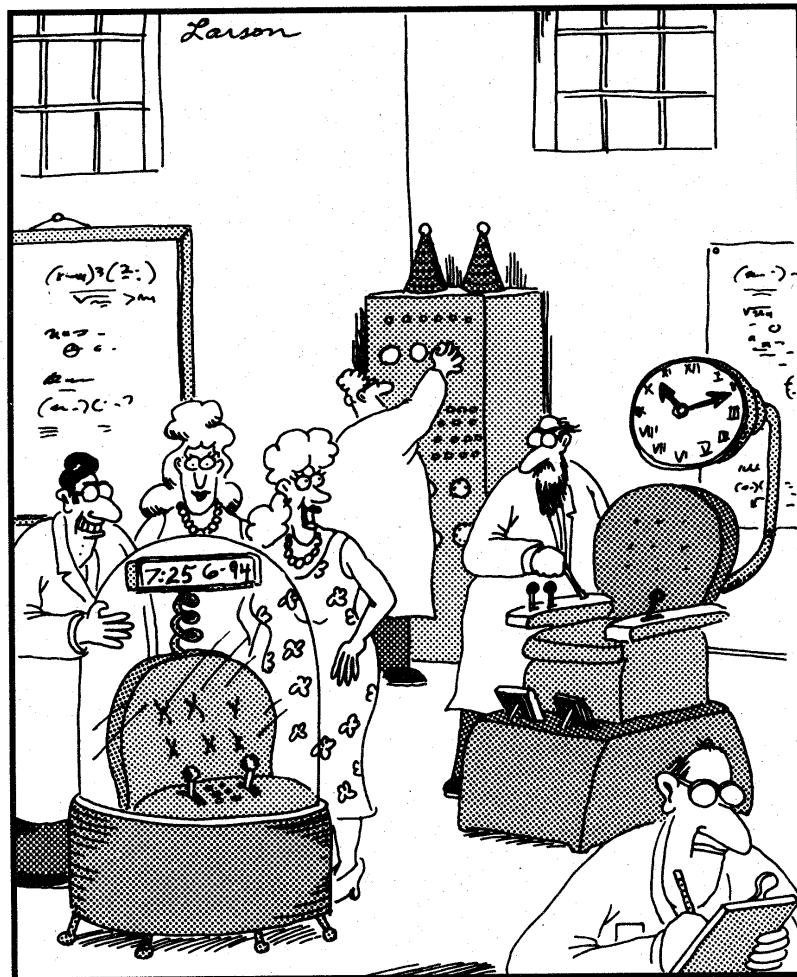




## Fourth Session

### Radionuclides, Targets, Processing and Automation

Co-Chairs: Gunter Firnau, Sven-Johan Heselius  
Jeanne Link, Nigel Stevenson



"Oh, Professor DeWitt! Have you seen Professor Weinberg's time machine? ... It's digital!"

## **Invited Talk:**

### **Surface Modification by Ion Beam Assisted Deposition**

**Prof. Gerhard K. Wolf**

University of Heidelberg, Institute of Physical Chemistry

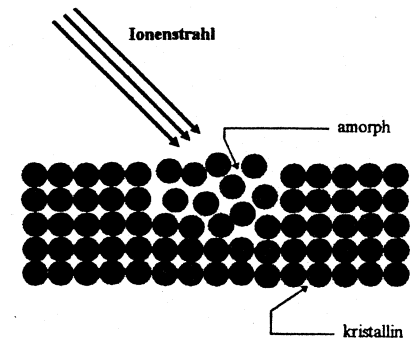
- 1. The Useful Tool “Ion Beam”**
- 2. Processes and Equipment**
- 3. Applications of Ion Beams**
  - **Structural Surfaces**
  - **Textured Surfaces**
  - **Metallizing of Polymers**
  - **Bactericidal Coatings**
- 4. Potential of Ion Beam Techniques for Targetry**



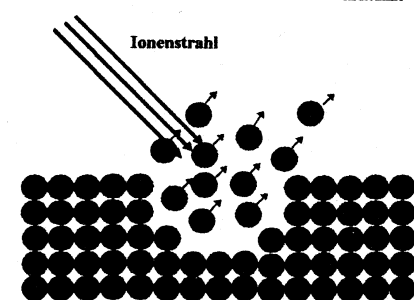


## 1. The Useful Tool “Ion Beam”

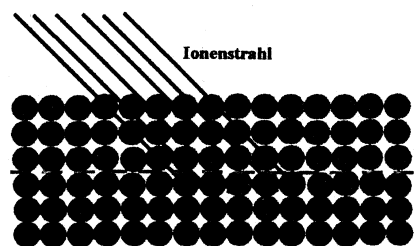
### Interaction of Ion Beams with Matter



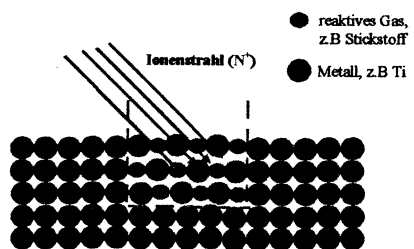
Destruction ( $\mu$  scalpel)



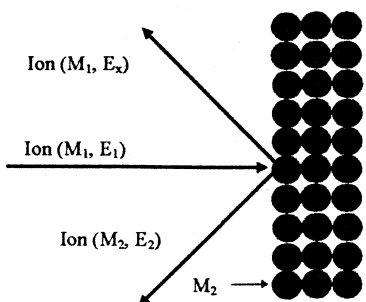
Sputtering ( $\mu$  sandblast)



Mixing ( $\mu$  stirrer)



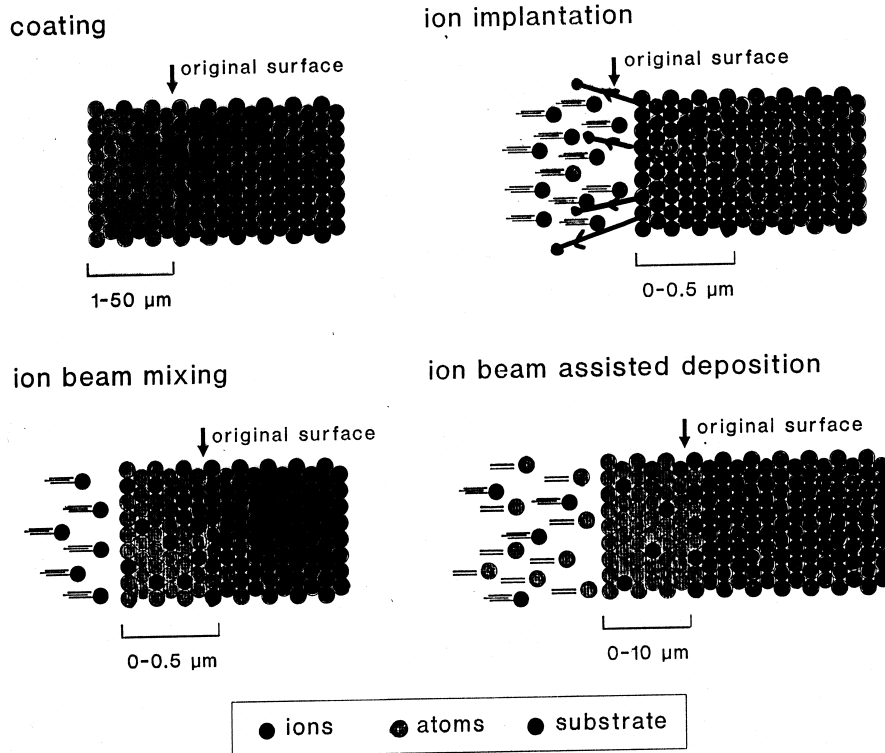
Alloying and Reaction ( $\mu$  alloying)



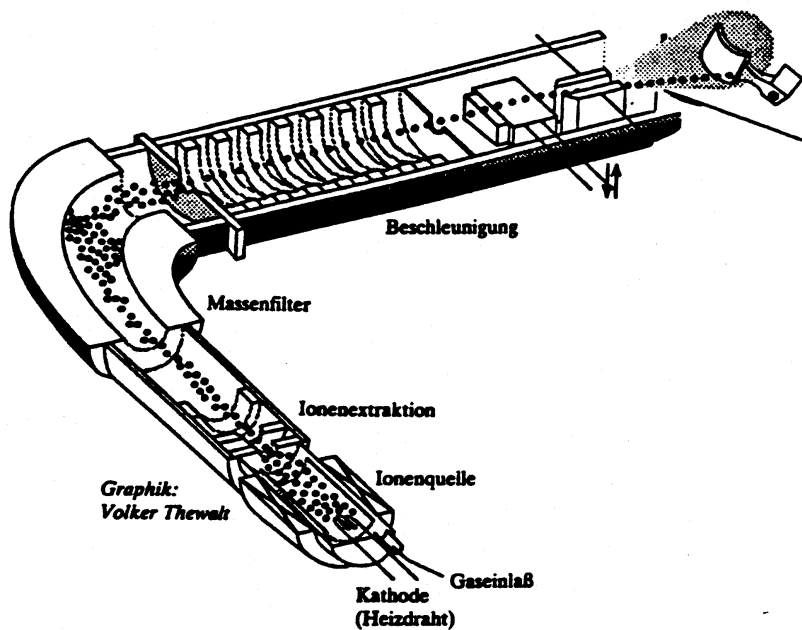
Compaction (lattice defects)

Analysis (non-destructive analysis of solids and depth profiles; RBS, NRA, PIXE)

## Different Ion Beam Techniques

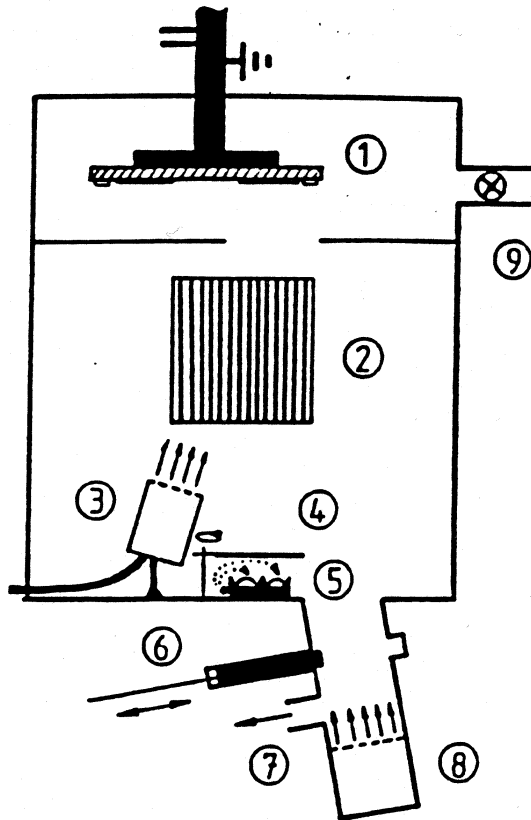


## 2. Process and Equipment

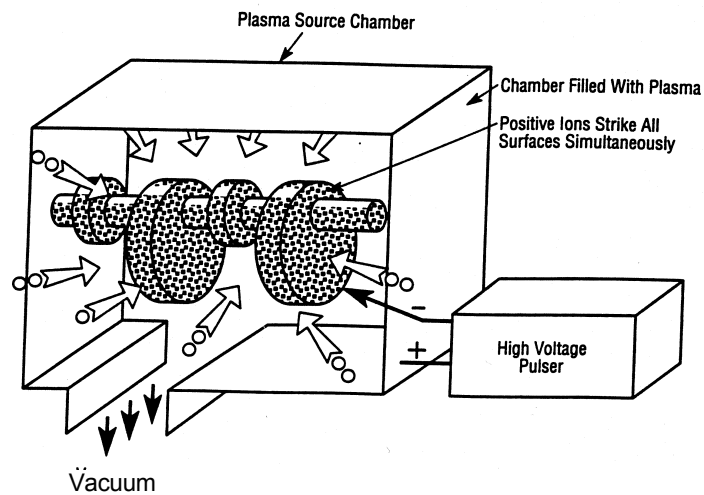


Ion accelerator with ion source, extractor, magnet, acceleration line and target station (typical parameters:  $V=200\text{ keV}$ ,  $I=1-10\text{ mA}$ )

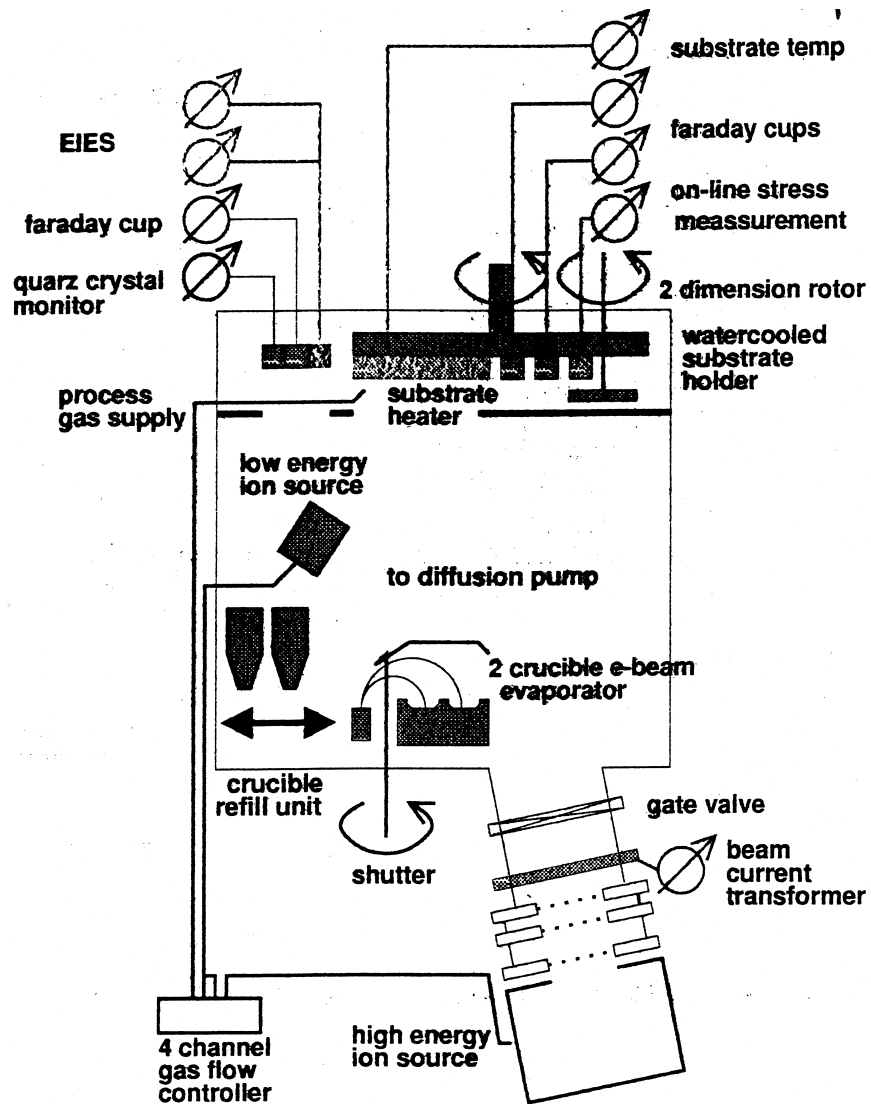
**Ion Beam Accelerator**



IBAD Nitride Layers



A plasma sheath surrounds the target in plasma immersion ion implantation. Therefore ions bombard all surfaces simultaneously without beam aiming or target manipulation.



Scheme of the Heidelberg IBAD system ALLIGATOR

### 3. Applications of Ion Beams: Surface Treatment

- **Irradiation and Sputtering (broad beam)**
  - Structured surfaces
  - Structures by masking
- **Focussed Ion Beam Treatment**
  - Structured surfaces
  - Ion beam CVD

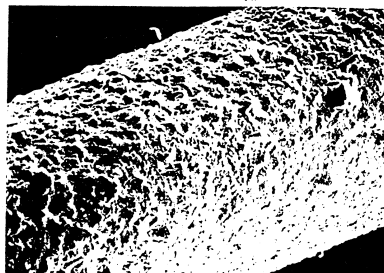


$\text{Pt} + 5 \times 10^{18} \text{ Ar}^+ / \text{cm}^2$

## Selected Structures and Uses: Structured Surfaces by Ion Beam Techniques

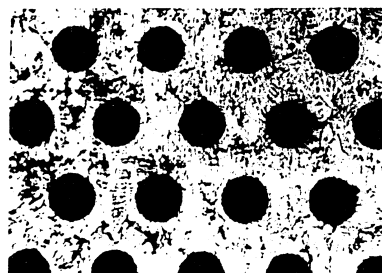
### Random Pattern (Hillock Structure):

- Pacemaker electrode tins, dental implants



### Precise Features with Straight Walls:

- Micro filters, contact lens molds, mammary prosthesis molds



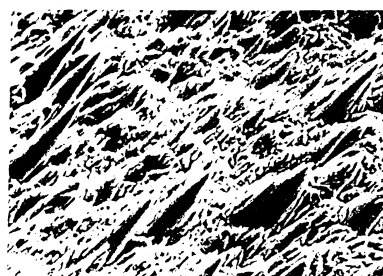
### Regular Pits with Angled Walls:

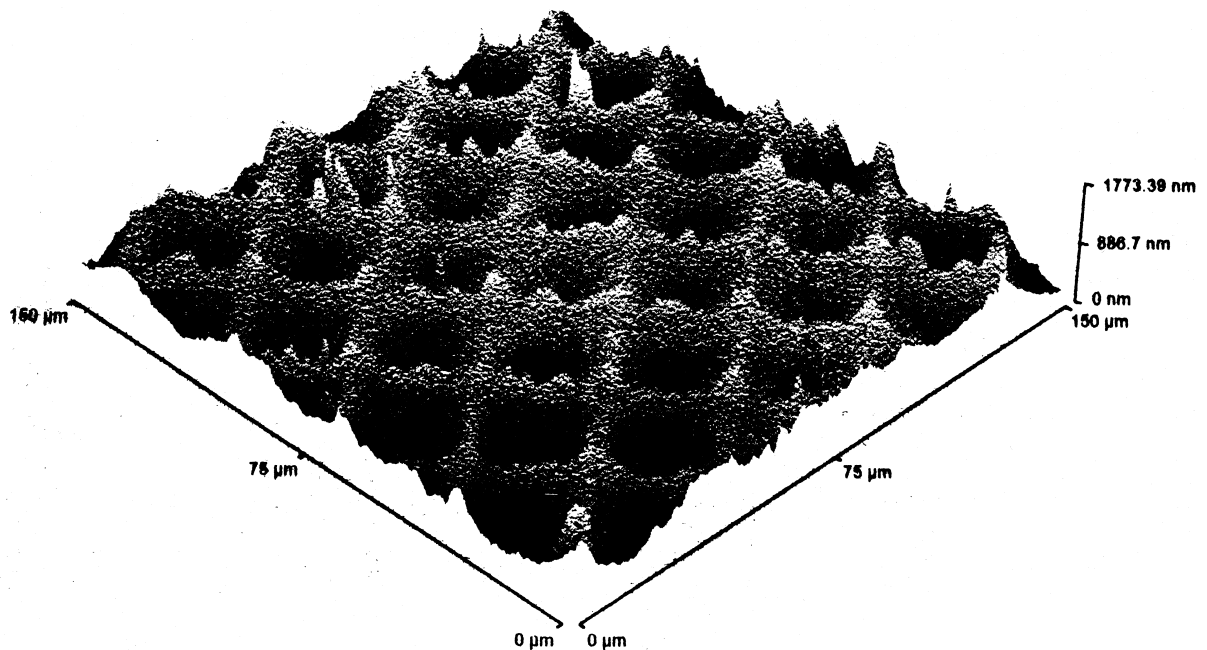
- Orthopaedic implants, heart valves, dental implants



