NEW PRODUCTION ROUTES FOR SOME THERAPEUTIC RADIONUCLIDES

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In modern internal radionuclide therapy, β^-- emitting radionuclides are often used. For this purpose the relatively high-energy β-particle emitting radionuclides ^{32}P ($T_{1/2}$ = 14.26 d), ^{89}Sr ($T_{1/2}$ = 50.53 d), and ^{192}Ir ($T_{1/2}$ = 73.83 d) are commonly employed and the Auger electron emitter ^{169}Yb ($T_{1/2}$ = 32.03 d) is of great potential interest. The conventional production routes of these radionuclides involve mainly the (n,γ)-reaction at nuclear reactors [1], leading to high product yields but low specific activities. In the case of ^{32}P and ^{89}Sr the (n,p)-reaction with fission neutrons has also been used. The achieved specific activity is then high, but the yield is quite low. In this work we report on new cross section measurements for the production of ^{32}P and ^{89}Sr in a fast neutron field. Furthermore, data measurements relevant to the production of ^{169}Yb and ^{192}Ir via charged particle induced reactions are also described.

Fast neutrons were produced via breakup of 14 MeV deuterons on a thick Be-target at the compact cyclotron CV 28 in Jülich [2]. Proton bombardments over the energy range of 5 to 18.5 MeV were also carried out at the CV 28 and at the MGC-20E cyclotron in Debrecen. Irradiations with higher proton energies (25 to 45 MeV) were done at the injector of COSY in Jülich. For measurement of the neutron spectrum-averaged cross section of the $^{32}S(n,p)^{32}P$ process, thin sulphur layers were irradiated and the activities determined via β^- counting using a gas flow proportional counter. Similarly, for the investigation of the $^{89}Y(n,p)^{89}Sr$ reaction, after neutron irradiation of Y_2O_3, a clean radiochemical separation was performed and the product analysed via β^- counting as well. Measurements on the $^{169}Tm(p,n)^{169}Yb$ and $^{192}Os(p,n)^{192}Ir$ reactions were performed using the stacked-foil technique. Thin samples of $^{169}Tm_2O_3$ were prepared using a sedimentation technique and those of enriched elemental ^{192}Os via electrolytic deposition. The measured neutron cross section data were compared with values calculated from the known (n,p) excitation functions and the d(Be)-neutron spectral distribution. This served as an integral test of the differential data. The (n,p) cross sections with the d(Be)-neutrons were found to be about four times higher than those with fission neutrons. No-carrier-added ^{32}P and ^{89}Sr can thus be produced more efficiently via the (n,p)-reaction using d(Be)-breakup neutrons than with fission neutrons.

Regarding the production of ^{169}Yb and ^{192}Ir via charged particle induced reactions,integral yields were calculated from the measured excitation function of the $^{169}Tm(p,n)^{169}Yb$ reaction up to 45 MeV and of the $^{192}Os(p,n)^{192}Ir$ reaction up to 20 MeV. The production possibilities of the two radionuclides at a cyclotron and at a nuclear reactor were evaluated, assuming at both optimum production conditions. The reactor route gives much higher yields. However, the advantage of the cyclotron route would be that a "no-carrier-added" product is obtained.

[1] Manual for reactor produced isotopes. IAEA-TECDOC-1340, 116-117/238-243, International Atomic Energy Agency, Vienna (2003).
[2] Spahn I., Coenen H. H., Qaim S. M., Radiochimica Acta 92, 183-186 (2004).

