^{169}$ Yb ( $T_{1/2}$  = 32.0 d, EC = 100%) and  $^{165}$ Er ( $T_{1/2}$  = 10.36 h, EC = 100%) are almost pure Auger electron and X-ray emitters and have been gaining interest in radiotherapy. Large scale production of these radionuclides could be done via the  $(n,\gamma)$  reaction at nuclear reactors. This production route yields a product of low specific activity, therefore, alternative routes resulting in no-carrier added product utilizing charged particle induced processes on the stable isotopes of holmium, erbium and ytterbium, seemed to be interesting.

**Experimental:** The excitation functions of the <sup>nat</sup>Yb(p,xn)<sup>169</sup>Lu (decays into <sup>169</sup>Yb), <sup>nat</sup>Er( $\alpha$ ,xn)<sup>169</sup>Yb and <sup>165</sup>Ho(d,2n)<sup>165</sup>Er processes were measured up to E<sub>p</sub> = 36 MeV, E<sub> $\alpha$ </sub> = 36 MeV and E<sub>d</sub> = 21 MeV by using the well known stacked-foil activation technique. Irradiations were done at the external beam of the CGR-560 cyclotron of VUB. The activity of the produced nuclei was determined by gamma- and X-ray spectroscopy.

**Results and Discussion:** The excitation functions of  $^{nat}Yb(p,xn)^{169}Lu$  and  $^{165}Ho(d,2n)^{165}Er$  reactions have been measured for the first time in this work. For  $^{nat}Er(\alpha,xn)^{169}Yb$  two earlier experimental data sets were found in the literature, showing large disagreement with our new results. The experimental data were compared to the results of the nuclear model codes ALICE-IPPE and EMPIRE-II, showing good agreement for proton and alpha induced reactions and being less successful for description of the (d,2n) process. For a comparison of the hitherto investigated charged particle induced reactions we calculated the integral yields of  $^{169}Yb$  and  $^{165}Er$  from various experimental excitation functions.

Energy		Integral yield of 169Yb (MBq/ $\mu$ Ah)				Integral yield of 165Er (MBq/μAh)	
(MeV)	169Tm(p,n)	169Tm(d,2n)	natEr(α,xn)	natYb(p,xn)169Lu decay	165Ho(p,n)	165Ho(d,2n)	
10	0.40	0.30	0.00	0.01	6.86	22.6	
15	1.43	2.22	0.00	0.03	58.2	178.3	
20	1.77	3.90	0.02	0.25	82.0	294.3	
30	2.41		0.28	3.23			
40	3.04		0.81	10.8			

**Conclusion:** Using optimal energy ranges the  $^{nat}Yb(p,xn)^{169}Lu$  indirect route is the most effective for production of  $^{169}Yb$ . The  $\alpha$ -particle beam is not competitive with protons and deuterons. We have found that the maximal cross section of the (d,2n) reaction on Ho is significantly higher than that of the (p,n) reaction.

Keywords: Ytterbium-169, Erbium-165, Therapeutic Radionuclides, Cross Section, Cyclotron

### P012 PRODUCTION OF 87,88Y AND 89Zr VIA ALPHA-PARTICLE INDUCED REACTIONS ON nat Rb AND nat Sr

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**Introduction:** The radioisotope  $^{87}$ Y ( $T_{1/2}$ = 79.8 h) finds application in SPECT. Furthermore, its use as  $^{87}$ Y/ $^{87m}$ Sr generator has also attracted considerable interest. The daughter  $^{87m}$ Sr ( $T_{1/2}$ = 2.8 h) is used substitutionally for SPECT studies of calcium metabolism. The long-lived  $^{88}$ Y ( $T_{1/2}$ =106.6 d) has gained some attention for investigation of slow metabolic processes, especially in animal experiments. The radionuclide  $^{89}$ Zr ( $T_{1/2}$ = 79.4 h) has been suggested for PET studies of labelled antibodies.

The three radionuclides are generally produced via proton or deuteron induced reactions. We studied their production by alpha-particle induced reactions.

**Experimental:** Measurements of the excitation functions of the reactions  $^{nat}Rb(\alpha,xn)^{87.88}Y$  and  $^{nat}Sr(\alpha,xn)^{89}Zr$  using thin  $^{nat}RbCl$  and  $^{nat}SrCO_3$  samples were done up to 26 MeV  $\alpha$ -particles using the stacked-foil technique. For the separation of n.c.a.  $^{87.88}Y$  from Sr and Rb, and of n.c.a.  $^{88.89}Zr$  from Y and Sr, ion-exchange and solvent extraction techniques were studied using the resins Dowex 50W-X8 and 21K and the complexing agents HDEHP and TPPO.