### Irregular Micron Size Features:

- Pacemaker electrode tips, heart valves, defibrillator electrodes





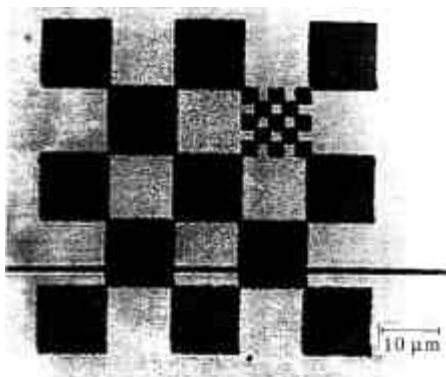
Structures generated by bombardment of a polymer with noble gas ions through a copper net and subsequent etching

#### Applications of Ion Beams: Ion Assisted Deposition of Single and Multilayer

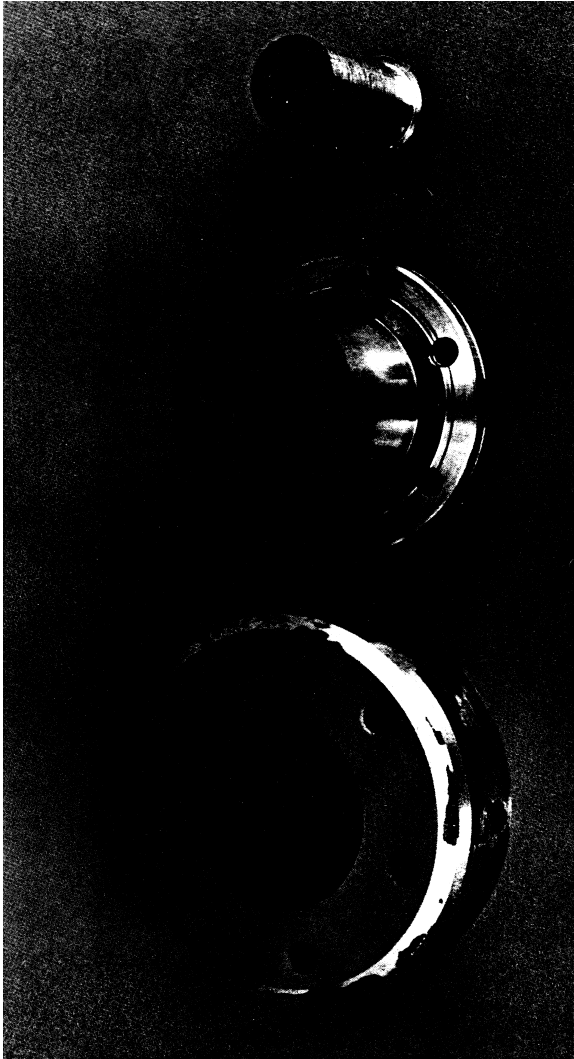
Elemental coatings

Compound coatings (e. g. bactericidal coatings on stainless steel, metallization of polymers)

Textured surfaces by ion beam assisted deposition



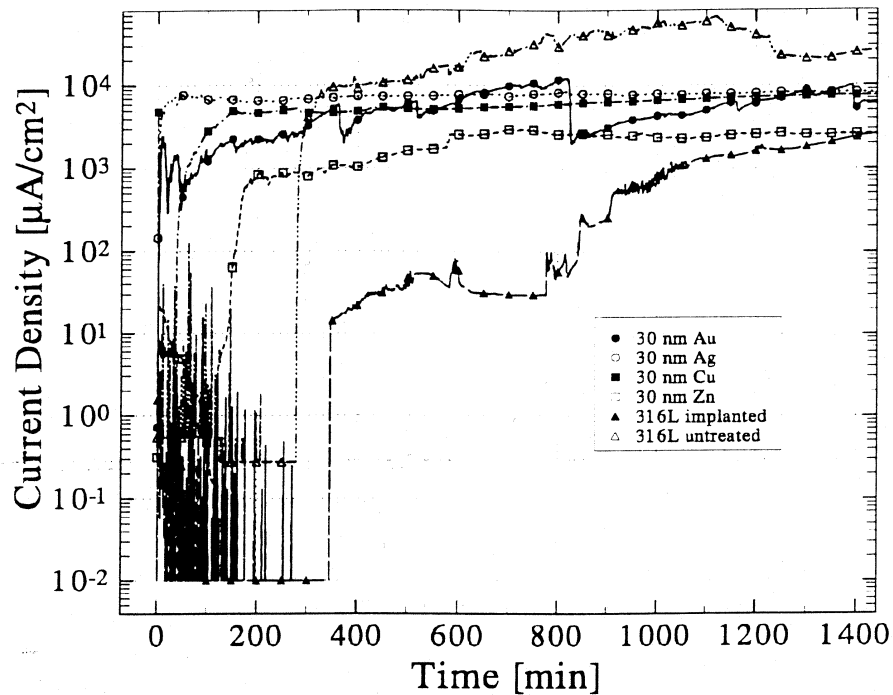
Optical pattern written with a gas field ion source (focussed  $\mu$ -Ne ion beam). The irradiated areas in polycrystalline SiC were amorphized. The smallest structures are approximately 2  $\mu$ m. Current densities of 10-100 A/cm<sup>2</sup> can be achieved.



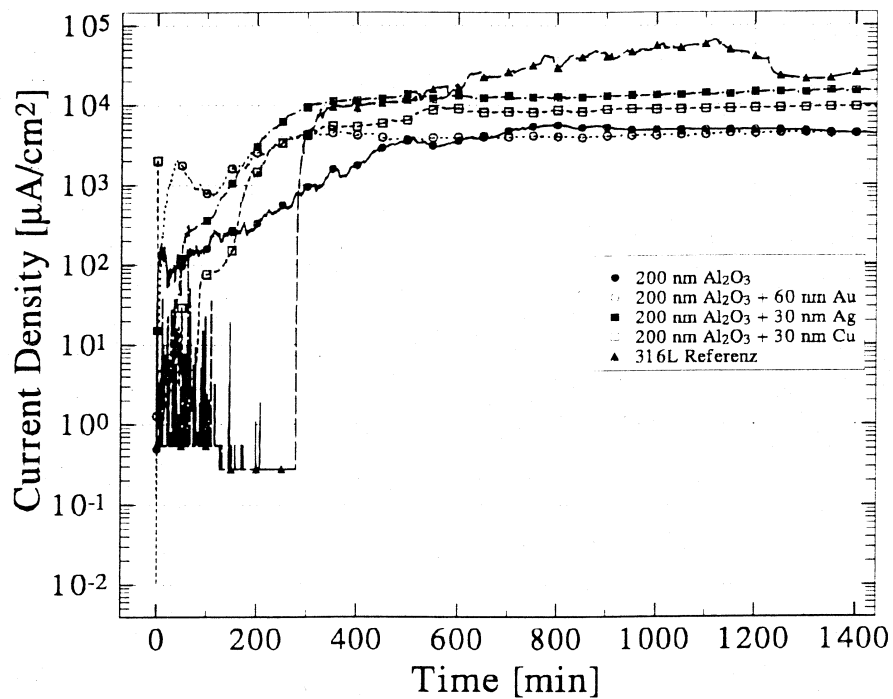
Ion beam modified set of press tools.  
The low tolerances required exclude  
regular PVD or CVD coating methods.  
(Photo: MAT Dresden)



## Chronoamperometrie in 0.9% sodium chloride solution



Corrosion of surgical stainless steel coated with different metals (ion beam mixing) in physiological saline. Increase of the corrosion current density indicates formation of holes.

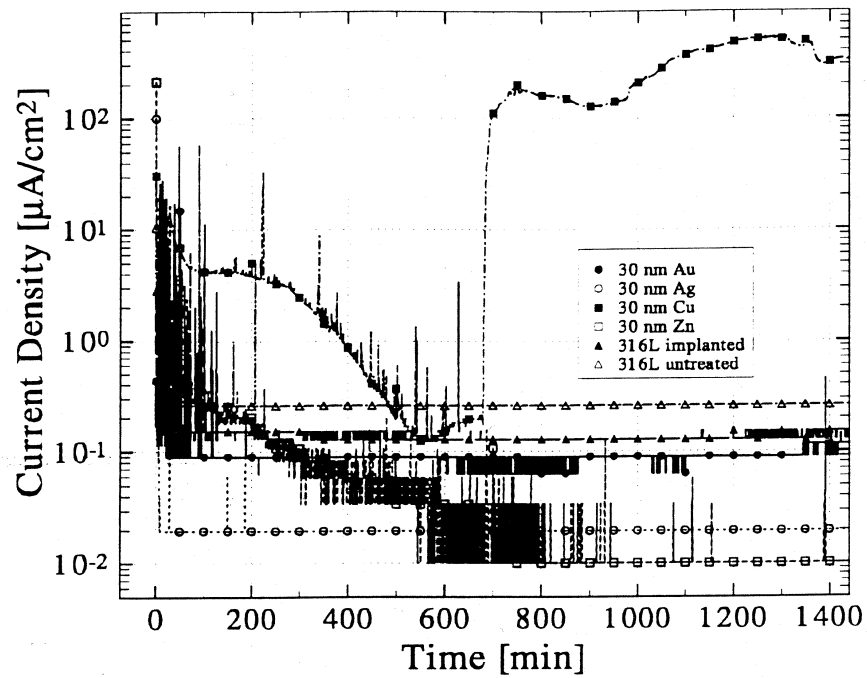


Same as above, but with aluminum oxide interface layer between stainless steel and metal layer.

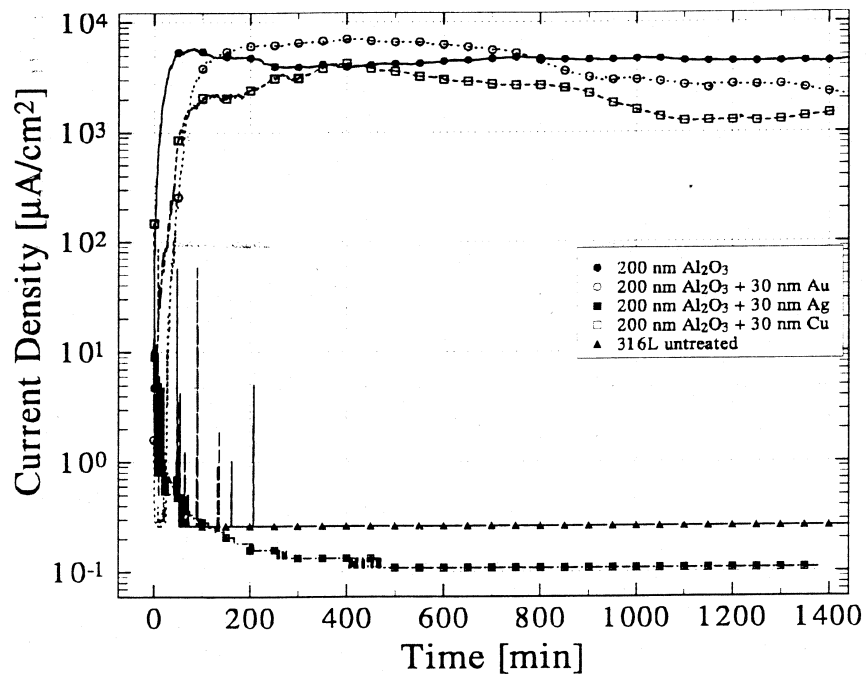




### Chronoamperometrie in human plasma



Same as above, but measured in human plasma (passivation of steel and metal coating except Cu)

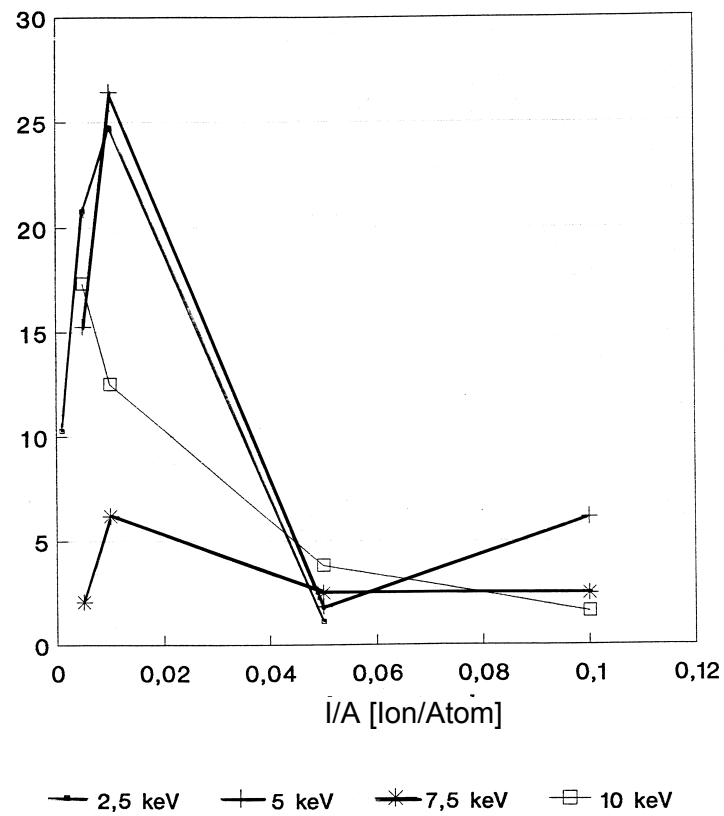


Same as above, but measured in human plasma (no passivation because of aluminum oxide interface layer; exception: Ag – AgCl precipitation)

## Adhesion of copper as a function of ion bombardment (ion / atom ratio)

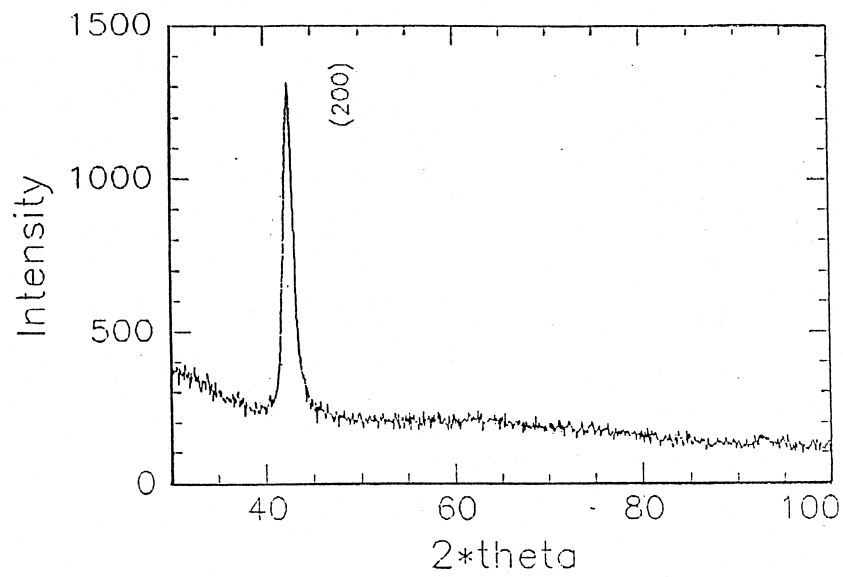
PPS (Polyphenylene sulfide), treated at 120 °C for 1 hr

Adhesion force [N/mm<sup>2</sup>]

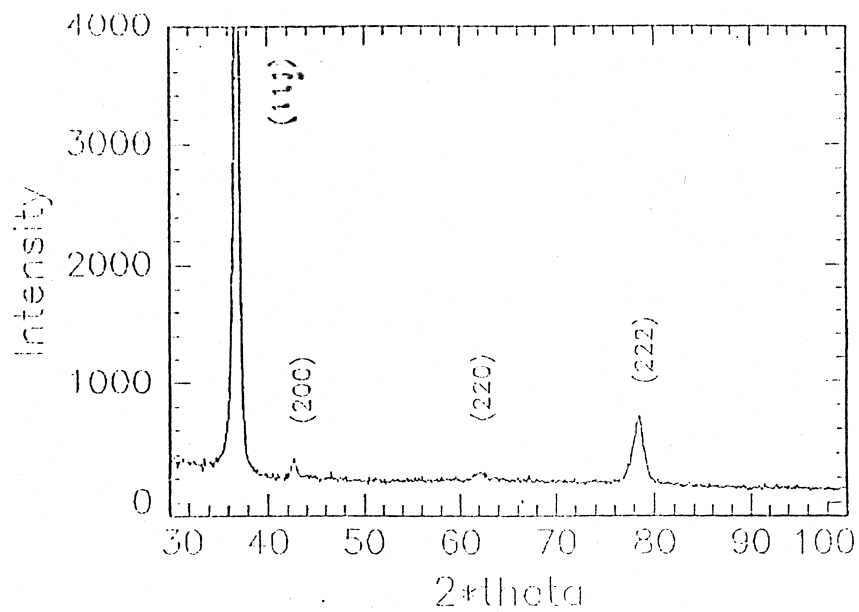




## Textures of IBAD TiN



TiN  $I/A = 0.1$   
(1 Ion : 10 Atoms)

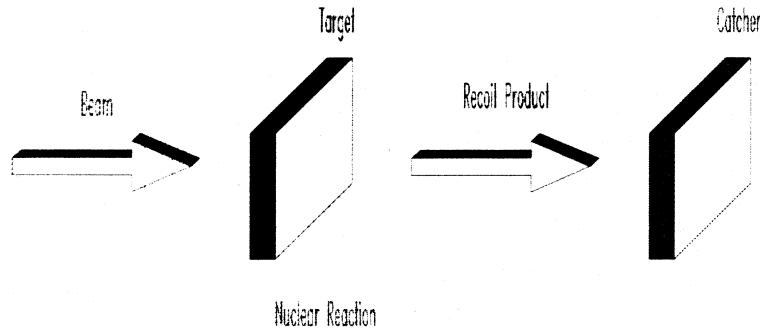


TiN  $I/A = 0.01$   
(1 Ion : 100 Atoms)



#### 4. Potential of Ion Beam Techniques for Targetry

- Target wheels with thin films for recoil separation



- Fine structured target: selective metallization of polymers and peeling off product
- Deposition of metals on metal substrates: good adhesion and complex shapes



Selective metallization of polymer through mask with copper

#### Discussion:

Q: T. Ruth: What are the limitations for the ion source? What kind of materials can you use, and what are the limitations?

A: It depends on the type of the source you are using. In principal you can use nearly every type. If you have the right source..... I wouldn't say plutonium or so. But any metal, if you have the right source, you can more or less evaporate any metal in the source and you get metal plasma and you can have metal beams or other types of source you can have all the rare gases or  $N_2$  or  $O_2$  and so on. So there is no principal limitation, it depends only that you need to have the right ion source.

Q: R. Ferrieri: ...(inaudible)...Is there any work on surface modification using cluster metal ion beams ...(inaudible)...

A: Yes, I mean not specially for the targetry, but there is also in Germany a group in Freiburg. They use cluster ion beams and they produce also very nice, smooth films. Cluster ion beams are very useful for having smooth surfaces, because they cluster like eggs that hit the surfaces. Smeared all over the surfaces.