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EVALUATION OF POLYOXOMETALATES TO EXTRACT LANTHANIDES

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Objective: To determine if polyoxometalates (POMs) can be used to extract lanthanides from aqueous solutions into an organic solution using alkylamines, for ultimate use in indirect production of carrier free lanthanides for radiotherapy. **Introduction:** There are currently two methods available to produce Lu-177; one is direct neutron activation of enriched Lu-176. Lutetium-177 can be produced from Lu-176 but only ~28% of the Lu-176 atoms are converted to Lu-177, which leaves mostly nonradioactive Lu-176 – yielding a specific activity of 20-30 mCi/mg. Another disadvantage of the direct approach is the production of the long-lived impurity Lu-177m ($t_{½}$= 160 days). The second method to produce Lu-177 and other lanthanides (including Pm-149 and Ho-166) is neutron capture followed by beta decay to the desired radioisotope. For example, neutron activation of enriched Yb-176 produces Yb-177 (half-life = 1.9 hr), which is followed by beta decay, producing Lu-177. When the indirect method of production is used, separation of the daughter material from the parent is possible, with the advantage that nearly all of the lutetium atoms produced are radioactive. An additional advantage is that indirect production produces far less of the long-lived Lu-177m impurity. A higher specific activity increases the effective dose delivered to the tumor. A number of chromatographic separations have been investigated at MURR to provide high specific activity separations of lanthanides. Most have not proven viable for production of radioisotopes for medical applications. The reasons for this include poor separation of the milligram amounts of target material from the picogram or less quantities of the desired radioisotope; the introduction of contaminants that result in the radioisotope being unusable; and/or because they did not allow for recovery of the expensive target material. The ideal separation needs to be rapid, efficient at separating the desired radioisotope from the target material, amenable to recovery and reuse of the target material, and adaptable to scale-up and remote handling in a hot cell. In this study, four different POM ligands designated a1, a2, w1, and w2 were evaluated to determine if useful in separating adjacent lanthanides. **Methods:** Radiolanthanides were produced by MURR and POM ligands were synthesized at Hunter College. Solvent extraction of radiolanthanides from aqueous solution into chlorofrom was evaluated using different alkylamines and POMs as complexants. Initially, aqueous solutions of the POMs and radiolanthanides with ratios of 0.5-1.0 of POM: radiolanthanide were prepared. Formation of the radiocomplex was determined by radio-TLC. After formation of the POM was confirmed, portions of the aqueous solution was then added to chloroform containing different amines. The aqueous and organic phases were vortexed, separated and counted by HPGe counting. **Results:** All of the POM ligands tested formed complexes with radiolanthanides at purities of 100% at concentrations of 1:1 and higher. Only the octylamine was shown to result in extraction of the radiolanthanide:POM complex into chloroform. Although all the POM ligands resulted in some extraction of the radiolanthanides into chloroform, the highest extraction into chloroform was observed for a1. The percent extracted appeared to increase as the lanthanide series was traversed. **Conclusions:** Radiolanthanides were selectively extracted by POM ligands into chloroform using octylamine. This suggests these ligands should be further evaluated to determine if they could be used to separate adjacent radiolanthanides as required for carrier free production.

Keywords: Polyoxometallates, Radiolanthanides, Separations

AN INCREASED SPECIFIC RADIOACTIVITY OF [^{11}C]HCN WAS OBTAINED USING IN-TARGET PRODUCED [^{11}C]METHANE

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Introduction: Hydrogen [^{11}C]Cyanide ([^{11}C]HCN) has long been used as an important ^{11}C-labelling precursor. Cyanide is used to form carbon-carbon bonds, and introduces nitriles that subsequently can be transformed into a number of functionalities such as amides, amines, aldehydes, esters and ketones. The most widely used way to produce [^{11}C]HCN is based on production of [^{11}C]CO2 in a nitrogen target. The [^{11}C]HCN is then produced in high yields in a two step reaction via [^{11}C]methane [1].

In ^{11}C-methylations it has been shown that it is possible to obtain extra high specific radioactivity (SRA) when using methane target [2, 3]. To investigate whether we could increase SRA for ^{11}C-cyanation reactions, we compared the preparations of a [^{11}C]benzonitrile from an aryl bromide precursor with either [^{11}C]HCN from target produced [^{11}C]methane or [^{11}C]carbon dioxide.

Method: [11C]CO2 was produced using 16.4 MeV protons in the 14N(p,a)^{11}C reaction on N2(g) with 0.5% O2(g) and reduced to [11C]CH4 over a nickel catalyst. [11C]CH4 was produced in the same target type on N2(g) and 10% H2(g). NH3 gas was added to the produced isotope and the mixture was passed through a quartz tube containing heated platinum (990°C). The produced [^{11}C]ammonium cyanide was bubbled through H2SO4 (50%, 2mL) at 65°C to generate [^{11}C]HCN and then trapped in KOH (0.5mg in 0.4mL DMSO) to get [11C]KCN. The [11C]KCN was transferred to a vial containing the aryl bromide precursor and palladium catalyst where the labelling (135°C, 5 minutes) took place [3]. Semi preparative reversed phase HPLC was performed to purify the product. The collected fraction from HPLC was evaporated and the residue redissolved into 8 mL physiological phosphate buffer (pH = 7.4). Radiochemical purity and SRA was analysed with reversed phase HPLC.