**Results and Discussion:** The measured excitation functions  $^{nat}Rb(\alpha,xn)^{87m,87m+g,88}Y$  and  $^{nat}Sr(\alpha,xn)^{89,88}Zr$  are in fair agreement with earlier measurements [1,2]. The present data strengthen the database especially below 15 MeV. The chemical separations via ion-exchange and solvent extraction not only provided the radionuclides in a pure form but also led to useful information on the distribution coefficients in dependence of the HCl-concentration and pH-value, respectively.

**Conclusion:** The calculated yield of  $^{87}$ Y over the energy range of interest is 8.2 MBq/ $\mu$ Ah, with relatively low  $^{88}$ Y-impurity. On the other hand, the  $^{88}$ Y-yield is 0.08 MBq/ $\mu$ Ah, and a high-purity product can be obtained after a decay time of about two months. The yield of  $^{89}$ Zr is 0.9 MBq/ $\mu$ Ah. These yields are much lower than in p- and d-induced reactions.

For the separation of n.c.a. yttrium, the cation exchange chromatography using Dowex 50W-X8 was found to be the best, while for the separation of n.c.a. zirconium solvent extraction using TPPO is superior. Those methods gave separation yields of about 80% and the separated products were of high radionuclidic and chemical purity.

**Acknowledgement:** This work was done under an Egyptian-German bilateral agreement and we are grateful to the concerned authorities for their support.

**References:** [1] Iwata, S., J. Phys. Soc. Japan 1962; **17:** 1323-1333. [2] Levkovskij, V.N., Activation Cross Sections by Proton ans Alphas, Inter-Vesti, Moscow, 1991.

Keywords: Alpha-Particle Induced Reactions, Chemical Separation, Y-87, Y-88. Zr-89, Production Yield

# P013 THICK TELLURIUM ELECTRODEPOSITION ON NICKEL COATED COPPER SUBSTRATE FOR 1241/1231 PRODUCTION

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**Introduction:** Due to its favorable nuclear properties, the 4.18 d radionuclide  $^{124}$ I ( $22\pm0.5\%$   $\beta^+$ ,  $E_{\beta}^+$  = 2.13 MeV, 78% E.C.) has great potential for application in nuclear medicine. Accelerator production of  $^{124}$ I is largely achieved via nuclear reactions  $^{124}$ Te(p,n) $^{124}$ I and  $^{124}$ Te(d,2n) $^{124}$ I which are well suited for medium to low-energy cyclotrons.

**Experimental:** In a 600 ml beaker, Te (6 g/l) was added, followed by the addition of KOH and deionized water (250 ml). The homogenized solution was filtered through fine glass filter (0.45 m) to remove any residual particles if necessary to be ready for electroplating. Non-reactive plating vessels are hollow Perspex cylinders (diameter 6 cm, height 20 cm) fitted with an axial Pt anode wire mounted at the bottom by means of a tube-end fitting with perforated septum. Four symmetrical windows (11.7 cm²) on the vertical side wall allow up to four copper targets backing positioning. Windows liquid-tight sealing is realized by stainless steel mechanical pestles mounted on a PVC ring surrounding plating vessel and by pressing the copper backing against O-ring seal. An external PVC ring is fitted with four supporting pins to hold a motor-stirrer combination in position. The stirrer is a hollow perforated Perspex cylinder mounted on the axis of a DC motor and surrounding the platinum anode. The stirrer rotation speed is set at 1000 rpm; during the process, its rotating direction is reversed after 8 seconds improving deposit homogeneity.

**Results and Discussion:** The electroplating efficiency of the new basic electroplating solution of tellurium which contains potassium hydroxide as complexing agent for tellurium has increased to nearly quantitative. The new system produced 20 high quality plated targets from each electroplating solution bath taking in consideration the depletion of the solution does not go more than 40%. When the same amount of NaOH solution was used to dissolve the proper amount of tellurium instead of KOH, electroplating progresses smoothly. However, after sometime, the electroplated targets started to dissolve and the whole solution turned to dark color. This dark color originated from undissolved tellurium oxides.

**Conclusion:** Tellurium electrodeposition on a nickel coated copper substrate was investigated for production of Iodine-124. The electrodeposition experiments were carried out by the alkali plating baths. The optimum conditions of the electrodeposition of tellurium were as fallows:  $6\,g/1$  Tellurium, pH=10, DC current density of ca  $8.6\,mA0cm^{-2}$  and room temperature. Tellurium thickness was obtained more than  $40~\mu m$ . Lambrecht et al. have reported the electrodeposition of tellurium but haven't reported electroplating condition and also their results have showed tellurium thickness was less than  $22~\mu m$ .