Q: R. Ferrieri: ....very drastic differences in the nature of the surface modification... vapor deposit the metal and modifying it with a simple monatomic ion beam as opposed to depositing that metal as a clustered ion beam...

A: Yes, if you have the cluster ion beam, in principal you can produce craters by the cluster. But because of the smearing effect, the surface really doesn't see these craters. They are sub-surface craters and it depends also a little bit on the chemical nature of the substrate, what happens.

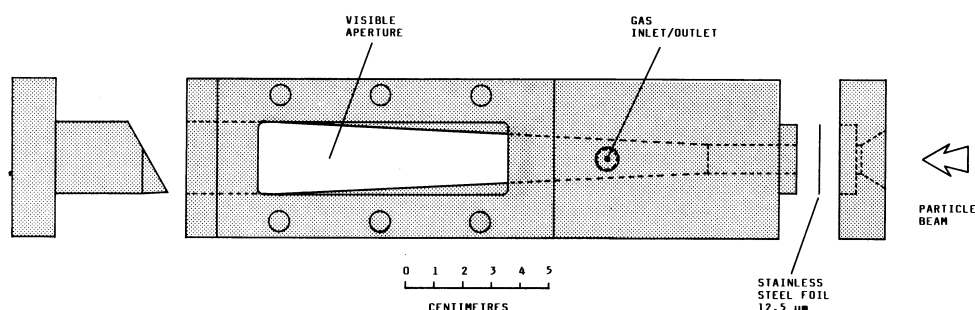
## Introduction to Session IVa: Particle Beam Penetration in Nitrogen Gas and Water Targets

**S.-J. Heselius**

Turku PET Centre, Accelerator Laboratory, Åbo Akademi University,  
Porthansgatan 3, FIN-20500 Turku, Finland

A report was given on the penetration of particle beams in gas and water targets. The following is a summary of this report covering the penetration of proton beams in nitrogen gas and water targets. This summary is based on results earlier reported in Refs [1] and [2]. The figures included in this summary are from Refs. [1] and [2] as well. The studies reported in Ref. [1] were carried out with the Åbo Akademi 103-cm isochronous cyclotron (Heselius, Malmberg, Solin and Långström) and the studies reported in Ref. [2] with the BNL 60-in. cyclotron (Heselius, Schlyer and Wolf).

### Proton-Beam Penetration in Nitrogen Gas Targets [1]



**Fig. 1:** Scale drawing of target chamber equipped with window for photographic beam

A conical target chamber equipped with a glass window was constructed (Fig. 1) and used in a photographic study of the proton beam penetration in nitrogen gas. The light emitted by nitrogen molecules in the beam volume of the target gas was photographed in both the vertical and the horizontal plane (Fig. 2). The proton beam penetration was measured from the photographs and the penetration increase in relation to the tabulated proton range [3] was determined. The target pressure was observed with a Kulite XTM-190 pressure transducer, with the exception of one study, where a pressure gauge was used. Two 12.5  $\mu\text{m}$  stainless steel foils (Goodfellow Metals Ltd.) separated the accelerator vacuum from the target pressure. The proton beam was collimated by apertures of 3 - 8 mm diameter before entering the target. The visible aperture of the target chamber was 87 x 25 mm (for location, see Fig. 1). The length of the target chamber was 187 mm and the volume 56  $\text{cm}^3$ . The target chamber was made of aluminum and equipped with channels for the cooling media. Two different cooling media, water at +5 °C and alcohol at -25 °C, were used in the studies. Nitrogen N4.8 ( $\text{N}_2$ , purity 99.998 %, Aga Special Gases) was the target gas.

The proton beam penetration in nitrogen gas was studied using four different target system volumes (56, 88, 150 and 1060  $\text{cm}^3$ ). The target system volume was varied by connecting different buffer volumes to the target chamber. Using the 150  $\text{cm}^3$  target system the beam penetration in flowing nitrogen (flow rate 100-500  $\text{cm}^3/\text{min}$ ) was studied.

Using the windowed target chamber the penetration of 11.5 and 10.4 MeV proton beams in the nitrogen gas target was studied as a function of the beam current for 750-1100 kPa target pressures. Using the four different target system volumes the beam penetration in the target was studied in detail for 10.4 MeV protons on nitrogen as a function of the beam



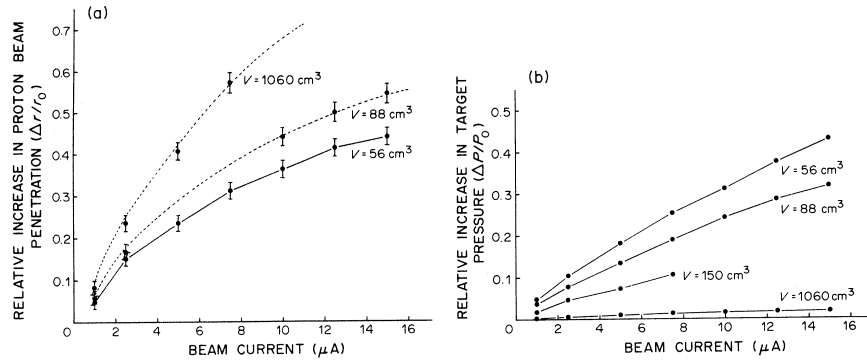
**Fig. 2:** Vertical view (left) and horizontal view (right) of nitrogen gas target at 1.05 MPa initial pressure, bombarded with 11.5 MeV protons of 1.0 – 5.0  $\mu\text{A}$  beam current. Target system volume = 56  $\text{cm}^3$ . Beam entrance from the right. Distance between two vertical lines is 1.75 cm. For location of visible aperture, see Fig. 1. Collimator diameter = 0.5 cm. [1]

current on the target. The initial target pressure was 1.088 MPa in all runs. An increased proton beam penetration in the target was observed when the beam current was increased, due to the reduction of the gas density in the beam volume of the target. The range of the lowest part of the proton beam, most clearly observed in the horizontal views for a low beam current on the target (e.g. 1  $\mu\text{A}$ , Fig. 2), corresponded within the experimental errors to the tabulated proton range  $r_0$  [3] at the initial target pressure and temperature. The experimental uncertainty was  $\leq 1.5\%$ . The increase in beam penetration related to the tabulated particle range ( $\Delta r/r_0$ ) is shown as a function of beam current in Figs. 3a and 4 for the four target system volumes. As can be seen, the proton beam penetration in nitrogen was strongly dependent on the target system volume. The relative increase in proton beam penetration for the 150  $\text{cm}^3$  target system volume was 50 % for a 7.5  $\mu\text{A}$  beam current on the target, whereas that for the 56  $\text{cm}^3$  target system volume and the same beam current was 31 %. The target pressure increase in relation to the initial pressure ( $\Delta P/P_0$ ) is shown in Fig. 3b as a function of beam current.

Using the 150  $\text{cm}^3$  target system the proton beam penetration in the nitrogen gas target was studied under flow-through conditions for the gas flow rates 100, 200, 300, 400 and 500  $\text{cm}^3/\text{min}$ . The calculated  $\Delta r/r_0$  values are shown in Fig. 4. A small variation in the  $\Delta r/r_0$ -values was observed at the flow rates used. This was within the limits of the experimental uncertainty (Fig. 4). No target pressure increase as compared to the initial target pressure (1.088 MPa) was seen at steady state conditions with beam on the target. No significant

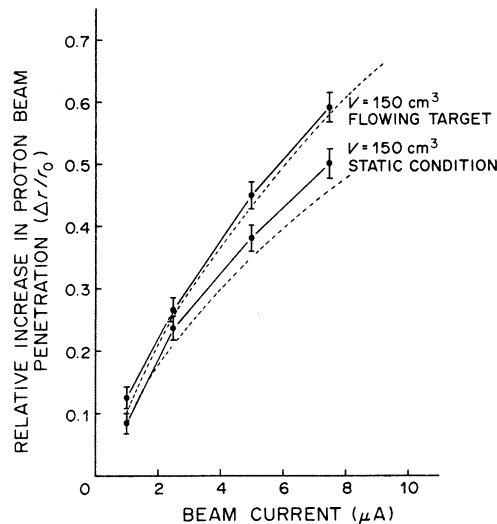


difference was observed in the proton beam penetration and scattering when alcohol at  $-25^{\circ}\text{C}$  was used as cooling medium, as compared to the values obtained in the target cooled with water.



**Fig. 3:** (a) Relative increase in beam penetration ( $\Delta r/r_0$ ) and (b) relative increase in target pressure ( $\Delta P/P_0$ ), for 10.4 MeV protons on nitrogen gas as function of beam current on target for different target system volumes ( $V$ ). Initial target pressure  $P_0 = 1.088 \text{ Mpa}$ ,  $r_0$  = tabulated proton range. The dashed lines represent the values for  $\Delta r/r_0$  predicted by the model (beam scattering angle  $\alpha = 2^{\circ}$ ). [1]

The photographic study showed that a target chamber equipped with a glass window is a convenient beam diagnostic tool for the determination of the target pressure required for thick target conditions at a certain beam current and energy. The photographic view reflects the shape of the beam in the gas target and approximately gives the maximum penetration of the beam. A strongly volume dependent beam penetration in nitrogen gas targets was observed. A large bulk volume in the target system allows the beam volume to expand. Thus, the volume of the windowed experimental target chamber has to be comparable to the volume of the corresponding production target chamber. The relative increase in beam penetration was found to be higher in a gas target irradiated under flow-through conditions than in the target under static condition. The volume of the target system in the flow-through



**Fig. 4:** Relative increase in beam penetration ( $\Delta r/r_0$ ) for 10.4 MeV protons on nitrogen gas under static and flow-through conditions as function of beam current on target. Gas flow rates =  $100\text{-}500 \text{ cm}^3/\text{min}$  for flowing condition. Target system volume =  $150 \text{ cm}^3$ . Initial target pressure  $P_0 = 1.088 \text{ Mpa}$ . The dashed lines represent the values for  $\Delta r/r_0$  predicted by the model (beam-scattering angle  $\alpha = 2^{\circ}$ ).

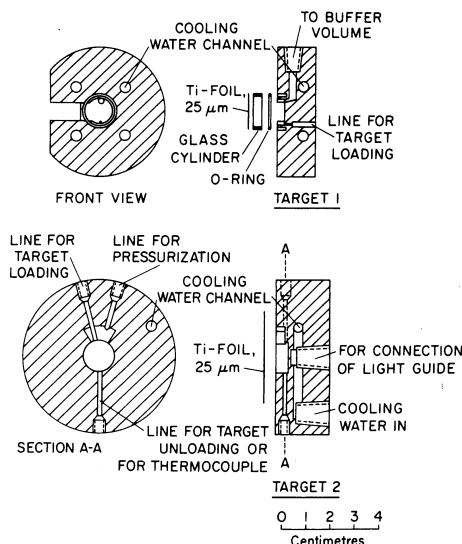




condition can be considered as infinitely large. As can be seen in Fig. 4, the relative increase in beam penetration was higher in the  $150 \text{ cm}^3$  gas target under flow-through conditions than in the  $150 \text{ cm}^3$  target system under static condition. As expected the cooling effect of the flowing target gas has no influence on the density reduction in the beam strike at the low gas flow rates used ( $100\text{--}500 \text{ cm}^3/\text{min}$ ).

### Study of Proton Beam Irradiated Water Targets [2]

The target chambers used for studying proton beam irradiated water targets are schematically shown in Fig. 5. Target 1 was used in a photographic study. The target water was photographed through a glass window. The width of the glass aperture was  $2.9 \text{ mm}$  and the height  $7.9 \text{ mm}$ . A titanium foil (thickness  $25 \mu\text{m}$ ) was glued to the  $12.5 \text{ mm i.d.} \times 15.0 \text{ mm o.d.}$  glass cylinder which was inserted into an O-ring groove of the aluminum target body. The glass cylinder was pressed into the groove by the water-cooled aluminum helium cooling unit and sealed with a Viton O-ring. The target water loading volume was  $0.3 \text{ mL}$ .

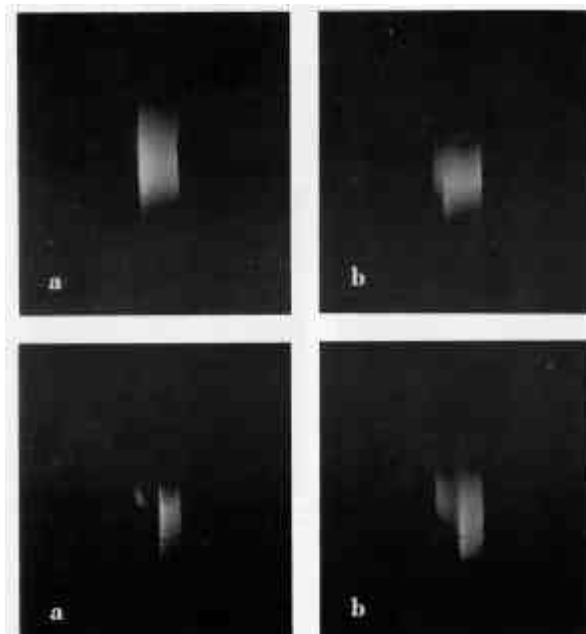


**Fig. 5:** Schematic diagrams of diagnostic target units. The upper target unit (Target 1) was equipped with a glass window for photography of target water during irradiation. The lower target unit (Target 2) was used in a multi-parameter study. [2]

The proton beam was collimated by a  $7.9 \text{ mm}$  aperture in an aluminum flange mounted at a  $3.5 \text{ cm}$  distance from the  $25 \mu\text{m}$  thick titanium target entrance foil. The  $17.7 \text{ MeV}$  proton beam energy was degraded by a  $0.116 \text{ g/cm}^2$  thick aluminum foil permanently mounted in the target assembly at a distance of  $2.2 \text{ cm}$  from the target entrance foil. Helium cooling gas was streaming in between the aluminum beam energy degrader foil and the titanium target entrance foil. The energy incident on the target water was  $14.7 \text{ MeV}$ .

When water is irradiated with a particle beam the water molecules are excited by the beam and emit visible blue light during the deexcitations. The light emitted in the target during irradiation was photographed through the glass window on the target. The light emission was photographed as integrated over a  $4 \text{ s}$  film exposure time at a  $90^\circ$  angle to the particle beam in the horizontal plane. Light emission from excited water molecules was used in order to study the penetration of the proton beam in water targets. In order to confirm that the bright light emission observed in the pictures really reflects the penetration of the proton beam in the target the light emission was photographed with the  $25 \mu\text{m}$  titanium target window and the  $0.116 \text{ g/cm}^2$  permanently mounted aluminum foil as energy degraders. Two exposures

were recorded, one for 5  $\mu\text{A}$  beam current and the other for 10  $\mu\text{A}$  beam current (Fig. 6). Then, without change of any parameter, another 0.116 g/cm<sup>2</sup> aluminum energy degrader was inserted into the target assembly in front of the collimator at a distance of 4.7 cm from the target entrance foil. Two exposures of the light emission from the target were again recorded for the same beam currents on the target as above.



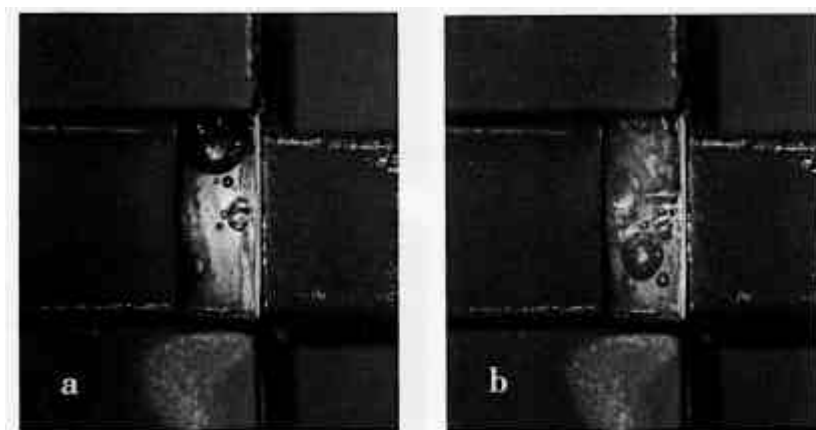
**Fig. 6:** View of light emission from the water target irradiated with 14.7 MeV protons (top) and 11.6 MeV (bottom). (a) shows light emission for a 5  $\mu\text{A}$  beam current and (b) for 10  $\mu\text{A}$ . Light emission from the target was integrated over a 4 s film exposure time. The vertical bright lines in the pictures reflect the intense light emission corresponding to the high value of the linear energy transfer in the Bragg peak. Beam entrance from the right. For beam collimation and energy degradation of the 17.7 MeV proton beam, see text. For location of aperture (width 2.9 mm, height 7.9 mm), see Fig. 5. [2]

The penetration of the proton beam in the target could clearly be observed by measuring the position of the intense light emission corresponding to the high linear energy transfer in the Bragg peak. A shift in the position of the bright lines in the photographs was clearly observed when the additional degrader foil was placed in front of the target. The proton energies entering the target water was calculated to be 14.7 MeV and 11.6 MeV for the arrangements without and with the degrader foil in place, respectively. The ranges of the proton beams as measured from the photographs agree with tabulated ranges. [3]

During irradiation of water at atmospheric pressure with an intense focussed proton beam (of the order of 20-30  $\mu\text{A}/\text{cm}^2$ ) the target medium is boiling and gas bubbles are formed in the target water. These bubbles were observed by photographing the target water through the glass window on the target body. Due to the formation of bubbles in the target medium the proton beam even penetrates a target calculated to be 200 % range thick, and further the target back wall into the cooling water if the wall is chosen to be thin enough.

In the photographic views showing the integrated light emission from the target water (Fig. 6) light can also be observed behind the vertical bright line. This is due to gas bubbles formed in the target during irradiation (Fig. 7), which allow the beam to penetrate further than the range calculated for liquid water. From the views in Fig. 6 we can also clearly see that the fraction of light behind the bright line is greater in the exposures recorded for a 10  $\mu\text{A}$  beam current than in the exposures recorded for 5  $\mu\text{A}$  beam current. This is also in agreement with the fact that fewer bubbles were formed for a lower beam current (Fig. 7). The fact that the

bright line remains visible suggests that the water was not boiling during some fraction of the time. When the water was in this state, the water stopped the beam at the predicted range. When there were bubbles present, the density was lower and thus the range of the proton beam was extended through the back wall of the target resulting in a less intense light emission throughout the beam path. All photographic views were recorded with the target irradiated at atmospheric pressure.



**Fig. 7:** Views showing formation of bubbles in the water irradiated with 14.7 MeV protons of (a) 5  $\mu$ A and (b) 10  $\mu$ A beam currents. Beam entrance from the right. For beam collimation and energy degradation of the 17.7 MeV extracted beam, see the text. For location of aperture, see Fig. 5. [2]

#### References:

- [1] S.-J. Heselius, P. Malmberg, O. Solin and B. Langstrom, Studies of Proton Beam Penetration in Nitrogen-Gas Targets with Respect to Production and Specific Radioactivity of Carbon-11, *Appl. Radiat. Isot.* **38** (1987) 49-57.
- [2] S.-J. Heselius, D. J. Schlyer and Alfred P. Wolf, A Diagnostic Study of Proton-beam Irradiated Water Targets, *Appl. Radiat. Isot.* **40** (1989) 663-669.
- [3] J.F. Janni, *At. Data Nucl. Data Tables* **27** Nos 4/5 (1982).