Results: For [^{11}C]HCN produced from the [^{11}C]CO2-target we were able to obtain a final product of (mean ± S.D., n=9) 417 ± 172 MBq with a synthesis time of 45 ± 4 minutes. The average SRA was 263 ± 174 Ci/mmol (calculated at 45min. after EOB). For [^{11}C]HCN produced from the [^{11}C]CH4-target we were able to produce a final product with a radioactivity of (mean ± S.D., n=3) 946 ± 472 MBq and a synthesis time of 44 ± 5 minutes. The average SRA for the three batches was 2701 ± 927 Ci/mmol (calculated at 45min. after EOB). The irradiation time was held constant at 15 minutes for all batches. The main cause of SRA increase must be avoidance of isotopic dilution from CO2 absorbed on inner surface of tubing, valves and other parts.

Conclusion: Switching from carbon dioxide target to methane target we were able to increase the SRA of [11C]HCN ten-fold without compromising total radioactivity or prolonging synthesis time.

1. Iwata, R., Ido, T., Takahashi, T., Nakanishi, H., Iida, S. Appl. Radiat. Isot. 1987; 38: 97-102
2. Buckley, K. R., Huser, J., Jivan, S., Chun, K. S., Ruth, T. J. Radiochim. Acta 2000; 88:201-205
3. Noguchi, J., Suzuki, K. Nucl. Med. Biol. 2003; 30: 335-343
4. Andersson Y, Långström B. J. Chem. Soc., Perkin Trans. 1, 1994: 1395-1400.

Keywords: Specific Radioactivity, Carbon-11, Hydrogen Cyanide

SELF-SUFFICIENCY AND OPPORTUNITY COST: NAVIGATING TOWARD EXPANDED DISTRIBUTION OF PET FEEDSTOCKS

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Introduction:

Opportunity cost is the penalty for waiting, playing it safe. Today, PET researchers track authentic ligands in small animals at 1 mm resolution. To lose a priceless preparation to logistical error upstream places a high value on a reliable, thus local, supply of the tracer feedstock. PET research at Wisconsin, nurtured on parsimony, employs two accelerators and four PET scanners spread over 5 km. Molded by these constraints, the ratio of the activity A needed at the user site to the initial activity at the point of production, $A/A_o = \exp(-R/v\tau)$, involves the distance R, velocity v and mean life τ. Obviously this is site-specific, not a binary relationship. The tracers listed below are transported (DOT 7A) across and beyond campus, ordered by increasing range, with only ^{14}O limited to in-house use.

Materials and Methods:]

Conventional PET tracers. The 6-minute delivery of $H_2{}^{15}O$ to the distant PET site softens with $^{13}NH_3$. Separate $^{11}CH_3I$ systems were developed at both the cyclotron and μPET site, calling for the transport of half-Curie activities of cryo-trapped $^{11}CH_4$ with a specific activity of ~ 3-4 Ci/μmole measured by capillary GC and a Valco pulsed discharge detector. After twenty years of in-house production, FDG is now purchased from commercial sources.

Unconventional PET tracers. The growing need for longer lived PET tracers now centers on ^{64}Cu and ^{124}I to label cancer specific agents. Earlier work surveyed over 100 thick target yields at 11 MeV; this work addresses the scale up to provide reliable access for a widening circle of users. A 133 mg/cm^2 layer of 96% ^{64}Ni (11 -> 6.8 MeV) is electroplated (<50 mA DC; ~36 hours) onto a gold disc. Irradiation at 30 μA reaches 1100 mCi in 7-8 hours. The 7 mm diameter nickel spot is dissolved in HCl and Ni-Co-Cu separation is performed on AG-1 X8 with 90% recovery of the ^{64}Ni stock for re-plating. High ^{64}Cu specific activity, requiring ultrapure reagents, is needed for mAb's and the nearly insoluble ATSM.