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Keywords: Thick electrodeposition, Tellurium target, <sup>124</sup>I, Production, TeO<sub>2</sub>

# P014 EXCITATION FUNCTIONS FOR PRODUCTION OF MEDICALLY RELEVANT RADIOISOTOPES IN DEUTERON IRRADAITIONS OF Pr AND Tm TARGETS

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**Introduction:** Radionuclides of rare earth elements are gaining importance as emerging therapeutic agents in nuclear medicine. Some of them such as <sup>169</sup>Yb are included in an ongoing IAEA-CRP on "Production of Therapeutic Radioisotopes" while others like the <sup>140</sup>Nd/<sup>140</sup>Pr pair attract interest for their possibility to combine therapeutic and imaging properties in a single radiolabelled compound. In the frame of our ongoing study of light particle induced reactions leading to medically relevant radionuclides we present here our measurements on deuteron induced reactions on Pr and Tm targets up to 40 MeV incident energy.

**Experimental:** Specially prepared  $Tm_2O_3$  or  $Pr_6O_{11}$  targets, deposited on Al or Ni backings, were arranged in several stacks. Ti monitor foils are inserted and irradiations performed in the external beam lines of the Tohoku University (Sendai) and Vrije Universiteit Brussels cyclotrons with deuteron beams of up to 40 or 20 MeV, respectively.

Remeasurement of cross sections of the monitor reactions  $^{nat}$ Ti(d,x) $^{48}$ V and  $^{nat}$ Al(d,x) $^{22,24}$ Na over the whole energy range results in determination of beam characteristics with reduced uncertainty and allows a check of the median energy in each target foil.

**Results and Discussion:** The HPGe gamma spectrometric analysis at different times after EOB of the Pr containing targets results in determination of direct and cumulative production cross sections (without chemical processing) of the radionuclides  $^{139\text{m.g}}$ Nd,  $^{141}$ Nd,  $^{139}$ Pr and  $^{139}$ Ce determined through their gamma radiation and of  $^{140}$ Nd measured through the annihilation radiation of its decay product  $^{140}$ Pr.

A similar data processing applied to the Tm targets allows determining cross sections for several longer lived Tm radioisotopes and for  $^{169}$ Yb where we extended results on the  $^{169}$ Tm(d,2n) reaction measured earlier [1] up to 21 MeV.

**Conclusion:** Thick target productions yields were deduced from spline fits to the experimental cross section points and comparisons with yields obtained by proton induced reactions on similar target materials are discussed.

The experimental excitation curves are also compared with the data obtained from theoretical model calculationd done by the Obninsk group (ALICE-IPPE and EMPIRE II).

**Reference:** [1] F. Tarkanyi, A. Hermanne, S. Takacs, F. Ditroi, I. Spahn, S.F. Kovalev, A.V. Ignatyuk, S.M. Qaim. Activation cross section of the  $^{169}$ Tm(d,2n) reaction for production of the therapeutic radionuclide  $^{169}$ Yb, submitted to Applied Radiation and Isotopes, in press.

Keywords: Excitation Functions, Medically Relevant Isotopes, Deuteron Reactions, Pr and Tm Targets

# P015 PRODUCTION OF HIGH SPECIFIC ACTIVITY 177 Lu FOR TARGETED RADIONUCLIDE THERAPY USING MEDIUM FLUX RESEARCH REACTORS

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Introduction:  $^{177}$ Lu  $[T_{1/2}$ =6.73 d,  $E_{\beta max}$ =496 keV,  $E_{\gamma}$ =113 keV (6.4%), 208 keV (11%)] is presently being considered as an attractive radionuclide for targeted radiotherapy owing to its suitable decay characteristics. The expected increase in clinical use of  $^{177}$ Lu necessitates its large scale and cost-effective availability in high specific activity. Towards this, two routes, viz.direct  $^{176}$ Lu (enriched)  $(n,\gamma)^{177}$ Lu or indirect  $^{176}$ Yb $(n,\gamma,\beta^-)^{177}$ Lu could be considered. However, due to the high  $(n,\gamma)$  cross-section of  $^{176}$ Lu (2100 b), irradiation of enriched Lu target in medium flux reactors produces  $^{177}$ Lu with sufficiently high specific activity and adequate radionuclidic purity for most radiotherapeutic applications.