#### Discussion:

Q: G. Wolber: Have you tried to make this last test with higher pressures to suppress bubble building ?

A: We really were not able to photograph it, but we used another technique. We made an experimental arrangement, just having quite a tiny wall behind our water volume. So we constructed a target in order to have the beam penetrating the water target and also the back wall in case we have bubbles. And then we were looking from the cooling water volume. We were looking if we can see light in the cooling water or not. And when we got bubbles in the target volume, we really could see quite intense emission of light in the cooling water. We were recording this also, so we could get quite interesting pictures of this. The more bubbles, the more intense light in the cooling water. But we can talk about this another time. To answer your question, this has been done.

Q: J. Steinbach: Back to the gas target. Please give us a comment on the angle of beam spreading.

A: This is of course partly due to spreading in the target entrance foils and also in the target gas.

J. Steinbach: Yes, that's clear, but do you have an idea how much is it?

A: As a mean we can talk about 2°, 3°, 4°. It depends on your beam, if you have an  $\alpha$ -particle beam or a proton beam. For a proton beam in this case of 11.5 MeV it was 2°.

Q: G. Firnau: So you would guess that our commonly 5° of a conical surface is fine?

A: Yes, it's fine.

### **Olof Solin, Turku: Spectroscopy of a High Purity Nitrogen Target during Irradiation**

I want to just briefly present some work that we have been doing on gas targets. It is a continuation of what Sven-Johan presented, but more recent work from the last two years. We study spectroscopically the light emission from nitrogen gas targets. The experiments have been done in a pressure range from 100 mbar to 150 bar. The target is not the same for all pressures, of course, we used four different targets depending on the pressure we wanted to study. At the highest pressure, we have a special gridded foil technique, similar to what has been described by Jerry Nickles already in 1980. And it still works.

We have the target, and an optical fibre to transmit the light of the target chamber, the spectrograph which is rather special because it can record very broad optical spectra in a single exposure from 200 nm to 1070 nm. This is in about 30,000 channels, so the resolution is also very good.

What we have been studying is pure nitrogen. If you put any additives into the nitrogen, what you will see is very much different. This is pure nitrogen, I would like to stress that.

The strong peak here, that's a molecular transition in nitrogen, the Gaden's Green it is called, that's a very strong emission, the Herman's infrared also, the nitrogen second positive system. Also you see the first positive system, this is of low intensity. Then we see, interestingly enough, the forbidden atomic nitrogen transition 2d-2p, that is also a very strong transition. If we have any oxygen in the gas, we see a very strong gamma system of nitrous oxide.

When we vary the pressure, you can see on the x-axis there is the gas pressure going from 100 mbar to 150 bar. On this axis we have the emission of the Herman's infrared, remember, I showed you the zero-zero transition at 805 nm. If we normalize that against the second positive system we get a quadratic increase in the light coming out from the target as a function of pressure. So this is a constant current. From the spectra recorded at different pressures we can calculate the peak area of the second positive system and of the other systems. We use the second positive system as the normalizing for all effects that happen when we vary the pressure because we had different targets, different geometries, when we vary the pressure, the beam is stopped in a shorter volume, so we get effects of that kind. But we can normalize that because the second positive system can be considered as a measure of direct excitation power of the beam coming into the gas. It is interesting that the infrared light that comes out increases to the second power as a function of the pressure. That means that the mechanism that generates this light must be a three body process.

In this diagram we have potential curves for the nitrogen molecule. On the x-axis is the molecular diameter, here is the energy, this corresponds to about 10 eV. For example, the Herman's infrared transition corresponds to the c'' - a' transition, and the Gaden's green is the h-g transition. The second positive system is the c-b transition here, deep inside the

molecule. What we are thinking is that the mechanism that generates the light is a three body recombination of two excited atomic nitrogen and a molecular ground state nitrogen. For example, for the Herman's infrared it will be the 4s-2d and the molecular nitrogen, and for the Gaden's green system the 2d and the molecular nitrogen. As you remember, we also saw the forbidden transition of the nitrogen 2p-2d, and that is interesting because we don't see any *molecular* transition involving the 2p atomic nitrogen here. It seems that this is already to energetic to be able to recombine in a similar process in molecular nitrogen.

Because this is a three body process, it requires to have three nitrogen species two atomic and one molecule, it will also be very sensitive to any contamination that is introduced into the target. What we show here as a function of irradiation time in a closed target with high purity nitrogen is that we see as a function of accumulated dose the emission from the Herman's infrared goes down, and simultaneously the emission from the nitrous oxide gamma system goes up. This is a high pressure aluminum target, and we interpret this as a slow sputtering or leaking into the target of oxygen from the aluminum walls.

Q: T. Tewson: Olof, at the Vancouver meeting you reported that when on your nitrogen target when the color changed, if I remember correctly from blue to green.....

Solin: Green to blue

Tewson:.....the carbon stopped coming out.

A: Oh, that one, that's right, but it changed from blue to green and then the carbon stopped coming out.

Tewson: Whereabouts on your species here does the carbon stop coming out? In other words, in the transition that you are looking at here, has the carbon stopped coming out?

A: If you means this.... the carbon starts to come out, when this transition stops.

Q: G. Wolber: It's amazing that you apparently can at least identify a few compounds arising during the reaction in the target. Would it perhaps be possible to get hints on the in target radiochemistry, on the hot chemistry within the targets, so perhaps you could make use of the compounds which arise and make perhaps use for further labeling procedures?

A: I certainly think so, yes. This is the technique.

Q: Have you actually done spectroscopy of the water target irradiations?

A: Yes, we have. Haven't seen much.

## Glass Target for Production of [ $^{13}\text{N}$ ]NH $_3$ from Methane Revival of an Old Method

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Production of [ $^{13}\text{N}$ ]NH $_3$  - used for measuring myocardial blood flow with PET - takes place mainly by the  $^{16}\text{O}(\text{p},\text{n})^{13}\text{N}$  nuclear reaction [1]. Although the  $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$  reaction gives higher yield and can be utilised on low energy machines, its use is less common because of the technical difficulties. For the latter reaction one can use graphite, carbon slurry or methane as target materials [2,3,4]. Producing [ $^{13}\text{N}$ ]NH $_3$  from methane was introduced by Tilbury et al. [4] in 1971, and Straatman et al. [5] proposed a distillation method for purification. However the method was not used mainly because of the radiation by-products of methane. We are using the Tilbury method for production of [ $^{13}\text{N}$ ]NH $_3$  with a new approach for purification.

The target is a 1 m long Pyrex glass tube through which research grade methane (99.995 %) is flowing continuously. The flow rate is 800-1000 mL/min at 1.5 bar target pressure. The gas leaves the target through a 0.8 mm inner diameter Teflon tube and bubbles into 10 mL distilled water. In a typical run we irradiate the methane with 14 MeV deuterons at 12  $\mu\text{A}$  for ten minutes. This results in 3500 MBq of  $^{13}\text{N}$  activity, mainly in the form of ammonia. After the irradiation the bubbling is continued for an additional two minutes then the solution is transferred into the hot cell. Here it is passed through a flow sensor, a C-18 Sep-Pak cartridge and a cation exchange column (3x25 mm Biorad AG $^{\text{®}}$  50W-X2, 50-100 mesh, H $^+$  form). The cation exchange column retains quantitatively the ammonia. Afterwards the column is washed by 5 mL water then the ammonia is eluted from the column by 10 mL 0.02 M disodium hydrogen phosphate 0.09 M sodium chloride solution. The isotonic column effluent is filtered through a sterile membrane filter (Millex $^{\text{®}}$ -GV, Millipore) directly into the injection syringe. The process takes about ten minutes and gives around 1200 MBq  $^{13}\text{NH}_3$  in a pH 6-7.5 solution.

The radiochemical purity of the final product is determined by the HPLC method of Gatley and Shea [7]. The conditions for the analysis is: Partisil P10 ODS3 (C $_{18}$ )250x4.6 mm column eluted at 2 mL/min with a 2:1 sodium-1-octansulfate (pH 7.0) and acetonitrile; ZA 212 plastic scintillator coupled with a photomultiplier radioactivity detector; KNAUER differential refractometer type 98.00. The typical analysis only shows one peak in the chromatogram at 3.1 minutes (k'0.54), which corresponds to the  $^{13}\text{NH}_3$ .

Our work shows that methane can be used as a target material for routine  $^{13}\text{NH}_3$  production. The system could be easily automated with the help of a photodiode based flow sensor on the Teflon tubing. This would provide a signal at the end of each step allowing the immediate continuation of the process; thus one can avoid the time loss of the usual, time based systems. To guarantee high quality, there are four major elements in our system that determine the purity of the final product:

1. *The bombarding deuterons are fully stopped in the target, thus the end of the tube remains cool allowing the long chained radiolysis products to condense.*
2. *The water insoluble products remain in the bubbling vessel.*
3. *The soluble unpolar products and the methyl and ethyl amines are retained on the C-18 Sep-Pak cartridge.*
4. *The anionic species (as [ $^{13}\text{N}$ ]CN) are not retained on the cation exchange column, thus will not be present in the final solution.*





The system has an overall 70 % decay corrected yield, and can easily produce the necessary activity for a myocardial study (usually 740 MBq).

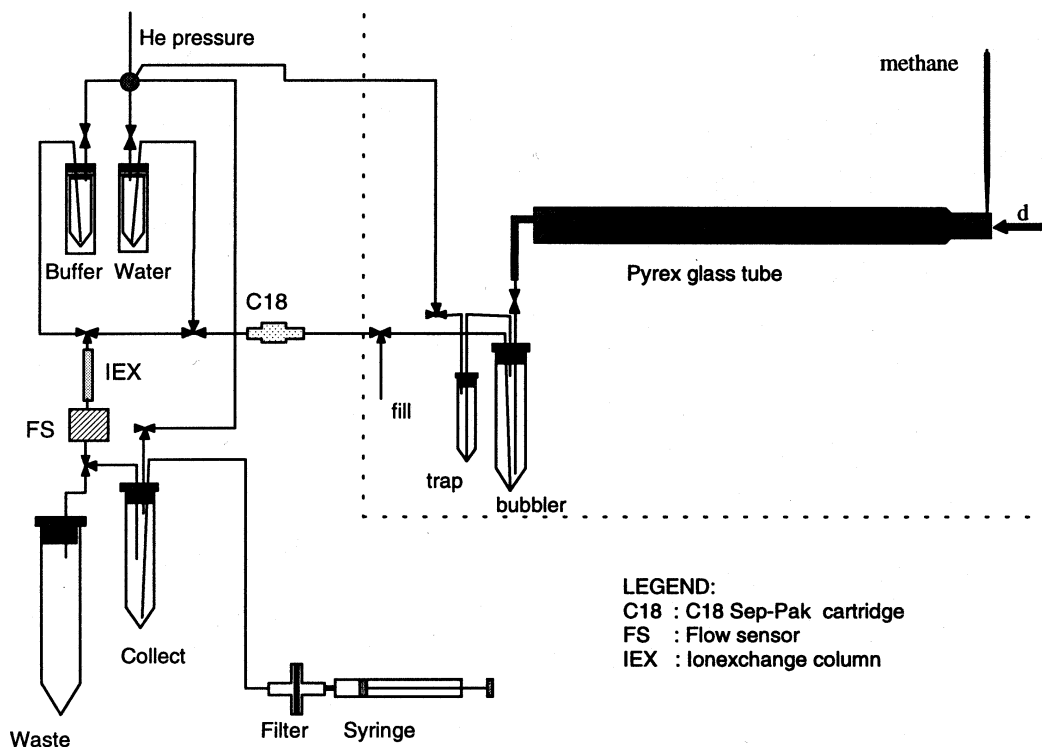


Fig. 1: Schematic outline of the  $^{13}\text{NH}_3$  production system

#### References:

- [1] B. Wieland, G. Bida, H. Padgett, G. Hendry, E. Zippi, G. Kabalka, J. Morelle, R. Verbruggen, M. Ghyoot, Appl. Radiat. Isot. **42**: 1095 (1991)
- [2] C.S. Dence, M.J. Welch, B.J. Hughey, R.E. Shefer, R.E. Klinkowstein, Nuclear Medicine and Biology **21**: 987 (1994)
- [3] G. Bida, B.W. Wieland, T.J. Ruth, D.G. Schmidt, G.O. Hendry, R.E. Keen, J. Label. Compds. Radiopharm. **23**: 1217 (1986)
- [4] F. Oberdorfer and W. Maier-Borst, J. Label. Compds. Radiopharm. **30**: 191 (1991)
- [5] R.S. Tilbury, J.R. Dahl, W.G. Monaghan and J.S. Laughlin, Radiochem. Radioanal. Lett. **8**: 317 (1971)
- [6] M.G. Straatman and M.J. Welch, Radiat. Res. **56**: 48 (1973)
- [7] J.S. Gatley and C. Shea, Appl. Radiat. Isot. **42**: 793 (1991)

#### Discussion:

Q: R. Dahl: I took part in the earlier work with Tilbury on this and I'm curious. Did you look at the spectrum of impurities that came out of the gas in the target before you bubbled it through the saline? And what did you see if you did look?

A: No, I'm afraid we didn't do it, we collected condensed product in the tubing. And I hope to analyze it with GC-MS. But no, we haven't analyzed the product.

Q: R. Dahl: Have you taken the glass target tube off yet? Have you had that disassembled?

A: Yes.

Q: R. Dahl: Did you notice any strong amine odor?

A: No.

C: R. Dahl: We did. That was one of the characteristics. That's why I was asking, because I looked back over our results, I've tried to duplicate them in my own lab now. And the experiments as they are coming out for me now cause me considerable puzzle compared with our earlier work.

A: No, we all planned it I collected the polymerized product in xylene. And I hoped to analyze it before this meeting, but.... We hope to do it soon.

C: R. Dahl: But you mean, the polymeric products off the walls.

A: Off the walls, yes.

C: R. Dahl: If you dissolve that I think and I would be interested in the results, because when we tried this and this was I remember now in the sixties, I was astonished because I was trying to clean the target any way I could, and I had handy benzene. And the residue from the walls dissolved and gave me an orange solution but didn't come off completely. So well, what else is there to try ? And you're looking around, this and chloroform.... Chloroform gave me a magenta solution. And the other material was not very soluble. Ultimately we threw the target out. And it wasn't until the people at Rice University had their publication on the cover of the American Chemical Society's C&EN and it showed the orange and the purple solutions and they had identified this as the „bucky balls“. And I really think that's what you may be getting there.

C: V. Pike: We also tried to use this method about twenty years ago at Hammersmith. And we had to transport the N-13 ammonia from the target to the lab. We found that after a while, the lines filled up with an oily substance. When we carried out an infrared on this, just turned out to be about something compatible with engine oil. We were making oil all over the place.

A: This is why they are bubbling through the ammonia immediately below the target, and they are carrying away just the solution of ammonia. And if there is some oily stuff coming out of the target, it's mostly sticking on the glass wall there.

Q: K. Dowsett: Did you take any action to prevent methylamine getting into your water as well? Methylamine from methyl radical plus ammonia and it's very water soluble.

A: Yes, we tried it and we analyzed it by HPLC, and we couldn't find anything. We had just one sharp peak of ammonia in our final product.

C: J. Clark: Can I just make a couple of comments. You run an HPLC, you didn't show us the trace. You should do. And the second thing is, you get the price for using the biggest Swagelok fitting in the whole meeting.

A: I'll show the HPLC spectrum on the poster. - I wanted to use one size less, but unfortunately what was available from Swagelok was one inch or two inch. Nothing between that.





## Production of $^{123}\text{I}$ by Photonuclear Reactions on Xenon at Linac LUE-40 of FLNP JINR

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In recent years the method of irradiation of highly enriched  $^{124}\text{Xe}$  with up to 30 MeV protons is considered to be best method [1]. Irradiation of target with intense proton beam in medium-size cyclotrons, the absence of corrosion problems in the target system, the simple chemistry, the high radionuclide purity of the product ( $^{125}\text{I}$  content  $< 10^{-3} \%$ ) and the high yield all make this method the most suit for routine production of  $^{123}\text{I}$ .

Highly pure  $^{123}\text{I}$  can be obtained from  $^{124}\text{Xe}$  by another method - via the photonuclear reaction  $^{124}\text{Xe}(\gamma, n)^{123}\text{Xe} \rightarrow ^{123}\text{I}$ . Despite the relatively low efficiency of this method the linear electron accelerators and microtrons can compete with the proton cyclotrons (more expensive to operate) and meet the requirements for clinics and regions.

This possibility has been studied at a number of centers [2-4] but there is still no regular production of  $^{123}\text{I}$  in electron accelerators.

The experimental  $^{123}\text{I}$  yield was measured for  $^{124}\text{Xe}(\gamma, n)^{123}\text{Xe} \rightarrow ^{123}\text{I}$  reaction at linac LUE-40 of Frank Laboratory of Neutron Physics ( $E = 40 \text{ MeV}$ ,  $I = 60 \mu\text{A}$ ). The gas target with  $^{124}\text{Xe}(0.1\% ^{124}\text{Xe})$  (Fig.1) was irradiated by vertical bremsstrahlung beam ( $E_e = 35 \text{ MeV}$ ,  $I = 40 \mu\text{A}$ ) generated via Ta converter (3.5 mm). The  $^{123}\text{I}$  yield extrapolated from our experimentally determined data ( $5 \text{ g/cm}^2 \text{ natXe}$ ,  $\Delta t_{\text{irr}} = 2 \text{ h}$ ,  $\Delta t_{\text{decay-accum}} = 10 \text{ h}$ ,  $A \approx 1 \text{ mCi}$ ) amount to  $\sim 1 \text{ Ci}$  for  $> 99\%$  enriched  $^{124}\text{Xe}$  [5]. Consequently the following parameters of LUE-40 experiment ( $I_e = 40 \mu\text{A}$ , irradiation time 12 h) make it possible production rate of  $^{123}\text{I}$  equal about  $\sim 8 \text{ Ci EOB}$ .

### References:

- [1] V. Bechtold, H. Schweikert, KfK Nachrichten **21**: 13 (1989)
- [2] B. Nordell et al., Int. J. Appl. Radiat. Isot. **33**, 183 (1982)
- [3] I.K. Kikoin et al., J. Radioanalyt. Nucl. Chem., Lett. **103**: 27 (1986)
- [4] Y. T. Oganessjan et al., Atomnaja mergia **68**: 271 (1990)
- [5] V.A. Arhipov et al., JINR Communication P6-94-402, Dubna (1994)

### Discussion:

Q: T. Ruth: Was the 1 mCi result an experimental result?

A: Yes, experimental result. This is an experimental model. The special construction group will develop another working target.