Two Te substrates have been studied for ^{124}I, fused TeO_2/6%Al_2O_3 and Al_2Te_3, both stable at ~ 20 μA. Scouting over irradiation variables: target support, cooling, beam angle leads to He target cooling (0°C, 3 l/sec, 30% heat removal), over a "just-thick" $^{124}TeO_2$ glass target fused onto a Pt support angled at 20-degrees and water cooled through an indium conduction pad. During distillation at 750°C, about 70% of the I-activity is trapped from the air stream downstream on quartz at -77°C, with the remainder left in the target. The ^{124}I activity is stripped with appropriate buffer and dried as a massless film on the inside of a V-vial for transport to the user. Losses of ^{124}Te are less than 1% per cycle for weekly production runs.

Results and Conclusions:

The UW RDS 112, a legacy cyclotron, makes Curie levels of ^{64}Cu. An installed base of several hundred small cyclotrons currently serves the PET community, mostly sleeping when not making ^{18}FDG. Enlisting even a few of these would provide a pool of feedstock sufficient for widespread application. Iodine-124 poses greater targetry challenges but makes an even clearer case for a cooperative consortium, a clearing-house to buffer global needs and reward those centers willing to shoulder the initial costs rather than miss the transient opportunity.

Tracer	half-life	A (EOSB) [mCi/μA]	Use	Challenge
$H_2{}^{14}O$	71 sec	2	rCBF	unveiling from ^{11}C
$H_2{}^{15}O$	2 min	70	rCBF	$^{15}N_2$ losses or $^{14}N(d,n)$
$^{13}NH_3$	10 min	6	rCBF	cross town logistics
$^{11}CH_4$	20 min	60	$^{11}CH_3I$	specific activity
^{64}Cu	12.7 hr	170	ATSM	^{64}NI recovery
^{124}I	4.2 d	37	many	volatile ^{124}Te and I

Keywords: Iodine-124, Copper-64, Local Production

MEASUREMENT OF CROSS SECTIONS FOR NATW(p,xn)$^{181\text{-}186}$Re REACTIONS UP TO 19 MEV

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186Re has been identified as a suitable radionuclide for therapy [1]. This is due to it's decay characteristics (t1/2 = 3.72 days, 92 % β emission). Currently 186Re is only available in low specific activity from neutron capture on rhenium targets in reactors. An alternative mode of production is the 186W(p,n) reaction. However, the literature values for these cross sections are not in agreement [2,3]. Cross sections for the production of 186Re along with 181Re, 182mRe, 182gRe, 183Re 184gRe from protons incident on natural tungsten have been measured using the stacked foil technique for proton energies up to 19MeV. Results are compared with the literature values. Our preliminary results indicate that up to 100 mCi of 186Re could be produced with a 15 hour irradiation of 100 μA of 17 MeV protons on an enriched 186W target.

[1] Postema EJ, Borjesson PK, Buijs WC, Roos JC, Marres HA, Boerman OC, de Bree R, Lang M, Munzert G, van Dongen GA, Oyen WJ, Dosimetric analysis of radioimmunotherapy with 186Re-labeled bivatuzumab in patients with head and neck cancer. J Nucl Med. 44 (2003):1690-9.

[2]Zhang X, Li W, Fang K, He W, Sheng R, Ying D, Hu W, Excitation Functions for natW(p,xn)181-186Re Reactions and Production of No-Carrier-Added 186Re via 186(p,n)186Re Reaction. Radiochimica Acta 86 (1999): 11-16

[3]Shitgeta N, Matsuoka H, Osa A, Koizumi, Izumo M, Kobayaski K, Hashimoto K, Sekine T, Lambrecht RM, Production Method of No-Carrier-Added 186Re. Radioanal. Nucl. Chem. 205 (1996): 85-92

Keywords: Rhenium-186, Cross-Section, Tungsten

ROLE OF X-RAY SPECTROMETRY IN MEDICAL RADIONUCLIDE DEVELOPMENT

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The present efforts relevant to radionuclide development for medical applications are oriented generally in two directions, therapeutic radionuclides and longer-lived positron emitters. Some of the therapeutic radionuclides decay almost exclusively via electron capture (EC) and can therefore be characterized only by X-ray spectrometry. In the case of longer-lived positron emitters, occasionally information on exact positron branching is missing. To determine this, resort is also made to X-ray spectrometry.