**Experimental:**  $^{177}$ Lu was produced by irradiating enriched (64.3%  $^{176}$ Lu) Lu<sub>2</sub>O<sub>3</sub> target at different flux positions (1.4x10 $^{13}$ -1.0x10 $^{14}$  n/cm<sup>2</sup>.s) and durations (7-21 d) in order to optimize the irradiation conditions for achieving maximum specific activity. The irradiated target was dissolved in dilute HCl. Assay of radioactivity and determination of radionuclidic purity of  $^{177}$ Lu was carried out by γ-spectrometry using an HPGe detector connected to a 4K MCA system.

Results and Discussion:  $^{177}$ Lu was obtained with a maximum specific activity of  $\sim$ 850 GBq/mg on 21 d irradiation at a neutron flux of  $1x10^{14}$  n/cm².s corresponding to  $\sim$ 21 atom% specific activity which is considerably higher compared to the theoretically calculated value. This could be possibly attributable to the contribution of epithermal neutrons. The radionuclidic purity of  $^{177}$ Lu was 99.98%,  $^{177m}$ Lu being the only radionuclidic impurity. In case of the production of  $^{177}$ Lu by (n,γ) activation of enriched target at high flux positions, the target burn up will be an important factor owing to the high σ value of  $^{176}$ Lu. Hence, a careful optimization of the time of irradiation is necessary to obtain the highest specific activity. It was observed that the specific activity of  $^{177}$ Lu was maximum after 21 d irradiation at  $1.0x10^{14}$  n/cm².s flux, beyond which, it decreased owing to the high target burn up. Although NCA  $^{177}$ Lu could be obtained via the indirect route, the yield of  $^{177}$ Lu will be significantly lower ( $\sim$ 1.1 GBq  $^{177}$ Lu/mg of 99% enriched lobation of NCA  $^{177}$ Lu from macroscopic level of Yb target.

**Conclusion:** A cost-effective large scale production of <sup>177</sup>Lu in adequate specific activity and high radionuclidic purity for use in targeted radiotherapy could be achieved in moderate flux rectors using enriched targets via the direct route thereby circumventing the tedious radiochemical separation involved in the indirect method.

**Acknowledgement:** The authors acknowledge IAEA for the enriched Lu target.

Keywords: 177Lu, High Specific Activity, Radionuclidic Purity, Targeted Radionuclide Therapy

# P016 A NEW MEASUREMENT OF THE EXCITATION FUNCTION OF THE <sup>75</sup>As(<sup>3</sup>He,2n)<sup>76</sup>Br REACTION

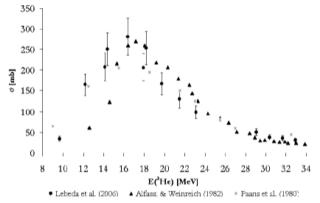
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**Introduction:** A positron emitter <sup>76</sup>Br (16.2 h) is promising radionuclide for PET imaging of processes with slower kinetics. It can be produced either by the <sup>76</sup>Se(p,n) reaction on enriched targets [1,2,3], or via the reaction <sup>75</sup>As(<sup>3</sup>He,2n) [4,5]. This work presents a new measurement of the excitation function of the latter reaction.

**Experimental:** The cross sections were determined by usual stacked-foil technique. Three stacks with 4-6 copper foils (12.3  $\mu$ m) covered by a thin As layer (0.45-2.16  $\mu$ m) were irradiated on the external beam of the cyclotron U-120M for 1.0-1.5 h at the beam intensity < 0.5  $\mu$ A. The beam energy was measured with use of a magnetic monochromator and its intensity was monitored by the <sup>nat</sup>Ti(<sup>3</sup>He,x)<sup>48</sup>V reaction [6]. The foils were analyzed by gamma spectrometry. The <sup>76</sup>Br activity was calculated from its 1 853.7 keV  $\gamma$ -line (14.7%) in order to avoid the contribution of <sup>75</sup>As (25.87 h) [5]. Precise thickness of the As layers was determined via neutron activation analysis.

**Results and Discussion:** The excitation function in the context of previously measured data [4,5] is displayed on Fig. 1. In lower energy region, it resembles more the work [4], but the maximum cross section value (280 mb) agrees rather with the data of [5]. An estimated total uncertainty of the cross sections was 12-16%. Calculated thick target yield as a function of the  ${}^{3}$ He energy for  $E_{out}$  = 10 MeV is displayed on Fig. 2. Its values (18.9 MBq/ $\mu$ Ah, resp. 440 MBq/ $\mu$ A at 25 MeV) makes the reaction suitable for production of  ${}^{76}$ Br in GBq amounts.