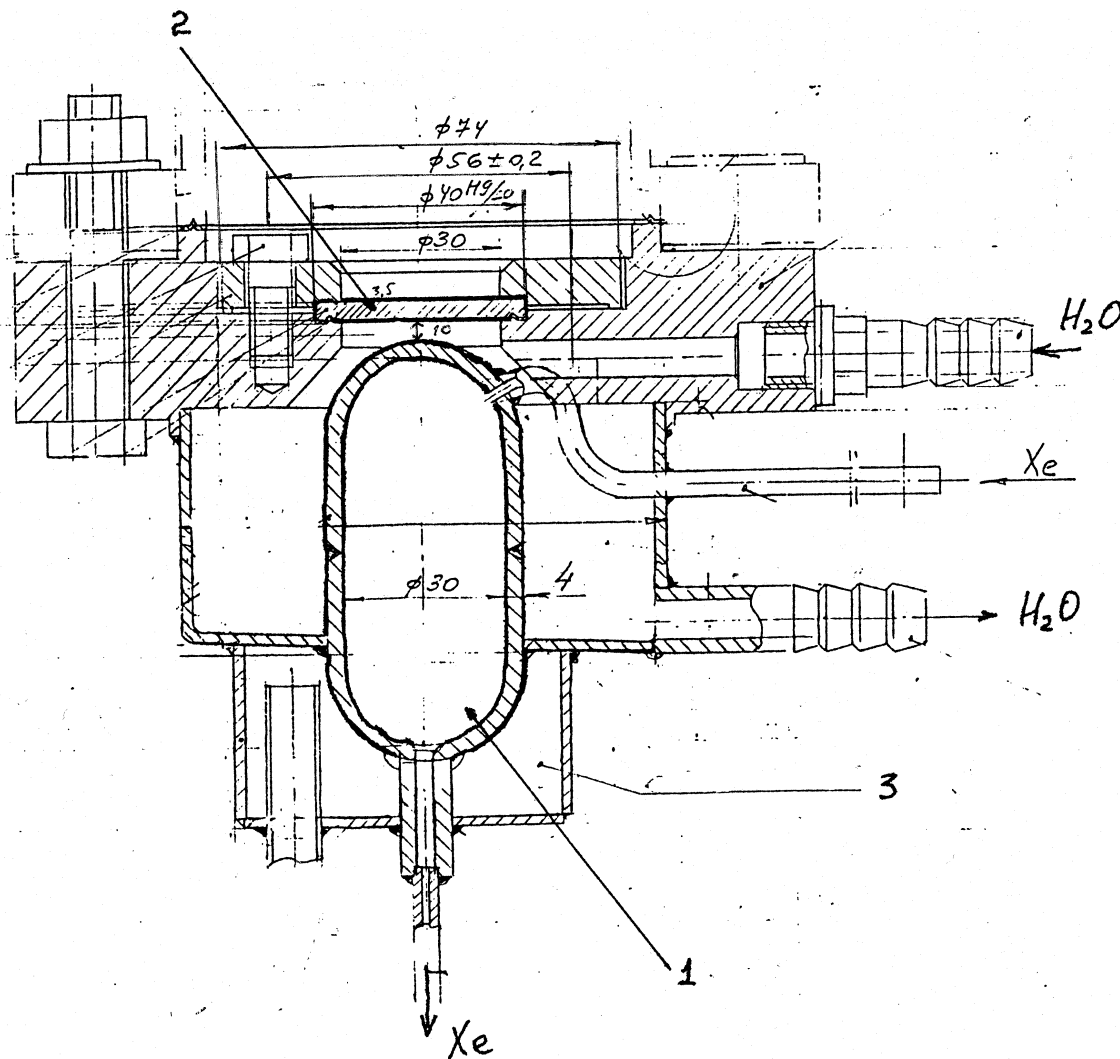


Fig. 1: Xe-gas target for  $^{123}\text{I}$  production.  
1) Target, 2) Ta-converter, 3)  $\text{N}_2$  cooler.

### Richard Ferrieri, BNL: $^{18}\text{O}$ Water Recovery and Purification

The first slide is just a picture of our resin recovery station. We process our target water in the back vault of our cyclotron, you'll see the reason for this in a second. The resin recovery station is a remotized unit that is located about three feet from the target. It involves emptying the water target through an anion exchange resin, it goes through a series of valves, here you can see the plumbing. Then we process that by rinsing the resin, we recover the water. We've rinsed the resin with carbonate and then deliver this carbonate charge to the hot lab through about 140 feet of transfer line. This is done nearly daily now in our hot lab. We have several transfer lines, these are comprised of about 1/32" o.d. Don't expect me to convert this to metric for you. It's polyethylene. This stands upright in a lead box, it's about 1.5 inch thick lead with a door. It allows us to go into the vault after an irradiation if we have to do any kind of target inspection or work on the target, which Dave and I frequently do. It does get a little toasty back there after we do a water target production run. We are using Lee valves as you can see here, these miniature valves. All the tubing in here for the most part is



polyethylene, it's got good radiation characteristics. The system's been in place since the Brookhaven workshop, six or seven years back. It had a small water sensor to tell us when the target had in fact unloaded. After I saw Ehrenkaufers little tiny bubble poster in Vancouver, this is one of these little ultrasonic sensors. It has since failed, it wasn't serviced for about eight months, but it sustained substantial radiation damage and it slowly deteriorated to the point that it's disfunctional now. These are just the two carbonate rinse tubes. We put in a 0.9 mL charge in the first rinse and a smaller volume in the second rinse, so that if we wanted to deliver a second charge to another hot cell of, say, 5 mCi of F-18 fluoride for any kind of research purposes we can.

This tube down here is our water recovery. And we actually will recover, the target volume, as Dave mentioned yesterday, is about 2.6 mL. The lines going to and from the target require us to fill with about a 3.5 mL charge for each irradiation. So it pays for us to recover this. We can reprocess our water. Mahmoud Firouzbakht just informed me this morning, he handles all our redistillation. And he has redistilled this water something like 32 times, without much deterioration of the isotopic enrichment of this water. What I wanted you to focus on is our little resin column here. We used to use these glass columns with the quartz glass frit in them and they had a Luer fitting on the bottom. And quite often from the pressure of the unloading, we tend to dry the system with about 30 pounds pressure of helium, to push the liquids through the resin column and deliver it to our lab. Quite often, we don't get delivery and after about half an hour Dave and I would suit up and go back into the lab and find this puddle of radioactive water or liquid on the bottom of our resin station due to the fact that the Luer fitting had popped off.

So we went to a Brownley HPLC guard column arrangement. And it allows us to take this column off with fingertight fittings and then we can load our resin column in there. Here is the guard column broken down. If any of you have ever used these things, they have like a teflon ring on each end and typically have a stainless 1/4 inch od guard column that can insert in. And as you screw this together it makes a nice gas/liquid-tight seal. The little hole drilled here, the only reason that is there, is to remind us to put this resin column in a certain orientation. Of course some of us aren't convinced of that, so we put X's all over the place to reinforce that action. The insert, we wanted something that was transparent, so that you can see the resin packing as you load it in and initially we used a 1/4 inch o.d. lucite tube. Of course we started off with a small bore lucite tube and packed it with resin. And interestingly enough, we were able to trap an extract of F-18. But the form of the F-18 was rendered inactive in terms of doing any subsequent synthesis. So we had to put an inert liner down this tube. You see here, it's a polyethylene sock, that's a polyethylene liner that's stuffed down. And you can see a hint of the resin that's packed in there. The resin is just a Dowex AG1X8 carbonate form resin, 200-400 micron. Nothing exotic about that.

The polyethylene sock is just 1/8 inch tubing, we use an Omnifit flaring tube just to create this little flare tip. And what I did is, I bore out the 1/4 inch lucite tube, the back end of that, so you can insert the sock. Once it's inserted we won't go replacing it. It stays in there, we've had these socks in these lucite tubes now for several years. What we do change, and this is important to the performance of the system, are these Alltech polyethylene frits. These are changed almost daily with the replacement of the resin. There is one frit that gets sandwiched into the back side of this column. You see here that the water target unload goes in this direction, and as this arrangement gets locked into the Brownley guard column. It's about 30 mg of resin, very small amount. That's one of the advantages of this, if you use any commercial packed resins, you're dealing with a lot of material. And this allows us, I think, and these resins have to be watered of course and the more of the resin you use, the faster the deterioration of your enrichment of the water you're going to have. So we have a very tiny amount, it's about 6 mL in the length down the column.



One of the things we've noticed is, by locating this whole apparatus back in the cyclotron vault, it allows us to retain some of the longer lived isotopes that come out of the target material, e. g. there is V-54, from our titanium window. We use a 2 mil thick titanium window on the target. And also Cd-109 coming from this silver target body. And it's interesting, some of that activity resides in the resin even though it's an anion exchange resin it has some capacity for metal ions. What's also interesting is that the polyethylene frit gets as hot as a pistol. So there is considerable retainment of the long-lived isotopes there. So in a sense this provides a way of processing our enriched water in the back vault and delivering the F-18 in a carbonate form to the hot lab. It also provides a mechanism for retaining the long-lived isotopes in a location away from the hot lab operation, where you really don't want to have those things accumulating.

#### Discussion:

Q: T. Ruth: Which volume of carbonate solution do you use to elute the fluoride in?

A: It's a 0.9 mL volume of 0.01 N. But we're using something different from that now. And it's usually a potassium carbonate. Although Sue Shin has an exotic synthesis that requires a cesium carbonate. We have the stock solutions back there. So that transport is quite nicely. Those lines I've described have been in place for years, and the only maintenance we do on them is the issue of cleaning systems with ethanol and everything. We don't go through any exotic cleaning. When we see transport slowing down, and... when the lines are cleaned the activity can come anywhere between three and five minutes to the back hot lab and it comes as a nice clean bolus. And when that starts to deteriorate, we start to see the transport times are maybe fifteen, twenty minutes, then it's time to wash the lines. And all I do is detach the line from the back vault, go into the hot lab, squirt through a couple of cc of distilled water and dry the line out with nitrogen. And the line is back into working very well.

I think one of the big issues about deterioration of transport line performance is this silver amalgam that comes out of the target. And these frits, the reason why they are changed with each run, they are grey to black. There is crap coming out of the target that's getting deposited in those transport lines. So you need a mechanism for getting that stripped out of your transport fluid. So that's another good reason for locating this resin recovery station or your processing station back in the vault where the target is, not in the hot cell, where the synthesis module is.

Q: K. Erdman: A couple of questions. First, why didn't you use just cross-linked polyethylene, I mean radiation hardened for that little container rather than going through that sock thing ?

A: Mainly because I didn't have any in stock at the time. Like I say, this has been working well. Yes, that's true, I probably could have made a research project of this and gotten grant support.

Q: K. Erdman : And the second question was, with that real small volume of resin you have in there, at what level of radiation in activity does that thing start to give you troubles?

A: Well, we've downloaded about a Ci of F-18 and actually have left it on the resin for some time. Half hours before we unloaded it to the hot lab. That's the nice thing also, we can do a water target irradiation first thing in the morning, leave it on the resin and then rinse it off and ship it to the hot lab, when they are ready to do synthesis. And we really haven't seen that much deterioration, although I have to admit, we really haven't done a stringent study on what goes on, what comes off, what's left behind, that sort of thing. So yes, there may be radiation damage involved there, but really for our operations that hasn't been a crucial issue.

C: We change the resin every day.

Ferrieri: Now one thing I should say, with the running of this resin. We've had on occasion a failed synthesis, and what do you do under those circumstances? Does somebody go back into the vault and replace that resin? What we've actually done is, we had that second rinse there. We actually have rerun the target and then processed it through the same resin and retained enough F-18 back on that resin that we can strip off with the second carbonate rinse. So, there is enough resilience in the resin to keep back things. But we tend to do this on a per run basis. If the vault is not hot, we change the resin and the frit, pack it in.

Q: K. Dowsett: Can you say a little bit about how you recycle your water? How you avoid trace organics coming back round and forming crap in the target for example?

A: Mahmoud Firouzbakht may want to comment on that. We have a Micro-still. He's got a methodology there.

C: M. Firouzbakht: When we collect, we have about 50 mL of water. And for each run we use about 3.5 mL. And we collect all the water and we distill it and collect about 45 or 50 mL. When we compare that water with the fresh water, it's almost the same. That means the isotopic purity doesn't go down that much.

Q: No trace of amines there?

A: M. Firouzbakht: No.

Q: J. Nickles: Why don't you elute the column with crypto-carbonated 80/20 acetonitrile water? I mean, have it ready for the standard fluorination procedures?

A: Yes, that's been entertained, no, we haven't looked into it though.

C: It works well.

A: Oh yes, I heard good things about that mixture.

C: J. Clark, Hammersmith: Bruno Nebeling isn't in the audience, but if anybody wants a very good GMP for O-18 water, contact Bruno. He's got all analytical techniques to show how good the water is when you've gone through this procedure. Or, if it hasn't worked out, don't use it. Actually recycle again.

### **Günter Firnau, Hamilton: Ion Exchange Columns and Cartridges**

<sup>18</sup>F-Fluoride that was produced in a silver-body target requires purification before it is used in nucleophilic substitution reactions. After bombardment of <sup>18</sup>O-water we pass the approximately 500 µL <sup>18</sup>O-water/<sup>18</sup>F<sup>-</sup> with 20 psi pressure through a microcolumn filled with the anion exchange resin AG-1-X8, 200 mesh in the bicarbonate form. The column is prepared from 1/16 inch O.D. polyethylene tubing size with an internal diameter of 1/32 inch. The resin bed in the tubing is 1.5 inch long. The resin is held in place by a 10 µm frit in a flanchless fitting (both from Upchurch). The column contains between 5 and 10 mg of moist resin. The column is home made and prepared fresh for each use. The retention of <sup>18</sup>F is nearly quantitative with this column. After elution with acetonitrile/water/KHCO<sub>3</sub>/cryptofix, only 1-2 % of <sup>18</sup>F remain on the column.

In our experience, it is important to have a small diameter column. When using the Waters QMA light cartridges (130 mg of resin), the results were not entirely positive. Although QMA cartridges work, there have been too many occasions when up to 50 % of the total  $^{18}\text{F}$  remained on the column after elution. Other commercially available cartridges (Varian or BioRad) contain up to 1 g of resin. They are expected to give more varying results. The manufacturers of automated FDG synthesis modules may want to invest some R&D into small cartridges with resin load of no more than 50 mg and a frit more porous than 10  $\mu\text{m}$ .

The use of a small resin column has another advantage in that the isotopic dilution of  $^{18}\text{O}$ -water can be kept minimal. This is important when the collected  $^{18}\text{O}$ -water is to be reused. In our hands the  $^{18}\text{O}$  enrichment drops from 97 % to only 88 %.

The other comment I would like to make refers to the colloidal silver that may emerge from a silver target when it is run at high currents (30-40  $\mu\text{A}$ ). The microcolumn also acts as a mechanical filter and retains the colloidal silver as a dark plug on top of the column.

C: J. Link: There is a company, I haven't tried it, I've ordered them. I use almost the exact size as you do and I've been doing homemade and trying to find them. There is a flow injection analysis company in Washington that sells and they're 10/32 fittings, they're upchurch fittings on the top and the bottom. It's almost the exact size that you have. There is an insert that fits inside. It's conical, it's not straight through. They will pack them themselves or it comes empty. They are 1 or 2 cm high and they are made for doing ion exchange, resin separations with process systems for flow injection analysis. So it's out there. And it's 10 micron frit.

A: By the way, this 10 micron filter here is an upchurch filter. It's called A423.

Q: T. McCarthy: I just wondered if you have tried to reduce the amount of cryptofix that you are using in your elution solvent, because that's considerably more than we are used to be using ? I think you'll find, that you can reduce that down a lot.

C: J.-L. Morelle: I just wanted to say something about the cartridges you display here. It's actually a repackaging of a product which I think existed some years ago, which is teflon based membranes, such as were used also in Karlsruhe. It's a Varian product repackaged by 3M. And they are small columns and you can't really notice, because here in the projector you mainly have the ring which you see, here is mainly the clamping ring, but it's 0.5 mm thick membrane, which is packed in a usual little cartridge. And we use it in our new system and we obtained more than 99 % recovery with it. The only problem we face, it's an experimental product, at the moment it's difficult to have it supplied. We had a few bags provided to us by 3M. It gives good yields, the only problem is that the clamping ring is not very tight on the cartridge inner surface and that in some cases part of the frit bypassed the membrane. So it's mainly a matter of getting through large production. I think it's useful that we put the pressure on them to convince them to put the product on the market.

Q: P. Larsen: I would like to know if anybody has tried to use this purification technique together with the FDG microlab ? Is it possible to control this very small amount of carbonate so you don't overload the column in the FDG microlab ?

C: J. Clark: Have you talked to Johan Ulin recently ? I think in Uppsala a refined version of the GE microlab will be shown where the original Mulholland resin is replaced by carbonate resin.





C: K. Erdman: People were asking if there is a little column. We make a little column now which is on our little unit out, you can have a look at it. It's roughly the size that he was talking about. Which we will sell commercially.

Q: G. Firnau: Is your column on the open market or only available for Ebco users?

A: No, we will sell it on the open market.

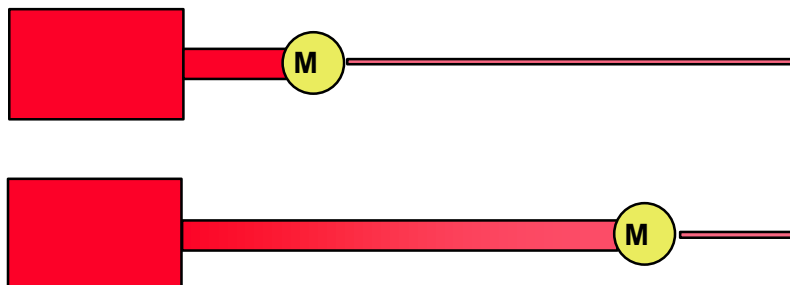
Q: M. Brüssermann: Same with us, we have a column on the Nuclear Interphase module which is as small as the first column you show there. The first column you made yourself. Also 20 mg, and we use a glass frit at the end, about 100 micron. It is available, it is working fine. We also tried to use the QMA Waters. It works sometimes fine as you said, sometimes not. We also asked producers of ion exchange columns and they said they will not make it, because it's too expensive to make such small columns.

Q: P. Mikecz: When I had protons in our CS-30 machine I used to use only fit columns. It's not ready packed, but you have a 3 mm diameter, 25 mm long columns, which again uses very tiny amounts of ion exchanger. It's very easy to fill and handle. Others, like Biorad had a few years ago an ion exchange membrane, which you could use like Jean-Luc's systems. Unfortunately Biorad stopped the production of this membrane.