This spectrometric technique is rather subtle and demands preparation of thin samples, so that the self-absorption effect is reduced to a minimum. A further difficulty lies in the determination of the efficiency of the low-energy Si(Li) detector. The accuracy of measurement is also affected by the uncertainty in the knowledge of the fluorescence yield, and by the presence of isotopic impurities. Since all the neutron deficient radioisotopes of an element decaying by EC emit the same X-rays, it is absolutely necessary that the radionuclide under investigation is highly pure. We describe below some studies using X-ray spectrometry, both for cross section measurements and determination of positron branching.

The excitation functions of the ^{103}Rh(p,n)^{103}Pd, ^{102}Ru(^{3}He,2n)^{103}Pd and ^{100}Ru($_{\alpha}$,n)^{103}Pd processes were determined using K_{α} X-rays of energy 20.21 keV. The radionuclide ^{103}Pd ($T_{1/2}$ = 17.0 d) is widely used in brachytherapy, but the database for its production has been hitherto rather weak. We also measured cross sections of the ^{85}Rb(p,4n)^{82}Sr reaction via X-ray spectrometry using the K_{α} X-rays of energy 13.39 keV. The data for this reaction had been obtained so far only via $_{\gamma}$-ray spectrometric assay of the 1.2 min daughter ^{82}Rb activity. Our X-ray spectrometric studies provide the first set of experimental data for the direct production of ^{82}Sr. This radionuclide is widely used in the preparation of ^{82}Sr/^{82}Rb generators for PET studies.

In connection with the determination of positron branching, we applied the X-ray spectrometric technique to ^{120}I ($T_{1/2}$ = 1.3 h) and ^{124}I ($T_{1/2}$] = 4.18 d), two very promising radionuclides, whose positron emission intensities, however, were rather uncertain. The two radionuclides were obtained in very pure form via the nuclear reactions ^{120}Te(p,n)^{120}I and ^{124}Te(p,n)^{124}I in combination with a radiochemical separation. From the ratio of the intensity of the 511 keV annihilation radiation to that of the K_{α} X-ray of 27.47 keV, the positron branching in each radionuclide was determined. It was found to be 56 ± 3 % in the case of ^{120}I and 22 ± 1 % in ^{124}I.

The above examples demonstrate the significance of X-ray spectrometry. It should, however, be mentioned that the X-ray spectrometry is used only in special cases, the $_{\gamma}$-ray spectrometry being the most extensively applied technique.

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Keywords: X-Ray Spectrometry, EC-Decay, Positron Branching

NEW NUCLEAR DATA FOR PRODUCTION OF ^{124}I *VIA* ^{3}He AND α-PARTICLE INDUCED REACTIONS ON ^{nat}Sb AND ^{121}Sb

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The radionuclide ^{124}I ($T_{1/2}$ = 4.18 d, EC = 78 %, Iβ$^+$ = 22 %) is both a diagnostic and a therapeutic radionuclide. It is generally produced using the low-energy reactions ^{124}Te(p,n)^{124}I or ^{124}Te(d,2n)^{124}I. The intermediate energy reactions ^{125}Te(p,2n)^{124}I and ^{126}Te(p,3n)^{124}I have also been investigated. The latter two reactions give high yields but the levels of radioactive impurities are also higher. The best method to date appears to be the ^{124}Te(p,n)^{124}I process, although the yield is rather low.

In view of the increasing significance of ^{124}I, we investigated the ^{3}He- and $_{\alpha}$-particle induced nuclear reactions on ^{nat}Sb, and the α-particle induced reactions on highly enriched ^{121}Sb. Some data existed in the literature but, since they were obtained in the context of ^{123}I production, the data base for ^{124}I production was rather weak. We prepared thin samples of ^{nat}Sb and ^{121}Sb (99.45 % enriched) by a sedimentation process, and irradiations were done in a stacked-foil arrangement at the compact cyclotron CV 28 in J$_{ü}$lich. The beam currents were measured using the monitor reactions ^{nat}Ti(^{3}He,x)^{48}V and ^{nat}Ti($_{\alpha}$,xn)^{51}Cr. The radioactivity of the products ^{121}I, ^{123}I, ^{124}I, ^{125}I and ^{126}I was measured using high-resolution $_{\gamma}$-ray spectrometry. The cross sections were calculated using the usual activation equation.