22 20 18 16  $[MBq/\mu Ah]$ 14 12 10 R 6 2 0 12 20 10 14 16 22 30 E (He) [MeV]

Fig. 1. Excitation function of the <sup>75</sup>As(<sup>3</sup>He,2n) <sup>76</sup>Br reaction.

Fig. 2 Physical yield of <sup>76</sup>Br in the thick As target.

**Conclusion:** The excitation function of the reaction  $^{75}$ As( $^{3}$ He,2n) was measured in the 10–33 MeV energy range. The results were compared with previously published data.

**Acknowledgement:** We are grateful to Mr. L. Cvrcek for preparing As layers and to Prof. J. Kucera for the NAA analysis.

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Keywords: Br-76, Excitation Function, He-3, Cyclotron, PET

# P017 EFFICIENT SEPARATION OF Y-86 AND RECOVERY OF STRONTIUM AFTER CYCLOTRON PROTON IRRADIATION OF STRONTIUM DISSOLVED IN AN AQUEOUS TARGET MATRIX

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**Introduction:** The production of metallic positron emitters is realized predominantly by applying solid target technology [1, 2] which can not be operated by an automated target processing. Those solid target systems, however, have several disadvantages. An alternative could be the irradiation of aqueous salts of target isotopes, allowing automated target operation. Here we report the separation of 86Y and the recovery of target-Sr after radionuclide production for potential automation using the new concept of dissolved target material in aqueous target matrices [3].

**Experimental:** After proton irradiation of the aqueous 2.8 M Sr(NO3)2 target solution, it was transferred into a hot cell. Additional fluid resulted from target rinsing. Ferric iron carrier was added and \*Y/Fe(OH)3 precipitation proceeded within 30 min at 80°C. The mixture was filtered, the precipitate was washed and re-dissolved in HCl. Carrier was removed by anion exchange SPE. The filtrate was evaporated to dryness and dissolved in dilute HCl for direct labelling purposes.

Similar to a reported procedure [4], recovery of target Sr was conducted by addition of an aqueous saturated (NH4)2CO3 solution to the clear filtrate obtained from \*Y/Fe(OH)3 precipitation. The precipitate was filtered and washed with dilute carbonate solution. Retained SrCO3 was re-dissolved by slowly reversed flow-through of a concentrated HNO3 solution. Nitric acid Sr(NO3)2 solution was evaporated to dryness and the residual salt was dissolved in water for further irradiations.

**Results and Discussion:** From the target solution obtained in the hot cell, more than 90% of radioyttrium were isolated in radiochemical (< 0.01% Sr) and chemical (< 0.01% Fe) pure form, ready for labelling procedures. Target Sr was recovered with overall yields of > 80%.

**Conclusion:** The described workup procedure is fast and efficient and has the potential to be conducted in a fully automated process. Thus, the new approach of operating an aqueous liquid target system allows an automated routine production of radioyttrium (e.g. 86Y, practical yields: 200 MBq calculated for 96% 86Sr enrichment, 1 h irradiation,  $6 \,\mu\text{A}\ 16 \,\text{MeV}$  protons) including the recovery of the expensive target nuclide.

Acknowledgement: We thank Prof. O. Marti and Prof. S.N. Reske for project support.

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Keywords: Y-86, Aqueous Target Matrix, Radionuclide Separation, Target Material Recovery

## P018 IN-TARGET PRODUCED (11C)METHANE INCREASES SPECIFIC RADIOACTIVITY

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**Introduction:** The most widely used way to produce the important 11C-labeling precursors [ $^{11}C$ ]methyl iodide ([ $^{11}C$ ]MeI) and hydrogen [ $^{11}C$ ]cyanide ([ $^{11}C$ ]HCN) is based on production of [ $^{11}C$ ]CO $_2$ . In the search for increased specific radioactivity (SRA) we investigated the influence of changing to target produced [ $^{11}C$ ]methane. The study involves the productions of five PET tracers, comparing SRA when produced from in-target [ $^{11}C$ ]CO $_2$  and [ $^{11}C$ ]CH $_4$ . Results from two and a half years with routine production of radioligands from [ $^{11}C$ ]CH $_4$  produced from target are included in this study.