### Richard Hichwa: Dimensioning of Transport Lines

**Problem:** How to deliver maximum activity within optimal volume/pressure conditions at the output.

**Standard Solutions:**



**Alternative Solutions:**

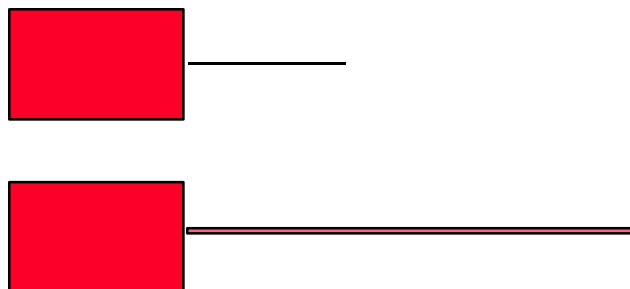
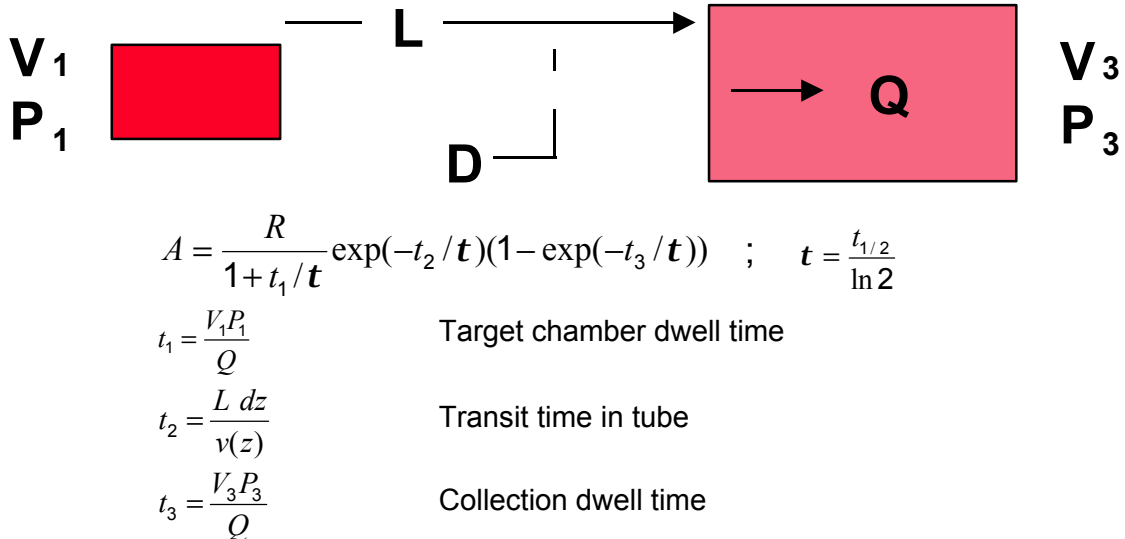


Fig. 1

Some years ago Jerry Nickles and I worked on an efficient gas transport model. The initial problem is simply one, namely how to deliver the maximum activity in gaseous form to an

optimal volume and a desired pressure at the output end of a gas delivery tube? A suboptimal system is shown in the upper Figure 1 in which a metering valve regulates flow.. High concentrations of radioactivity appear darker while lower concentrations are lighter. If the metering valve is moved closer to the output end, then large quantities of radioactivity are resident in the delivery tube and the concentration varies significantly along the tube length. An alternative solution is to eliminate all valves and tune the system to deliver the radioactivity at a prescribed flow rate and concentration so that high qualities of radioactivity are delivered at a useful output pressure (typically low pressure) as shown in the lower panel of Figure 1.

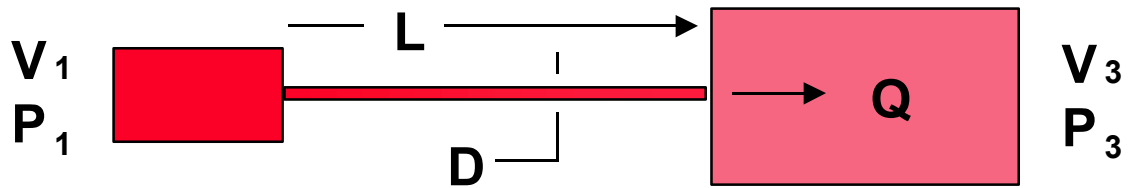


**Fig. 2**

To that end, one can model the system as shown in Figure 2 with a series of volumes at various pressures.  $V_1$  corresponds to a cyclotron target which operates at pressure  $P_1$ . In reality any volume of interest which is connected to the input of the delivery line (length  $L$  and diameter  $D$ ) can be modeled. The output quantity  $Q$  is defined as the mass flow rate from the tube.  $V_3$  is highly variable and can be a vial or container of fixed size, a vessel containing absorbers, or even human lungs (time varying volume). The equations shown here describe the optimal activity ( $A$ ), target chamber dwell time ( $t_1$ ), tube transit time ( $t_2$ ) and collection chamber dwell time ( $t_3$ ).  $R$  is the radioactivity production rate and is related to beam current, target pressure, beam area, reaction cross section, and beam energy.  $t_{1/2}$  is the nuclide half-life.

The delivery tube length ( $L$ ), target volume ( $V_1$ ) and target pressure ( $P_1$ ) are constrained to some extent by the cyclotron production parameters. Target chambers are typically operated at pressures that have been optimized for specific nuclide production. Only minor modification of parameters are really possible. The delivery tube diameter ( $D$ ) is fully adjustable in the equations shown in Figure 2. In the process of optimizing flow, all valves, flow controllers and constrictions are removed from the system in order to permit realistic characterization of the system impedance. The lower panel of Figure 3 succinctly lists the criteria for the optimization process. The first criterion is to minimize the time ( $t_1$ ) radioactivity spends in the target. This is achieved by making the volume of the target chamber as small as possible. Secondly, one tries to match the time spent in transport through the delivery tube ( $t_2$ ) to the collection dwell time ( $t_3$ ) and make both approximately equal to the mean life of the nuclide of interest. Minimizing  $L$  is desirable, but may be impractical. Overall activity collection is increased by maximizing  $V_3$  which can be achieved through efficient tapping mechanisms. A reference to this work appears at the bottom of Figure 3.





$L$ ,  $V_1$  and  $P_1$  are dictated by experimental setup.  
However,  $V_1$ ,  $P_1$  and  $D$  can be adjusted for optimal conditions. No metering valves, flow controllers, etc to impede flow.  $L$  and  $D$  govern system impedance.

**Bottom Line:**

- 1) Minimize  $t_1$  by reducing  $V_1$
- 2) Attempt to match  $t_2 = t_3$  to  $t$
- 3) Minimize  $L$
- 4) Trap at output to increase  $V_3$

**Reference:**

R.D. Hichwa, R.J. Nickles: The tuned pipeline-A link between small accelerators and nuclear medical needs. IEEE Trans Nucl Sci NS-26: 1707-1709, 1979

**Fig. 3**

## The Rossendorf Radionuclide Transport System for Gases and Liquids

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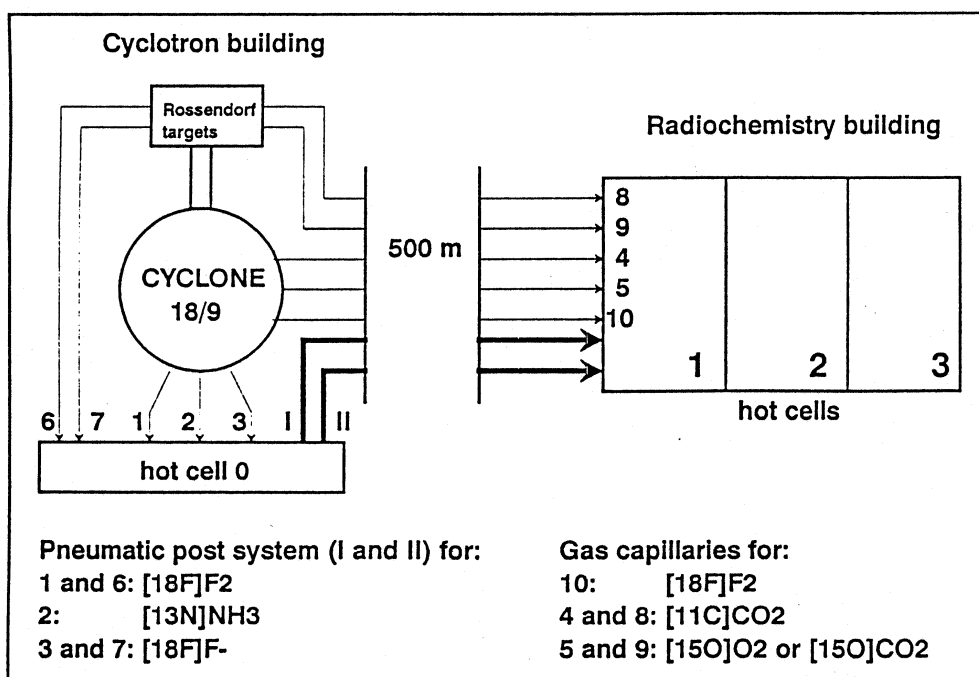
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On grounds of special local conditions the Rossendorf PET cyclotron "CYCLONE 18/9" is located 500 m far from the radiochemical laboratories [1]. The radionuclide transport system (RATS) was designed to transport small volumes of the irradiated liquids  $[^{18}\text{O}]\text{H}_2\text{O}/[^{18}\text{F}]\text{F}^-$  and  $\text{H}_2\text{O}/[^{13}\text{N}]\text{NH}_3$  with a pneumatic post system and radioactive gases within copper tubes from the PET cyclotron to the radiochemistry laboratories [2]. Since the end of September 1996 the cyclotron/RATS complex works with high reliability.

### Technical Parameters

The general layout of RATS is shown in Fig. 1.



**Fig. 1:** General Layout of RATS

#### cyclotron building:

Hot cell 0 contains:

- loading unit for the pneumatic post boxes
- Rossendorf distribution unit for further handling and transfer of  $[^{18}\text{F}]\text{F}^-$ ,  $[^{18}\text{F}]\text{F}_2$  and  $[^{13}\text{N}]\text{NH}_3$

#### radiochemistry building:

Hot cell 1 contains:

- unloading unit for the pneumatic post boxes
- IBA hot gas distribution unit for transfer of the radionuclides into the hot cells 2 and 3
- IBA  $[^{11}\text{C}]\text{HCN}$  module and  $[^{11}\text{C}]\text{CO}_2$  trap
- Rossendorf distribution unit for transfer of the radionuclides into the hot cells 2-5 and 11-14

### **Pneumatic post system**

The pneumatic post system consists of 2 polyethylene tubes with 33 mm inner diameter and 8.5 mm wall thickness. The pneumatic post box is made of aluminum (length = 110 mm) and contains a 10 mL vial for transport of e.g. 1.5 mL of activated liquid. With a pressure of 4 - 5 bar of compressed air the pneumatic post box is pushed through the polyethylene tube and a transfer time of 1.30 minutes is reached. The whole transfer process (unload the target into the vial, transport, unload the vial into the module) takes about 10 minutes.

### **Gas transport system**

The gas transport system consists of 6 copper tubes with 1.5 mm inner diameter [3] and 0.75 mm wall thickness. For [ $^{11}\text{C}$ ]CO<sub>2</sub> we reach an overall transfer time (EOB to BOS) of 4.30 minutes with 28 bar of pushing gas.

### **Control System**

RATS can be controlled both from the cyclotron site and from radiochemistry site. We use the MASTER-SLAVE principle: Access to control has only the master terminal whereas all information is displayed on both monitors. The use of PLC Simatic S5 system makes data transfer with cyclotron's control system very easy. The coupling of PLC and PC allows the control of the processes via the graphic interface by mouse click. All necessary information is given on screens (e.g. position of distribution valves, activity level, status of RATS).

### **Safety Aspects**

Both, cyclotron's and RATS' control systems are coupled by four interlock signals (kind of target, beam on/off, CYCLONE 18/9 and RATS ready/not ready to unload the target) to prevent the unloading procedure of a target if the cyclotron or RATS are not in the right status. In case of water target a laser sensor gives special information about the status of pneumatic post box to the control unit to prevent the unloading without a vial in the pneumatic post box.

In the poster details of RATS (e.g. layout, control system, radiation protection, interlock system) and our experiences of operation are given.

### **References:**

- [1] St. Preusche et al., The New Cyclotron of the Rossendorf PET Center, in: Cyclotrons and their Applications, World Scientific Publishing C. Pte. Ltd. (1996)
- [2] St. Preusche et al., The Radionuclide Transport System of the Rossendorf PET Center, XXX European Cyclotron Progress Meeting, Catania, Italy, Sept. 04-06 (1996)
- [3] O. Solin, National PET-Center Turku, Finland, private communication

## Long Distance Transport of [ $^{18}\text{F}$ ] $\text{F}_2$

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Electrophilic  $^{18}\text{F}$  is usually produced by the  $^{20}\text{Ne}(\text{d},\alpha)^{18}\text{F}$  reaction using elemental fluorine as a carrier gas. Among the factors which must be taken into account to achieve satisfactory activity yield of [ $^{18}\text{F}$ ] $\text{F}_2$  in routine production [1], as for instance target design, target gas, target gas handling and target operating conditions, the transport of the target gas after end of bombardment to the radiochemical laboratory plays also an importance role. The main factor that determines [ $^{18}\text{F}$ ] $\text{F}_2$  recovery is the chemical state of the target and the surface of the transport tubing. The transport of [ $^{18}\text{F}$ ] $\text{F}_2$  becomes an important problem, whenever the distance between the cyclotron and the radiochemical laboratory is so long that the surface of the transporting tube exceeds the target surface significantly. PTFE tubing has a good chemical resistance against  $\text{F}_2$  but is unsuitable as it becomes permeable with use [2]. At present stainless steel lines are recommended for transport, but the yield of [ $^{18}\text{F}$ ] $\text{F}_2$  activity in case of long distance transport is not sufficient. A good preparation of the whole target system is achieved when 75 to 80 % of [ $^{18}\text{F}$ ] $\text{F}_2$  can be recovered in the production module. But it is difficult to determine the exact activity yield of the target itself. The recovery of [ $^{18}\text{F}$ ] $\text{F}_2$  is not a constant value but strongly depends on the regime of use of the target and the transport facility.

The Rossendorf PET-Center has the feature of a 500 meter distance between the cyclotron and the radiochemical laboratories and therefore the problem to connect them. This is done by a special radioactivity transport system.

The [ $^{18}\text{F}$ ] $\text{F}_2$  is produced by the  $^{20}\text{Ne}(\text{d},\alpha)^{18}\text{F}$  reaction using the 9 MeV deuterons of the cyclotron CYCLONE 18/9 with  $\text{Ne} + 0.2\% \text{F}_2$  (12 bar, about 60  $\mu\text{mol F}_2$ , prepared from a stock fluorine mixtures of 2 % fluorine in neon) as target gas. The target volume is 50 mL. After EOB the [ $^{18}\text{F}$ ] $\text{F}_2$ -target gas is forced by its own pressure (12 bar at the beginning) through the 500 m transport line. After being discharged from the target by its own pressure, the [ $^{18}\text{F}$ ] $\text{F}_2$  is transported to the synthesis unit using  $\text{N}_2$  as a carrier gas with a pressure of 5 bar.

We have tested the suitability of two kinds of transport tubes in relation to radioactivity and chemical  $\text{F}_2$  - recovery:

1. polyethylene tube:  $d_i = 1.3\text{ mm}$ ;  $d_o = 1.8\text{ mm}$
2. copper tube:  $d_i = 1.5\text{ mm}$ ;  $d_o = 3.0\text{ mm}$

Before transporting [ $^{18}\text{F}$ ] $\text{F}_2$  the tubes must be cleaned carefully. The polyethylene tube was flushed with an alcoholic solution (70 %) to clean, blown through with a  $\text{N}_2$ -gas flow to dry and prefluorinated with the stock fluorine mixture.

The copper tube was cleaned and passivated by the following procedure.

1. flushing with 400 mL heptane
2. flushing with 200 mL acetone
3. flushing with 200 mL water
4. flushing with 200 mL about 5 M  $\text{HNO}_3$
5. flushing with about 3000 mL water up to neutrality
6. flushing with 200 mL p.a. acetone
7. flushing with  $\text{N}_2$  with 10 L/h for 24 h

8. passivation with fluorine by flushing with the stock fluorine mixture up to break through of fluorine (color change of a potassium iodide solution).  
The liquid-flushing was carried out by using a high-pressure pump (HPLC).

For routine operation the whole target system is daily passivated (prefluorination) by pre-irradiation (20 min, 10  $\mu$ A) and discharging the target gas through the transport tube. If the target and transport line has not been in use for a longer time it is required to intensify the passivation by repeated prefluorination with the stock fluorine mixture. Average recovery for  $^{18}\text{F}$  and  $\text{F}_2$  for the transporting process

	$^{18}\text{F}$	$\text{F}_2$
polyethylene tube	40-45 %	20-25 %
copper tube	75-80 %	90-95 %

### Conclusion

- Polyethylene tubes are not suitable for  $[^{18}\text{F}]\text{F}_2$  transport.
- Well prepared and passivated copper lines enable a sufficient recovery of  $[^{18}\text{F}]\text{F}_2$ .

At our PET center we have a 550 m copper tube in routine use. The specific radioactivity of the  $[^{18}\text{F}]\text{F}_2$  in the radiochemical laboratory was about 80 GBq/mmol.

### References:

- [1] G. Stöcklin and V.W. Pike, Radiopharmaceuticals for PKT. Kluwer Academic Publisher, Dordrecht, Boston, London (1993)  
[2] V.R. Casella, A.P. Wolf, J.S. Fowler, R.R. MacGregor and T.J. Ruth, J. Nucl. Med. **21**: 750 (1980)

## Diffusion Based Separation Methods in Production of Non-Conventional Positron Emitters with Low Energy Cyclotrons

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### Introduction

The PET-technique is mainly associated with the use of a few short-lived positron emitters like  $^{11}\text{C}$ ,  $^{15}\text{O}$  and  $^{18}\text{F}$ . However, there is an increasing interest to extend this number for several reasons. Longer half-lives can extend the kinetic time window of PET, labeling techniques using chelators can be applied, and physiological studies of several essential elements may be possible. We here present an on-going program for the development of simple production methods of such nuclides by the use of low energy cyclotrons.

A pre-requisite for introducing new nuclides in this area is that they should be possible to produce in useful quantities (for one or two patient studies) with accelerators available at PET-centers. The production cost of these nuclides (cost for target material, equipment and personnel) should be of the same order as the production cost of commonly used radionuclides like  $^{18}\text{F}$ .

In order to produce these nuclides with high radionuclidic purity, enriched target material often has to be used. Proton energy in the range of 12-16 MeV (typical for the PET-center cyclotrons) means fairly thin targets (200-300  $\mu\text{m}$ ) but the target cost may still be high. Low scale production and low production cost therefore necessitate re-use of enriched target material. Standard wet separation techniques are not practical, because losses in the recovery of expensive enriched target material will be large. In this context, thermal diffusion-based radionuclide separation methods seem to be more attractive. Produced nuclides are rapidly removed from the bulk of target, expensive enriched targets can be re-used several times without working-up and the technique allows a high degree of automation.

### Theoretical and Practical Considerations

Heat diffusion of the produced radionuclide plays an important role in a number of radiochemical separation methods such as ISOL, dry distillation, and surface etching. All these methods run close to the melting point of the target, when diffusion coefficients in solid state have the same order of magnitude as in liquid.

In an irradiated target the produced radionuclides can be looked on as being dissolved in the target material. When the boiling temperature of the produced radionuclide is low the radioactivity will be distilled when the target is heated close to the melting temperature. This is typical for the production of the halogenes  $^{75}\text{Br}$  [1,2],  $^{123}\text{I}$  [3] and  $^{211}\text{At}$  [4] but it is also applicable in other systems like in the separation of IIB group elements from target metals belonging to the IB group of the periodic system [5]. The surface etching technique seems to be applicable when produced nuclide has lower melting point than target material, but high boiling point. In this case the solved radionuclides tend to concentrate on the target surface during heating but do not evaporate. In many cases it is then possible to remove the produced nuclide from the surface by rinsing with very diluted acid, as it has been done with

$^{110}\text{In}$  and  $^{66,68}\text{Ga}$  [6,7]. The losses of target material under such conditions are in the order of one per cent or less.

The following criteria can be stated for a successful application of the dry distillation or etching techniques:

- The target material should not undergo sublimation at working temperature.
- The melting point of the produced element should be lower than that of the target material.
- If the boiling temperature of the produced element is high compared with the working temperature then the etching technique may be applied.
- If the boiling temperature of the produced element is low compared with the working temperature then dry distillation may be applied.

## Results and Discussion

Within our program, diffusion based separation methods were successfully applied to production of radionuclides given in table 1. As is seen the separation temperatures has been well below 1200 °C which enable us to use simple and cheap oven in the separation setup.