The excitation functions of the ^{nat}Sb(α,xn)$^{123,\ 124,\ 125,\ 126}$I reactions, measured over the energy range of 8 to 27 MeV, showed that ^{124}I can be produced with a thick target yield of about 1.2 MBq/μA,h, the levels of ^{126}I and ^{125}I impurities being about 10 %. Similar measurements on enriched ^{121}Sb led to a thick target yield of ^{124}I of about 2.2 MBq/μA,h with no longer-lived impurities. The level of ^{123}I is rather high (about 100 %), but it could be reduced in two ways: either by limiting the $_{\alpha}$-particle energy to about 17 MeV (with consequent decrease of yield) or by allowing a decay time of about 4 days prior to the application of ^{124}I.

The excitation functions of the ^{nat}Sb(^{3}He,xn)$^{121,\ 123,\ 124}$I reactions, measured over the energy range of 8 to 36 MeV, gave the calculated thick target yield of ^{124}I of about 1.0 MBq/μA,h. An ^{125}I or ^{126}I impurity was not detected. The levels of ^{121}I and ^{123}I impurities can be decreased by limiting the ^{3}He-particle energy to about 30 MeV or by allowing a decay time as mentioned above. A comparison of the presently investigated routes of ^{124}I production with the known methods mentioned above will be given. In general, the yields of ^{124}I in ^{3}He- and α-particle induced reactions on antimony are much smaller than those in the p- and d-induced reactions on isotopes of tellurium.

Keywords: Iodine-124 Production, alpha Particle Induced Reactions, He-3 Particle Induced Reactions

SEPARATION OF ^{143}Pr AND ^{143}Ce USING HEXAGONAL TUNGSTEN BRONZE INORGANIC SORBENTS

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Pr-143 has potential for therapeutic applications in nuclear medicine. It has a beta emission ($E_{b\text{-}avg}$ = 315 keV %) with a half-life of 13.57 days. It can be generated from the neutron bombardment of enriched ^{142}Ce (n,g) ^{143}Ce - ^{143}Pr + b^- or as a by-product of the U-235 fission process. As the hydrated tungsten trioxide (hexagonal tungsten bronze form) have shown potential as ion-exchangers and the treatment of radioactive waste,[1-3] we were interested to evaluating their potential for the separation of ^{143}Ce and ^{143}Pr. The present study investigates the binding behaviour of a molybdenum doped hexagonal tungsten bronze (Mo_{10}-HTB) with Ce^{3+}, Ce^{4+} and Pr^{3+}.

The Mo_{10}-HTB was synthesized using hydrothermal methods. Ce-141 [$t_{1/2}$ = 30.5 days; g = 145 keV, 48.2 %] and ^{142}Pr [$t_{1/2}$ = 19.12 hr; g = 1576.6 keV, 3.68 %] were produced via neutron irradiation in the High Flux Australian Reactor (HIFAR). The distribution coefficients (K_d [mL/g]) or binding affinity of the sorbents for each radiometal ion was determined under various acid conditions: (0.1 – 8.0 M HCl, H_2SO_4, 0.01 – 2.0 M $HClO_4$; H_2SO_4 0.1 – 8.0 M and 0.1M $(NH_4)_2S_2O_8$; 0.01- 2.0 M $(NH_4)_2S_2O_8$). Selected data of the binding affinity of Mo_{10}-HTB for the various radiometal ions in the presence of varying acid solutions is presented in Figure 1. The data show optimum binding for Ce^{3+} and Pr^{3+} is at 0.5 M for $HClO_4$ and 1.0 M for both HCl and H_2SO_4. The $K_{d\ Ce}{}^{3+}$ / $K_{d\ Pr}{}^{3+}$ in 1.0 M H_2SO_4 and 0.5 M $HClO_4$ are ~2.4 and ~12, respectively. In all conditions, the binding affinity for each radiometal ion decreases as the acid concentration increases. Furtherwork involves optimising conditions for *in situ* separation of ^{143}Ce and ^{143}Pr.