**Experimental:** [ $^{11}$ C]MeI: [ $^{11}$ C]CO $_2$  was produced using 16.4 MeV protons in the 14N(p,a)11C reaction on N $_2$ (g) with 0.5% O $_2$ (g) and [ $^{11}$ C]MeI was synthesized utilizing a GEMS MeI Microlab® system. [ $^{11}$ C]CH $_4$  was produced in the same target type on N $_2$ (g) and 10% H $_2$ (g) [1]. [ $^{11}$ C]MeI was synthesized via a radical reaction with iodide, and was converted to [ $^{11}$ C]methyl triflate [2]. [ $^{11}$ C]HCN: In target produced [ $^{11}$ C]CO $_2$  and [ $^{11}$ C]CH $_4$  was prepared as above. [ $^{11}$ C]CO $_2$  was reduced to [ $^{11}$ C]CH $_4$  over a nickel catalyst. NH $_3$  gas was added to the produced isotopes and reaction with platinum and subsequently H $_2$ SO $_4$  generated [ $^{11}$ C]HCN.

**Results and Discussion:** By switching to the [\(^{11}\)C]CH<sub>4</sub> target we could show a 25- to 40-fold increase of SRA for [\(^{11}\)C]Raclopride and [\(^{11}\)C]MADAM. We see a lower SRA increase for \(^{11}\)C]SCH23390 and [\(^{11}\)C]PE2I, which is believed to be due to reference contamination in precursor material. For [\(^{11}\)C]HCN as radiolabelling precursor we were able to get a 12-fold increase of SRA.

Table 1. SRA at EOS for five different radioligands

Ligand	SRA [ <sup>11</sup> C]CO2 (Ci/mmol)	N	SRA [ <sup>11</sup> C]CH4 April 2004 - June 2006 (Ci/mmol)	N	SRA [ <sup>11</sup> C]CH4 August - December 2006 (Ci/mmol)	N
[ <sup>11</sup> C]Raclopride	1243	10	26255	69	44925	23
[ <sup>11</sup> C]MADAM	737	9	28644	25	_	_
[ <sup>11</sup> C]SCH23390	1519	16	10111	69	12367	36
[ <sup>11</sup> C]PE2I	1782	23	10699	8	_	_
[ <sup>11</sup> C]Benzonitrile	263	9	3274	15	-	_

**Conclusion:** By switching from carbon dioxide target to methane target we were able to significantly increase the SRA of [<sup>11</sup>C]methyl iodide and <sup>11</sup>C]HCN, and the SRA is still rising after several hundred irradiations.

**References:** [1] Bjork, H. et al. [2004] 10th Workshop on Targetry and Target Chemistry, Madison, Wisconsin, abstract A02, p.12. [2] Schou, M., Halldin, C., Sovago, J., Pike, V., Gulyas, B., Mozley, D., Johnson, D., Hall, H., Innis, R., Farde, L. [2003] Nucl. Med. Biol. 30:707-714.

Keywords: Specific Radioactivity, [11C]Methane, Hydrogen Cyanide, Methyl Iodide

#### P019 NEUTRON FLUX MEASUREMENT FROM WATER TARGETS DURING PROTON BEAM IRRADIATION

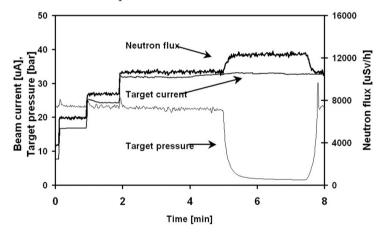
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**Introduction:** Diagnostics of radionuclide production targets are important when the yield and quality of the desired product is optimised and maximised. We describe selected parameters monitored during radionuclide production at the CC-18/9 cyclotron at Turku PET Centre and their use in target diagnostics.

**Experimental:** Signals, such as temperatures, target pressures, cyclotron vacuum levels, gamma and neutron flux and beam currents are routinely monitored and logged during radionuclide production target irradiation. Irradiation of water targets for production of either  $^{13}N$  or  $^{18}F$  is carried out with a high-pressure target system equipped with water recirculation and a heat exchanger for efficient removal of heat during high current irradiation. Measured water temperatures during irradiation are in excess of  $200^{\circ}C$  at entrance into the heat exchanger. Neutron fluxes were measured from both  $H_2^{16}O$  and  $H_2^{18}O$  targets during irradiation with up to  $50~\mu A$  beams of 18~MeV protons. The flux measurements were correlated to beam currents on targets and pressures.

**Results and Discussion:** During irradiation of  $\rm H_2^{16}O$ -targets for  $^{13}N$ -production a clear increase in neutron flux was observed when the target pressure was radically decreased, see fig 1. This pressure decrease was achieved by stopping the recirculation. We interpret the neutron flux increase as a result from void formation in the target with subsequent removal of water from the beam strike region. The beam then hits the target chamber material, which has different neutron production yields at these conditions. No such behaviour was observed during  $\rm H_2^{18}O$ -target irradiation for  $\rm ^{18}F$ -production.