**Table 1:** Production of non-conventional positron emitting nuclides with (p,n) or (p,2n) reactions.

Target	Nuclide	$T_{1/2}$	$\beta^+$ (%)	Separation method	Typical temp (°C)
$^{66}\text{Zn}$	$^{66}\text{Ga}$	9.5 h	57	etching technique	400
$^{68}\text{Zn}$	$^{68}\text{Ga}$	68 min	89		
Cu	$^{62}\text{Zn} (^{62}\text{Cu})$	9.3 h	8 (97)	dry distillation	1060
$^{70}\text{Ge}$	$^{70}\text{As}$	53 min	80	dry distillation	1105
$^{72}\text{Ge}$	$^{72}\text{As}$	26 h	88		
$^{76}\text{Se}$	$^{76}\text{Br}$	16 h	54	dry distillation	1100
$^{110}\text{Cd}$	$^{110}\text{In}$	69 min	62	etching technique	305

Diffusion rate of the produced nuclide is one of limiting factors. By increasing the temperature a number of other systems may from theoretical considerations be candidates for thermal diffusion separation techniques. A wide search for the optimal conditions for different material is necessary.

The thickness of the target foil affects the separation rate [8]. A thickness of 200-300  $\mu\text{m}$  may be too much in order to obtain an appropriate separation yield within a reasonable separation time. This problem can be solved by the use of a slanted target which gives an optimal energy degradation (due to the angle to the beam) with a physical thickness of 100  $\mu\text{m}$  or less.

## Conclusion

The thermal diffusion/etching and the dry distillation methods do not require expensive equipment and are easy to perform. They can be applied using relatively low-energy cyclotrons available at PET-centers. The use of the (p,n)-nuclear reactions enables production of useful amounts of desired nuclides with the use of isotopically enriched target material. Recovery of enriched isotopes and recycling of targets can be avoided due to low target losses during separation.

## References:

- [1] Z. Kovacs et al., Int. J. Appl. Radiat. Isot. **36**: 625 (1985)





- [2] W. Vaalburg et al., Int. J. Appl. Radiat. Isot. **36**: 961 (1985)
- [3] J.F.W. Tertoolen et al., J. Lab. Comp. Radiopharm. **13**: 232 (1977)
- [4] R.M. Lambrecht and S. Mirzadeh, Int. J. Appl. Radiat. Isot. **36**: 443 (1985)
- [5] V. Tolmachev, H. Lundqvist and L. Einarsson, Diffusion-based separation methods: dry distillation of zinc, cadmium and mercury isotopes from irradiated targets, Int. J. Appl. Radiat. Isot., in press
- [6] H. Lundqvist et al., Appl. Radiat. Isot. **46**: 859 (1995)
- [7] V. Tolmachev and H. Lundqvist, Appl. Radiat. Isot. **47**: 297 (1996)
- [8] G.J. Beyer et al., IAEA report INDC(NDS)-195/GZ, 77 (1988)

#### Discussion:

Q: J. Nickles: Do you have any intention of looking at the rhodium/palladium system? To get  $^{103}\text{Pd}$  out of rhodium spheres for example ?

A: No, we didn't check. The problem with that period is, it's pretty high melting points, it's about 2000. It's too high. The system might be too expensive. The only hope is in the future, that you can have some transition between allotropic modification. And since the system is very unstable you might have diffusion in this situation, even below the melting point. But I didn't check, it was just a calculation, just an idea.

C: T. Tewson: I had a silver target to make  $^{18}\text{F}$ , where the times between cleanings were getting shorter and shorter. And I tried to anneal it, to see if it would improve its lifetime and all the  $^{109}\text{Cd}$  distilled out very nicely from the silver target and condensed on the furnace walls. So there is a nicely contaminated furnace. So don't try, the reports were just phenomenal here, I just want to warn you, don't try and anneal your silver target.



## Introduction to Session IVb: Radionuclides, Target, Processing and Automation

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The papers were divided into three subjects; developments in solid targets for the production of longer-lived radionuclides for diagnostics and therapy; targets for new radionuclides; including targets for the production of generator nuclides; and automation in targetry and chemistry systems.

G. Blessing et al. from Jülich described four improved target systems, three solid targets and one gas target for irradiating highly enriched materials at high current without losing the target material. Their "pellet target" and "Disk Target" used water cooling successfully on targets with poor heat transfer. Their Wedged Targets utilized a shallow angle of 20° for the beam strike and irradiated molten or electroplated metallic elements on copper backing. A conical gas target used steam for removal of the Sr and Rb from the target. The systems for processing the targets were also described.

W.Z. Gelbart et al. from TRIUMF presented two targets. One paper described a target for production of radioiodine from tellurium oxide. The TRIUMF group used an existing solid target with a grooved Ag face onto which the  $\text{TeO}_2$  was melted. The target was irradiated with 150  $\mu\text{A}$  without loss of radioiodine. The second paper described a solid "encapsulated" target. The unique aspect of this target was a concentric cooling method that created a jet of water. Calculations of heat transfer and cooling in this target were presented and generated quite a bit of discussion with regard to the limits and advantages of these calculations.

The research group from Dubna also had two contributions to the session. The first by S.N. Dmitriev et al. who presented their procedures for production of ultrapure  $^{237}\text{Pu}$ ,  $^{236}\text{Pu}$  and  $^{97}\text{Ru}$ . The very pure products should be useful for biomedical and environmental studies. The second work by O.D. Maslov et al. described the target used by the Dubna group for production of the high-purity  $^{236}\text{Pu}$ .

F. Nortier and his colleagues at the National Accelerator Center in South Africa use 66 MeV protons to produce  $^{139}\text{Ce}$  from Praseodymium metal in a capsule target surrounded by water. The authors presented the excitation function for this reaction for proton energy up to 100 MeV, they calculated thick target yield from the cross section and showed that the measured yield of 0.37 to 0.39 mCi/ $\mu\text{Ah}$  for 16 to 1807  $\mu\text{Ah}$  of irradiation agreed well with the 0.37 mCi/ $\mu\text{Ah}$  calculated from the excitation functions.

R. Shefer from Newton Scientific, Inc. and colleagues at Washington University in St. Louis, presented their target system for production of  $^{64}\text{Cu}$  from nickel and the automated remote system for removal of the target from the accelerator and recovery of the  $^{64}\text{Cu}$  from the nickel. These researchers are also investigating production of six other medically useful radionuclides using this system. D. McCarthy from this same group of collaborators presented the production parameters for  $^{60}\text{Cu}$ ,  $^{61}\text{Cu}$ , and  $^{64}\text{Cu}$  using  $^{60}\text{Ni}$ ,  $^{61}\text{Ni}$  and  $^{64}\text{Ni}$  using this target system and 15 MeV protons or 8 MeV deuterons. In a different solid target, T. McCarthy from St. Louis described his work with a commercially available inductive heater to develop a portable device to remove  $^{13}\text{N}$  from graphite targets.

S. Smith and the investigators at ANSTO (Australia) also described production of  $^{64}\text{Cu}$ . These investigators use protons to produce  $^{67}\text{Ga}$  from  $^{68}\text{Zn}$  on a copper backed nickel target. Several other radionuclides are made in this target including a significant amount of  $^{64}\text{Cu}$ , presumably from the Ni in the target. The authors described their chemical methods to take advantage of the extra production reaction and obtain 2 to 5 Ci of  $^{64}\text{Cu}$  with specific activity of 700 to 15,400 mCi/mg from the  $^{67}\text{Ga}$  "waste". The challenge is to avoid contaminating the  $^{64}\text{Cu}$  with nonradioactive Cu from the target backing.

D. Phillips and colleagues at Los Alamos National Laboratory (LANL) and B. Zhuikov and colleagues from the Institute for Nuclear Research (INR) in Moscow described two collaborative targetry efforts. The first was the production of  $^{72}\text{Se}$  for an  $^{72}\text{As}$  generator. LANL uses wet chemistry after 800 MeV proton spallation on RbBr to obtain 0.005 mCi/ $\mu\text{Ah}$  EOB of  $^{72}\text{Se}$  with a  $^{72}\text{Se}/^{75}\text{Se}$  ratio of 1.0. The Troitsk (INR) group uses the (p,4n) reaction with 60-45 MeV protons on As in a GaAs target. Gas-chemical processing yields 0.5 mCi/ $\mu\text{Ah}$  EOB of the  $^{72}\text{Se}$  with a  $^{72}\text{Se}/^{75}\text{Se}$  ratio of 1.5. The second work presented by these authors was the production and recovery of  $^{109}\text{Cd}$  from indium. INR uses 140-80 MeV protons and has a 0.3 mCi/ $\mu\text{Ah}$  EOB production yield. LANL uses 800-600 MeV protons to yield 0.005 mCi/ $\mu\text{Ah}$  EOB of the  $^{109}\text{Cd}$ . A high temperature gas-chemical processing method (thermochromatography) is under investigation by these researchers as a more efficient lower waste solution to current wet-chemistry  $^{109}\text{Cd}$  purification techniques.

The use of separation techniques other than wet chemistry was also described by V. Tolmachev from the Kurchatov Institute who has been working with researchers in Uppsala, Sweden. He described the use of thermal diffusion as a method to bring a radionuclide with a melting point lower than that of the target material out of a target. The product radionuclide either distills out of the target, a technique that many use, or if it has a higher boiling temperature than the target, migrates near the surface of the target, where it can be etched off. This is a technique that has not been commonly used. The practical advantage of the technique is that expensive target materials can be conserved and reused. These same researchers also described the routine production of  $^{76}\text{Br}$  using 17 MeV protons on  $\text{Cu}_2^{76}\text{Se}$ . The dry distillation technique gave 150-300 MBq of activity 1-1.5 hrs EOB with a separation yield of 65-75 %. The target  $\text{Cu}_2^{76}\text{Se}$  pellets could be reused as many as 20 times before they needed to be redone.

P. Goethals and colleagues from Gent, Belgium described the production of  $^{55}\text{Co}$  as a tracer. The reaction used was

$\text{Fe}(p,2n)^{55}\text{Co}$ . The authors described their purification methods and had a 0.3 GBq yield of the cobalt.

## Improved Target Systems for Irradiation of Solids and Gases at the Compact Cyclotron in Jülich

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Several improvements have been achieved in the target systems available at the compact cyclotron CV28 in Jülich. The basic aim behind these was to irradiate highly enriched target materials at high currents with the minimum loss of the material. Efficient irradiation facilities were developed for amorphous materials in the form of pellets, foils and disks of metals and alloys, thin layers of electroplated materials on a wedged target holder, and for inerted highly enriched gases like  $^{82}\text{Kr}$ ,  $^{83}\text{Kr}$ , etc. The first three facilities are available on a common target holder. The latter constitute an extra irradiation and processing unit. We consider them individually.

### **Pellets of amorphous material**

One problem in the use of metal-oxide pellets (for example  $^{92,94}\text{MoO}_3$ ,  $^{147,144}\text{Sm}_2\text{O}_3$ ,  $\text{As}_2\text{O}_3$ ) is the removal and transfer of the fragile pellet from the pressing-tool to the target head without damage. We overcame this difficulty by introducing a steady counter pressure while removing the pellet. With this device we were able to use almost every produced pellet for production purposes.

The second problem encountered while using metal oxides as target material is the bad heat conduction, resulting in a high danger of evaporation of the target material during the irradiation. The two commonly utilized targets at Jülich, i.e. the  $4\pi$ -water cooled and the  $2\pi$ -water cooled systems, could often not be used advantageously. We modified the known  $2\pi$ -water cooled system to enhance the cooling of the pellet from the back and to add additional side cooling. A ring of a Cu-Al-foil (Al side facing the target) is then placed in front of the pellet and is fixed with a large sized copper nut to give it mechanical stability and allow transfer of heat to the nut.

After the high current irradiation (for example up to 15  $\mu\text{A}$  of 36 MeV  $^3\text{He}$  particles on enriched  $^{147}\text{Sm}_2\text{O}_3$ ) the pellet could be easily removed by opening the target with a special wrench. The loss of target material during the irradiation is below 5 %.

### **Foils and disks of metals and alloys**

A critical problem in irradiating poor heat conducting metal and alloy foils and disks is the bad contact between the target material and the target holder. Disks (or foils) of 14 mm diameter and 1 mm thickness are placed on the target holder (cooled from the back by a stream of water) and tightened in front using an O-ring and a screw cap which is also cooled with water. Such an arrangement withholds 20 MeV proton beam currents of about 20  $\mu\text{A}$ .

### **Thin electroplated layers on a wedged holder**

It is often difficult to produce electroplated layers of target material thicker than 150  $\mu\text{m}$ . In those cases the use of a slanting beam is advantageous since the effective target thickness increases by a factor of  $1/\cos \varphi$ . We developed a wedged target holder where the beam falls at an angle of  $20^\circ$ . An added advantage of this irradiation facility is that the power density is considerably reduced and even substances with low melting points (e.g. Se, Te, As, etc.) can be irradiated at moderate to high currents.



### **Gas target system for irradiation highly enriched gases**

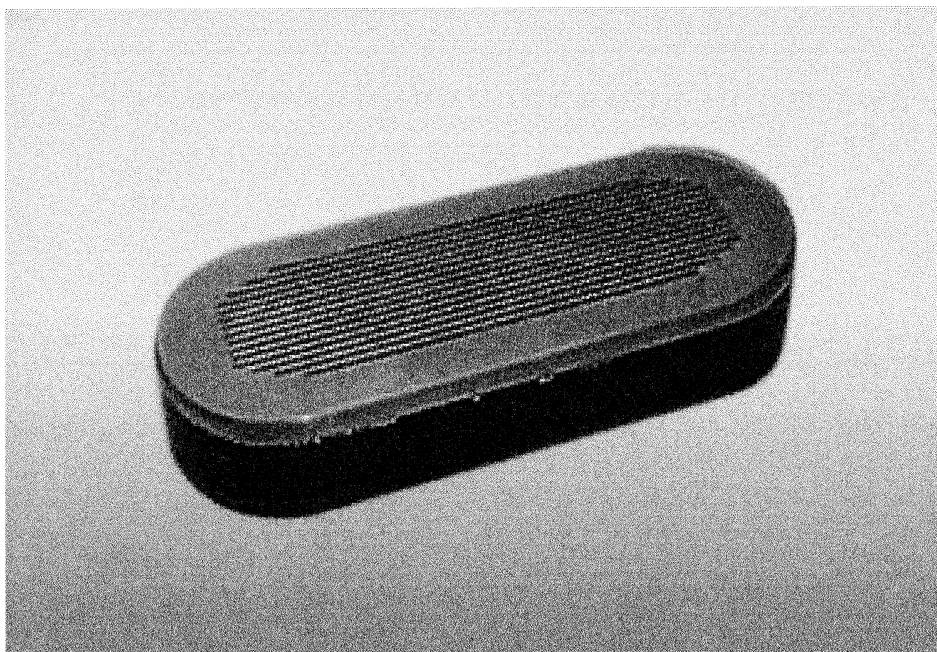
A safe gas handling and irradiation system has been developed. The reaction products formed are deposited on the inner walls of the target; their removal is effected by introducing steam (500 °C). The recovery yield of the radioactive products is > 90 % within 5 min and the total volume of the rinse solution amounts to about 6 mL. The system has been routinely used during the last year for the production of  $^{82\text{m}}\text{Rb}$  and  $^{83}\text{Sr}$  via  $^{82}\text{Kr}(\text{p},\text{n})^{82\text{m}}\text{Rb}$  and  $^{82}\text{Kr}({}^3\text{He},2\text{n})^{83}\text{Sr}$  processes. So far the loss of  $^{83}\text{Kr}$  was negligible.

## Solid Target for the Production of Radioiodines from Tellurium Oxide

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The existing solid targets [1] for radioisotope production accept plated target materials only. In order to allow the irradiation of powders and other non-plateable substances a new encapsulated target is being developed [2]. A similar target for producing radioiodines from  $\text{TeO}_2$  powder with a PET cyclotron [3] has been tested with beam but difficulties with the vacuum integrity of the target has hindered any significant progress. In the interim we have explored the possibility of using the existing commercial solid targetry to irradiate tellurium oxide by employing grooves in the surface of the solid targets which are filled with the oxide powder (see Figure 1). To ensure good adhesion of the oxide and efficient heat transfer during irradiation, the grooves were made narrow (0.6 mm) and run the full length of the target surface parallel with the water cooling channels immediately below the silver surface. The  $\text{TeO}_2$  was melted (around 735 °C) within the grooves and on resolidification was found to adhere strongly within the containing channels.



**Fig. 5:** A commercial solid target adapted to irradiate tellurium oxide

Beam irradiation tests were performed at 29 MeV with beam currents up to 150  $\mu\text{A}$  at which point the cooled target surface temperature is about 100 °C. No significant releases were observed during the irradiation process. Upon subsequent analysis the expected radioiodines were found to be present in the target material. By the appropriate choice of energy either the (p,n) or (p,2n) reactions can be favored on selected enriched tellurium targets to produce the specific radioiodine of choice. We will explore these options, the make-rates and impurities produced in the near future. In addition, the lower melting point of the monoxide,  $\text{TeO}$ , (370 °C) and the higher atom ratio of tellurium possibly makes this a more desirable target material so this option will also be examined.





The major challenge associated with this technique that remains to be solved is the extraction of the radioiodine from the target material. We intend to do this by dry distillation at 750 °C.

#### **References:**

- [1] W.Z. Gelbart et al., High Current Radioisotope Production with Solid Target System, Proc. Of the 1993 Particle Accelerator Conference, Washington DC, p 3099
- [2] W.Z. Gelbart et al., High Current Encapsulated Target and Target System for radioisotope Production, presented at this workshop
- [3] N. R. Stevenson et al., On-line Production of Radioiodines with low Energy Accelerators, Proc. 6<sup>th</sup> Workshop on Targetry and Target Chemistry, Vancouver B.C., p82 (1995)

#### Discussion:

Editors' remark: Part of the discussion was not recorded.

Q: J. Link: How do you remove the Te?

A: T. Ruth: We don't remove the Te, we sublime the iodine from the target.

Q: R. Weinreich: What is the thickness of the target backing?

A: N. Stevenson: The silver is 3 mm thick.

Q: What model did you use for your heat transfer calculations?

We used a heat transfer finite element analysis for our encapsulated target. The water is coming up from the bottom, being jet-streamed through a nozzle and the top few layers of this finite element analysis program is actually where the encapsulated material is. The bottom two or three elements represents where the return water squeezes by the back of this target material and exits. So you can see what is happening, the center of the beam, this represents half of the nozzle, half of the target. The center of the target, which has the highest beam current as well, is a Gaussian distribution across the front, it's the hot spot. And in the case of the water flow, there is a dead spot, a theoretical dead spot right in the center and that causes the typical shortfall of these finite element analysis programs that they do that. And as a result, they often overestimate the surface temperature. We actually put thermocouples on this target and irradiated it to almost 200  $\mu$ A and we found the surface temperature was much lower than predicted at the center. So that's the limitation of these finite element analysis programs, they can end up with these theoretical zeros, division by 0.

Q: J. Link: Who else uses these kind of heat calculations on their solid targets? Anybody?

C: We were doing something similar. It was not a pellet target if you remember, it was an ordinary solid target, very similar design as you have, but we haven't faced any problems with these division by zero problems.