1. B. Gerand G. Nowogrocki, J. Guenot and M. Figlarz, J. Solid State Chem. (1979) *29*, 427.
2. K. P. Reis, A. Ramanan and M. S. Whittingham, J. Solid State Chem. (1992) *96,* 31.
3. C. S. Griffith and V. Luca, Chem. Mater. (2004) *16,* 4992.

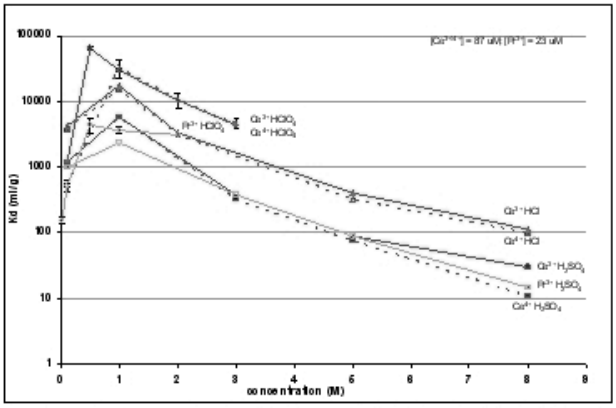

Figure 1. Comparison of $Ce^{3+/4+}$ and Pr^{3+} binding with Mo_{10}-HTB

Keywords: Cerium-143, Praseodymium-143 Separation, Inorganic Sorbents

MINIMIZING ISOTOPICALLY ENRICHED [N-15]NITROGEN GAS USAGE IN THE PRODUCTION OF O-15 LABELLED WATER

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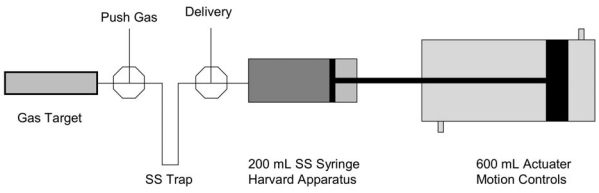
[O-15]Water has been widely used for measurements of cerebral blood flow, cerebral oxygen utilization, and extravascular water. (1) The production of O-15 on low energy proton only medical cyclotrons is limited to the $^{15}N(p,n)^{15}O$ nuclear reaction which employs relatively expensive isotopically enriched nitrogen-15 as the target material. Also intrinsic to many of these studies is the need for multiple scans spaced over a considerable time period. This requires continuous target irradiation and "burping" activity from the target to minimize relatively expensive $^{15}N_2$ usage at the cost of tying up valuable cyclotron time.

During our development of [O-15]CO_2 for radioactive ion beam experiments (2, 3), which requires sending batches of radioactive gas over 350 meters to the ion source of the Nuclear Science Division's 88" cyclotron every 120 seconds, we conceived of a system which allows for target loading, irradiation, separation and recovery of the N-15 containing gas and reloading of the target in an automated manner with virtually no loss of enriched $^{15}N_2$.(4)

The system consists of a 600 mL pneumatically controlled driving cylinder (Motion Controls) coupled to a 200 mL high pressure stainless steel syringe (Harvard Apparatus). This is used to load and unload the target through a small 1/16 in stainless steel tubing trap. The syringe is loaded with 5% Hydrogen/Nitrogen-15 and then pushed into the small volume gas target of our CTI RDS-111 cyclotron to a final pressure of 280 psi. The target is valved off and irradiated. Next the target is unloaded by pushing back the pneumatic cylinder passing the gas through the stainless steel cold trap (-40 C) catching the [O-15]H_2O and allowing the N-15 enriched target gas to return to the syringe. The [O-15]H_2O is released by resistively heating the trap and delivered to a vial with helium push gas. The process is repeated by actuating the pneumatic cylinder thus recharging the target. The entire process of loading, unloading and delivering the [O-15]H_2O is controlled by an in-house developed LabVIEW program.

Application of this device to production of medically useful quantities of oxygen-15 labelled water will be presented.