**Conclusion:** Neutron flux measurement can be used to monitor production of  $^{13}$ N during irradiation of  $^{12}$ O. The information thus gained can also be applied to  $^{12}$ O targets.

Acknowledgement: Prof. Jerry Nickles is gratefully acknowledged for discussions.

Keywords: Radionuclide Production, Neutron Flux, Cyclotron, Water Targets, 13N

#### P020 UPDATING COMPUTER CONTROL FOR AN RDS-112 AND A CPCU

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**Introduction:** At the University of Wisconsin, we have been able to modernize our RDS-112 (#001) by building a new LabVIEW based control system which replaces the original iRMX control system. Additionally, we have responded to an increased need for 18F-fallypride on campus by reviving an CTI CPCU designed for 18F-FDG production with LabVIEW control to create a flexible system for 18F nucleophilic chemistry.

**Experimental:** Both devices ran under the control of a PC running software developed by CTI with the iRMX operating system, which is restricted to a 386-based processor. To ensure an extended life for the cyclotron and rebirth of the CPCU, the control of these devices was coupled with new National Instruments data aquistion hardware.

Using a combination of NI I/O hardware (2 x PCI-6033E, PCI-6703, and PCI-6509 on the cyclotron and 2 x USB-6501 on the CPCU) and NI's DAQ assisant we developed a link between the new PC and old hardware, replicating all previous functionality.

The new RDS software emulates the old system's data I/O structure and runs in what would formally be called "maintenance mode". Its new features include a graphical user interface (GUI), data logging with MS Word document output, neutron and gamma counters, a leaky integrator for estimating in-target activity, and web-based remote observation capabilities. For the CPCU, early synthesis similarities in nucleophilic 18F substitution chemistry allowed us to program in LabVIEW using much of the same timing structures from the original FDG program. New features include a GUI to permit complete synthesis or manual interruptions at each synthesis step, followed by a cleaning operation.

**Results and Discussion:** Since May 2005, the cyclotron system has been in full operation with no complications with system control. For the CPCU, we have 100% synthesis success rate out of more then 50 trials since May 2006. Decay corrected yields have consistently been on average 33% out of the box with average specific activities of 2.5 Ci/mmol.

**Conclusion:** The cyclotron and CPCU improvements have helped to keep the UW's CTI equipment in full operation. Using the software structure developed for the CPCU, the cyclotron interface could be automated to eliminate the need for maintenance mode. The CPCU program is easily adapted for other nucleophilic 18F substitution syntheses. With these new computer interfaces being more robust than the originals, we hope to considerably increase the lifetime of these devices that were built to last.

**References:** [1] Mukherjee, J., et al. Nucl Med Biol 22: 283-296 (1995). [2] Padgett, H.C., et al. Appl. Radiat. Isot., 40: 433-444 (1989).

Keywords: 18F-Fallypride, RDS-112, LabVIEW, CPCU, Automation

# P021 IRRADIATION OF BISMUTH TARGET FOR CYCLOTRON PRODUCTION OF $^{211}$ At/ $^{211g}$ Po AND $^{210}$ At $\rightarrow$ $^{210}$ Po

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**Introduction:** Astatine-211 ( $t_{1/2} = 7.214$  h) is an  $\alpha$  emitter at secular equilibrium by EC with its ultra-short-lived  $\alpha$  emitting daughter polonium-211g ( $t_{1/2} = 516$  ms), with a high Linear Energy Transfer (LET) of 130 eV nm<sup>-1</sup> and a half-life suitable for high-LET metabolic radioisotope therapy onto humans (HL-RIT). The overall α branching of the  $^{211}$ At/ $^{211g}$ Po system is  $^{100\%}$  and it has a CF specific activity of 76 GBq· $\mu$ g<sup>-1</sup> for  $^{211}$ At and  $^{3.8}$ · $^{106}$  GBq· $\mu$ g<sup>-1</sup> for  $^{211g}$ Po. A good choice for an <sup>211</sup>At production is minimizing radioisotopic contamination and the direct method based on the nuclear reaction  $^{209}$ Bi $(\alpha,2n)^{211}$ At, using  $\alpha$  energies greater than the reaction threshold of 20.72 MeV seems the most practical, because it can be carried out in a medium energy cyclotron. The most relevant impurities co-produced are <sup>210</sup>At and <sup>209</sup>At because their half-lives and physical reaction thresholds are similar to <sup>211</sup>At one but, selecting an opportune energetic window, their production can be avoided; however a small amount of 210At - produced at energies above 28.61 MeV - is acceptable because it can be used like y-emitting internal spike, for radiochemical processing and labelling.