References:

(1) Frackowiack, R.S.J., Lammertsma, A.A. in Reivich, M. and Alavi, A., eds., Positron Emission Tomography. New York, Alan R. Riss Inc. 1985, pp 153-181.

(2) Powell, J. et al. "BEARS: Radioactive Ion Beams at Berkeley" Nuclear Instruments and Methods in Physics Research A 455 (2000) 452-459.

(3) Powell, J.; O'Neil, J.P.; Cerny, J. "Production of an Accelerated Oxygen-14 Beam" Nuclear Instruments and Methods in Physics Research B 204 (2003) 440-443.

(4) O'Neil, J.P "Oxygen-15 Preparation: Radioactive Ion Beam Considerations" Conference on the Application of Accelerators in Research and Industry. (2004)

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Keywords: [O-15]water, Isotope Production, Isotope Recovery

AN IMPROVED QUALITY CONTROL PROCEDURE FOR [^{18}F]FDG BY HPLC WITH UV DETECTION

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Introduction: Quality control of 2-deoxy-2-[^{18}F]fluoro-D-glucose ([^{18}F]FDG), the most commonly used radiopharmaceutical for positron emission tomography, has received attention due to its increasingly widespread use in clinical studies. In USP, the permissible chemical impurity level is defined for 2-deoxy-2-chloro-D-glucose (ClDG) formed during the synthesis of [^{18}F]FDG. The EP has also designated a test for amounts of inactive (cold) FDG and ClDG. Although high-performance anion exchange chromatography with a pulsed amperometric detector (HPAEC/PAD) has been listed in USP and EP as the recommended method for the determination of these substances, this method is not widely accepted in PET facilities due to the need for a comparatively expensive detector and frequent maintenance of the electrode surface to retain sensitivity. In addition, this method could not be utilized to determine the radiochemical purity of [^{18}F]FDG. Under HPLC conditions (strong alkaline solution), non or partially hydrolyzed 2-[^{18}F]fluoro-1,3,4,6-tetra-*O*-acetyl-D-glucose ([^{18}F]TAG) is further hydrolyzed to [^{18}F]FDG during the analysis, thus requiring an additional TLC-test for this impurity. In an attempt to resolve the problems with current methods in the present study, we established a new detection technique for the determination of cold FDG, ClDG and radiochemical purity in [^{18}F]FDG solutions.

Methods: This method was based on a normal phase chromatography coupled with a post-column photometric derivatization with 2-cyanoacetamide (2-CA). FDG and ClDG were separated on an aminopropyl-modified silica column, Inertsil NH_2 (4.6 mm i.d. x 250 mm; GL science), using acetonitrile/water as the mobile phase at a flow rate of 1.0 ml/min. The effluent was mixed with 2-CA in sodium borate buffer solution at 0.5 ml/min, and the reaction was carried out at 100°C in a 10 m x 0.5 mm i.d. coil. The UV absorbance of the resultant product was monitored at 276 nm using UV detector.

Results: Under optimal conditions, FDG and ClDG were successfully separated and detected with detection limits of sub-ppm levels for a 20-µl injection volume; these levels were far beyond the requirements for FDG and ClDG stipulated in USP and EP, although slightly worse than those of the HPAEC/PAD method. Sufficient linearity (0.5-100 ppm) and reproducibility were obtained. The R.S.D. for replicate injections (n=6) was better than 2.2 % (below the USP mandated limit of 5.0 % for FDG and ClDG). Day-to-day reproducibility, which was insufficient with the HPAEC/PAD method, was also satisfactory. The analysis was completed within 30 min and could be applied to [^{18}F]FDG preparations obtained with commercially available equipment. Moreover, this HPLC separation procedure allowed the analysis of [^{18}F]TAG, partially hydrolyzed [^{18}F]TAG and [^{18}F]F^-; it is also useful for the determination of radiochemical purity of [^{18}F]FDG preparations.

Conclusion: This HPLC technique was shown to be useful for the quality control of [^{18}F]FDG. Furthermore, this method might be acceptable at many PET facilities since it does not require an expensive and sophisticated electrochemical detector.

Keywords: [^{18}F]FDG, Post-Column Derivatization, Quality Control