Experimental: To check the reliability of the procedure and to measure the activity and the thick-target yield (TTY), irradiations of  $^{209}$ Bi targets at different  $\alpha$  energies have been carried out at the Scanditronix MC40 cyclotron of JRC-Ispra of the EC. The target thickness was chosen large enough to assure the absorption of all α particles with energy above 20 MeV. Studying the γ-X spectra collected at the end of each irradiation were identified the radionuclides produced and was calculated their activity; knowing the activity and the integrated charge was computed also the TTY, under different irradiation conditions.

Results and Discussion: The activity measured has confirmed that the irradiation conditions can be set up in order to obtain high activity of 211At and, at the same time, a wished amount of the 210At spike (from 0.001 to 7%). Under the irradiation conditions adopted also the long-lived impurity  $^{210}$ Po ( $t_{1/2} = 138.4$  d) is produced from  $^{210}$ At decay; however its activity is negligibly small compared to  $^{211}$ At/ $^{211}$ gPo one, due to the favourable ratio of correspondent half-lives. Gamma spectra acquired some months after the irradiations have shown the presence of the only long-lived  $^{207}$ Bi impurity and a more detailed evaluation of the  $^{207}$ Bi activity produced was investigated too.

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# P022 APPLICATION OF EXTRACTION CHROMATOGRAPHY TO THE PREPARATION OF RADIONUCLIDES FOR MEDICAL USE

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**Introduction:** Extraction chromatographic resins, especially resins comprised of extractant coated silica, increasingly find application in the preparation of radionuclides for medical use. Examples include: Dietz et al. [1] showed that a generator comprised of Sr and RE Resins, allows obtaining Y-90 with high yield ( $\sim$ 95%) in very high radiochemical purity (Sr decontamination factor  $>10^9$ ). Some patents [2] describe the use of extraction chromatography for the separation of Ac-225 from U-233/Th-229, using DGA Resin for Ac fixation, resulting in Ac-225 solutions with high radiochemical and chemical purity. Horwitz et al. [3] describe a method for the separation of Lu-177 from irradiated Yb-176 targets (up to 300 mg), using LN2 Resin. Yb decontamination factors of  $10^6$  for a Lu-177 recovery of 73% at an activity level of 12-15 Ci Lu-177 were reported.

**Preparation of In-111 solutions using extraction chromatography:** Extraction chromatographic materials were applied to the preparation of In-111 solutions from irradiated targets. Weight distribution ratios  $D_w$  were determined for Cd and In on two extraction chromatographic resins, Ac Resin [4] and LN Resin [5]. On both resins a difference in  $D_w$  values of two orders of magnitude between In and Cd was found, at HCl concentrations of 1M for the Ac Resin, 0.1M HCl for the LN Resin respectively, thus allowing an easy In/Cd separation. Indium could be eluted in both cases near quantitatively with 10 mL 6M HCl. For both resins Cd decontamination factors in the order of  $10^4$  for one separation cycle were found, as well as good separation from chemical impurities like Al, Fe and Cu. Further it was shown that TEVA Resin [6] has high selectivity for Cd over In at 2M HCl. This fact might be used in two applications. A TEVA disc can be used for further in-hospital purification of In-111 solutions. It was shown that Cd was fixed on the disc, even at flowrates >20 mL.min<sup>-1</sup>. Cd decontamination factors in the order of  $10^4$  were obtained at near quantitative indium recovery. The same selectivity also allows Cd target material recovery. The TEVA Resin has a Cd capacity in the order of 5 mg.mL<sup>-1</sup> resin. The amount of resin has thus to be chosen according to the amount of Cd used as target material. Cadmium can then, after indium removal, be recovered near quantitatively (93  $\H$  100%) using 0, 1M EDTA in 0, 1M HCl as eluting agent.

**References:** [1] M.L. Dietz, E.P. Horwitz, *Appl. Rad. Isot.*, <u>43</u>, 1992, 1093 f. [2] Bond et al., US Patent US7,087,206 B2, 08.08.2006. [3] E.P. Horwitz et al., *Appl. Rad. Isot.*, <u>63</u>, 2005, 23-36. [4] Horwitz, E.P.; et al., *React. Funct. Polym.*, **33** (1997) 25-36 (1997). [5] Horwitz E.P. et al., **37** (1975) 425-434. [6] Horwitz, E.P.; et al., *Analyt. Chim. Acta*, **310** (1995) 63-78