A: No, you don't have a place where there is a theoretical void of water motion. This is what happens, when you have a jet coming up at the center and it then returns around completely. There's this theoretical point where the water does not move and that's simply an artifact of the program. In reality of course what happens is, there are imperfections causing some lateral motion, so there is no complete dead point.



C: J.-L. Morelle: We did similar simulations when we had designed the Pd-103 target. And the problem that we had was actually the input that you put in the program. One of the things that we didn't include was the variation of heat conductivity with temperature. And that was very important, once you go above 300°, 400° with copper. And that I would advise anybody who does that, to include that effect.

A: That's right. This program does have that, but some of the earlier ones don't have a temperature dependence of thermal conductivity.

Q: If I may ask another question. It's again concerning your solid target design. While you're irradiating TeO<sub>2</sub>, do you need some additional helium cooling from the front?

A: No, because we know the surface temperature of our targets typically go no higher than 150° with the water cooling that we have.

Q: I was just wondering about the losses of iodine?

A: That's a question that was asked before by Dr. Weinreich and I don't know the answer to that.

Q: J. Link: How thick is the layer of the oxide?

A: It fills the groove.

Q: J. Link: So a mm or two then?

A: 0.6 mm.

Q: J. Link: If it's 0.6 mm and there's such poor heat conductivity, how are you modeling the heat conductivity across that gradient? Are you just using the oxide?

A: We haven't done it for the tellurium oxide, we've done calculations for a straight electroplated target, which doesn't have the grooves in it. This is the encapsulated target, so it's a different thing.

Q: J. Link: No, I wasn't looking at this one, I am asking about your grooved target. On the grooved target, what is the distance between the grooves.

A: The grooves are very thin, 0.6 mm, they stagger in between the water cooling fins at the back. So they are actually quite close to the cooling water. We haven't modeled heat transfer in this particular case, we haven't done this for that specific target. What we have done is for the straight, electroplated target.

Q: J. Link: But will the grooved target have good heat conductivity?

A: I'm not sure, because you have this variable and somewhat unknown barrier between the electroplate and material, so you can get good targets and bad targets. It's difficult to have some quality control in that. And what we did, we modeled it for an unplated target and we modeled it for a silver target and then the plate that goes on it, we were unable to do that....

C: M. Nortier: We also modeled a 0.1 mm thick layer of thallium onto the silver target, but we assumed a very good thermal contact and no thermal resistance between the silver and the thallium.



A: Right. And what we find in practice is, that some of these materials will take much higher beam currents than others. And therefore the unknown quantity there of how well it's adhered is much more important quite often than many of the other factors, such as the thickness of the silver or the flow rate of the water. So the real emphasis has to be on the plating quality, the consistency of the plating quality.

C: G. Blessing: Maybe we had the same problem like this or a similar problem and solved this with our solid pellet target. This was the reason why we designed this pellet target head, especially because of low heat conductivity. This was our normal common used target and this target was done as further development. This little spike here (editor's note: the back of the pellet target head has a spike which extends toward the jet of cooling water entering the back of the target and improves the cooling water flow pattern) is especially because of the water stream, comes here and normally you got water circulating here, which is not cooling. And we insert here this little spike and make a cooling from the side and a very thick nut because we could not use a front cooling, because we needed maximum energy on the target system. In comparison with this target system, with our normal target system, just back cooling without this pike, we could irradiate lanthanide oxides with maximum of about 4  $\mu\text{A}$ . With this target system, we could irradiate the same pellet without a front cooling and even without a front covering with up to 15  $\mu\text{A}$  of beam. So we can get sufficient quantities of current on the target. I think this is the reason for the better cooling from the back and from the sides. Just with this little spike we could solve this problem of water turning around and not cooling.

Q: J. Link: Dr. Tolmachev, you have targets that are both etched and electroplated. Do you notice any difference in heat transfer in those two methods? Have you seen this problem of no heat transfer between layers or poor heat conductivity between layers?

A: Not really. We actually haven't measured the heat transfer, we're just using rule of thumb and increase the beam current in natural targets in steps of 2. And when the target starts to be visibly affected after short runs we backdown 2  $\mu\text{A}$ . What we have observed is, when we irradiate selenium targets, it's interesting, because it looks like when the beam is not stable, it's fluctuation of beam. So the target surface starts to be affected, but not immediately, it starts to be visible beam spot after heating and dry distillation process. Some sort of development of damages on the target surface. We can irradiate for example Ni-targets for Cu production with up to 14  $\mu\text{A}$  without any problems.

C: J. Link: The next topic is automation for recovery of the solid targets. And partly it's automation, partly it's chemical methods for recovery, and when do you use one, when do you use the other. Two days ago we heard from LBL that they can take out a target with a long stick and one screw and that's one method for making it remote. I know we can look at what Los Alamos does, or TRIUMF and they have these huge systems, big pneumatic systems, but for a smaller lab you don't have that ability. I was hoping Ruth Shefer could talk first about her automation system and then we can talk about chemical methods vs automation methods for removing targets.

## High Current Encapsulated Target and Target System for Radioisotope Production

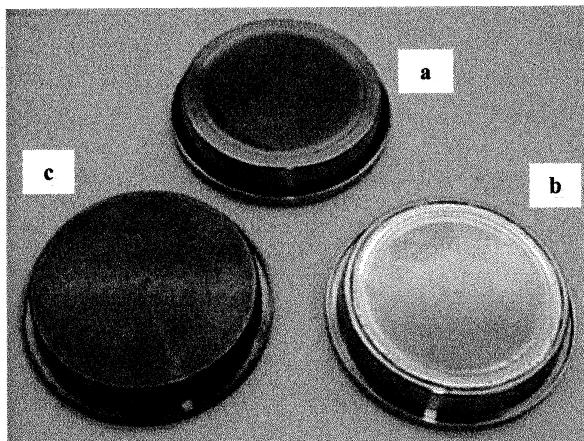
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### Introduction

Solid target systems for use with vertically oriented targets are currently used at TRIUMF for radioisotope production. These solid isotope production targets can only be used to irradiate metallic and electroplatable materials with a high melting point. In order to irradiate liquids, powders, nonelectroplatable materials, and materials with low melting points, a new horizontally oriented encapsulated target and target station is being developed. The encapsulated target is concentrically water-cooled and must withstand a 240  $\mu$ A 30 MeV proton beam. The thermal performance and the cooling and heat-induced stresses of the encapsulated target are being analyzed using finite element analysis (FEA). The results are then compared with actual measurements obtained using surface embedded thermocouples. This paper presents these results and conclusions and also summarizes the design of the encapsulated target station and the current status of the project.

### Encapsulated Target

The encapsulated target is machined out of a 70 mm diameter stainless steel ring with a silver soldered copper or silver central portion. This bimetallic construction enhances thermal performance and allows easy electron beam welding of the encapsulating foil to the periphery of the target. The target material, contained in a shallow recess of the central metal portion, is pre-placed prior to the welding of the foil. Figure 1a shows the target prior to encapsulation and an encapsulated target can be seen in Figure 1b. The system allows the use of conventional, plated targets as well. Figure 1c shows the round solid target made entirely out of copper in this case.



**Fig. 1:** Encapsulated Targets

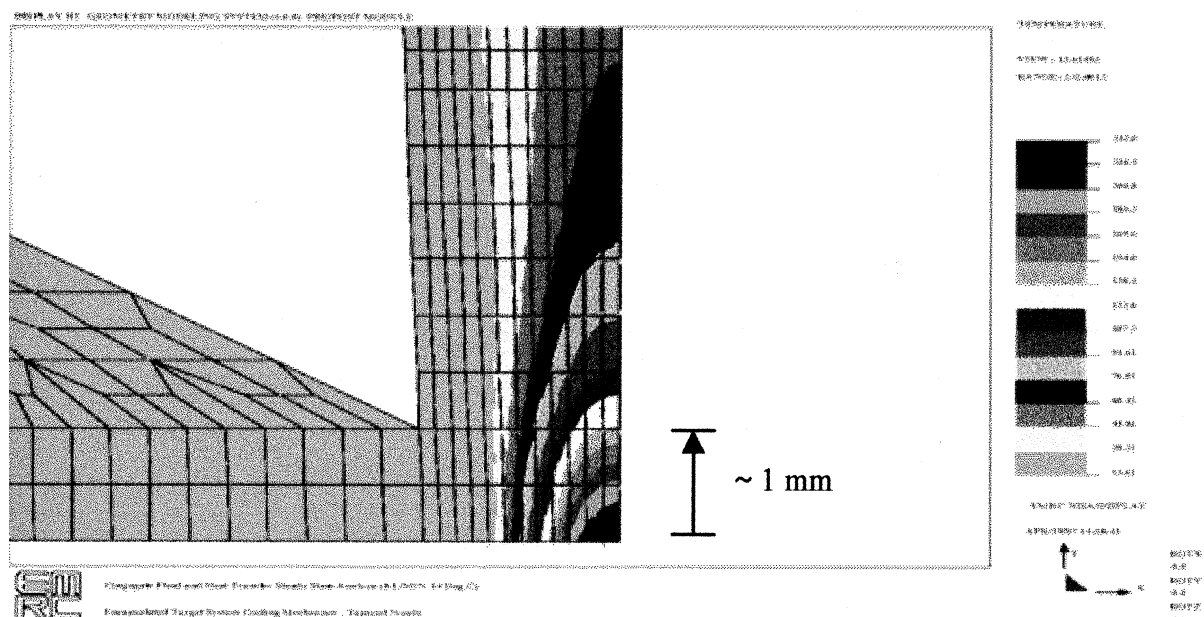


**Fig. 2:** Actuator Head

The target is positioned in the horizontal irradiation orientation by a remotely controlled actuator, which also creates a concentric coolant circuit against the back surface of the target during irradiation (see Figure 2) Water (at  $\sim 14^\circ\text{C}$ ) exits through the center hole, spreads radially across the back surface of the target, and drains through the holes on the outer rim of the actuator head. The entry water flow rate ranges from 8 L/min to 12 L/min. The thermal performance of the encapsulated target is examined using FEA computer modeling and is experimentally verified.

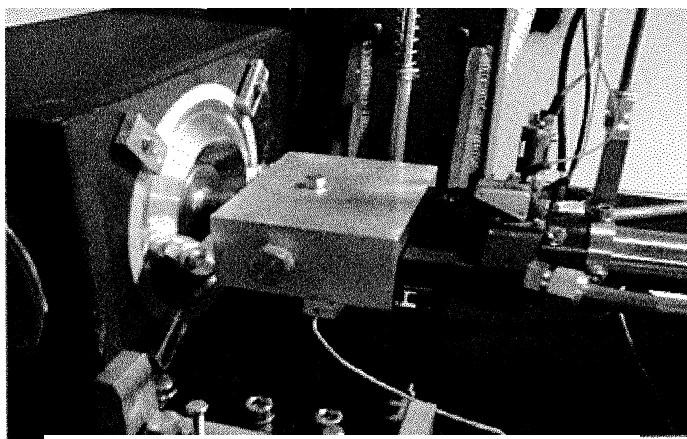


The thermal performance of the target is first investigated using computer modeling the 3D Fluid Module of the FEA program by EMRC, DISPLAY III/NISA II<sup>®</sup>. With the symmetric geometric properties of the encapsulated target and the assumption that the power density of the proton beam is Gaussian distributed over the target, the entire problem can be assumed to be axis-symmetric. The center metal portion of the target and the water coolant circuit are modeled using 3D axis-symmetric quadrilateral solid and fluid elements, respectively, and the heat source on the target (from the bombardment of the proton beam) is represented as a source of element heat generations distributed over three layers of elements of the target surface. Conjugate fluid flow and heat transfer steady state analysis (with temperature dependent material properties) is then performed for different combinations of target disc materials (copper and silver) and entry water flow rates (from 8 L/min to 12 L/min.) Figure 3 shows the typical temperature profile obtained from the FEA analysis.



**Fig. 3:** Temperature Profile of the Encapsulated Target during Irradiation

The results were then compared with actual temperature measurements obtained by irradiating a prototype encapsulated target with surface embedded thermocouples on an existing solid target station. A few components of the existing solid target station were modified to accommodate the round target and the new cooling method. A picture of the actual test setup is shown in Figure 4.



**Fig. 4:** Thermal Performance Test Setup on a Solid Target Station

### Target Station

Since the targets are irradiated in a horizontal position, a bending magnet is used to provide a 12.5° horizontal angle to the beam. The encapsulated target station (see Figure 5) has a modular assembly consisting of a landing terminal, a vacuum/irradiation chamber, and a

manipulator mounted on an aluminum stand. During operation, as the target (in its carrier called "rabbit") arrives at the target station from the hot cell through the pneumatic transfer system, it is brought to a soft landing and oriented in the required way by a dynamic braking and magnetic orientation system inside the landing terminal. The dynamic braking system utilizes the pressure build up at the closed end of the landing terminal and a reverse air flow injected into the receive housing of the landing terminal to slow and bring the rabbit to a "soft landing", while the magnetic orientation system simply takes advantage of the attraction/repulsion of opposite/similar magnetic polarities to orient the rabbit.

A manipulator grabber removes the target from the rabbit and pulls it along the manipulator guide until the target is directly underneath the target flange of the vacuum/irradiation chamber. The manipulator grabber then disengages from the target and an actuator head mounted on an air cylinder lifts the target off the manipulator guide and pushes the target against the target flange for irradiation. This actuator head (see Figure 2) creates a concentric coolant circuit against the back surface of the target and seals the vacuum/irradiation chamber at the same time. Additional cooling is provided for the top surface of the target by a forced flow of helium gas that is incorporated into the design of the target flange.

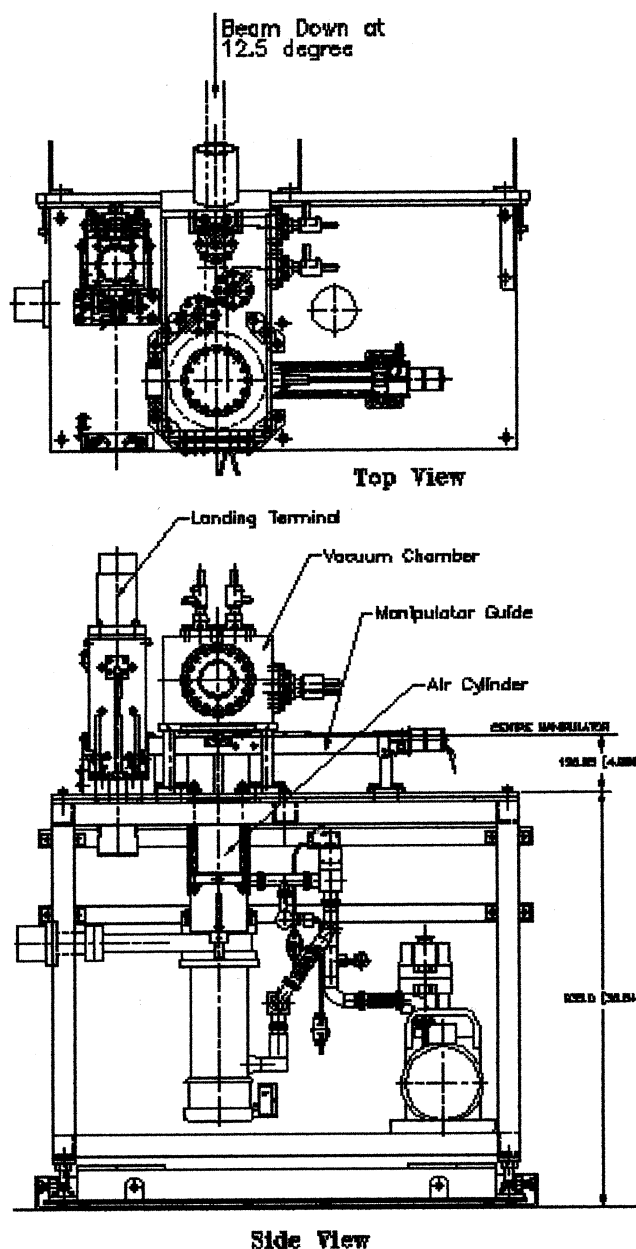


Fig. 5: Encapsulated Target Station

#### References:

- [1] W.Z. Gelbart, N.R. Stevenson, R.R. Johnson, F.M. Nortier, J. Orzechowski, F. Cifarelli, "High Current Radioisotope Production with Solid Target System" Proceedings of the 1993 Particle Accelerator Conference, pp. 3099-3101
- [2] F.M. Nortier, N.R. Stevenson, W.Z. Gelbart, "Investigation of the Thermal Performance of Solid Targets for Radioisotope Production," Nuclear Instruments and Methods in Physics Research, Section A **335**: 236 (1995)
- [3] F.M. Nortier, N.R. Stevenson, "Thermal Analysis of the TRIUMF Solid Targets", Symposium on Isotope Production and Applications in Medicine, Science and the Environment, Vancouver, Canada, 1993

Discussion:

Q: J. Clark: Nigel, your encapsulated target ....(inaudible)

A: I've got another drawing somewhere, you bring it in at about 20 degrees. What you have to do is you have to put a bending magnet in because you have solids that become liquids -- it's just gallium. You have to have the target horizontally. So we have to bend the beam through a magnet down onto this target. So I think it glances at 20 degrees.

Q: J. Nickles: The front window of the encapsulated targets, is it just an energy degrading foil, or is there a back stop, e. g. in the case of Rb going to  $^{82}\text{Sr}$ , you are going down from 70 to something like 40 MeV....

A: Which encapsulated target? -

J. Nickles: The encapsulated target for Rubidium.

A: So the 70 MeV one. That's not my target system.

J. Nickles: But you could be having a back stop of natural Rhodium for example, for making Palladium-103.

A: You mean piggy-back the targets effectively? Sure, you could do that I guess, but it's not done. But yes, I see no reason why it couldn't. You have to think about the design of it.

Q: T. Ruth: Is there a demand for Pd-103 ?

A: Nickles: I think there is going to be an infinite demand for Pd-103 and I think that this group is all going to be discovering it perhaps at the same time or separately. But waste beam or beam stops or slits or however you are dumping 200  $\mu\text{A}$  of beam at 30 MeV, that could do a job...

Stevenson: Well that's a good idea. It's possible. There's numerous ways of making that product anyway other than the way it's conventionally made at 18 MeV.

C: J. Nickles: True, but rhodium is such a benign target material, it's such a wonderful backstopper.

N. Stevenson: Yes. It's 100 % the right isotope as well.

Q: How do you prepare the solid targets? By electroplating or evaporating?

A: Yes. The target material is electroplated. So the solid target is just the conventional electroplated method, we have been using those for years. The new thing, the encapsulated target allows us to produce isotopes that have some difficulty with the target material, whether it be a salt, a powder of some sort, a foil or like gallium, that becomes a liquid at 30 degrees. So it's a more general target, but it's high beam current, several hundred  $\mu\text{A}$ .

Q: J. Clark: Any touch on material problems with gallium ?

A: Yes, we wondered what the container should be made of. We tried stainless steel and then Niobium has been suggested, I think they do that at Brookhaven. But yes, it's not a very nice one to start with in that sense, you're right.



Q: Do you reuse the solid target backings ?

A: Yes, ten to twenty times. The backing is made of aluminum, copper-plated.





## Development of Target Chemistry for Long-Lived Radioisotopes, Production for Biomedical and Environmental Studies

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The works on isotopically pure nuclides production are initiated by intensive researches of their applications in biomedical and environmental practice. With this aim in view a number of nuclides such as  $^{26}\text{Al}$ ,  $^{97}\text{Tc}$ ,  $^{97}\text{Ru}$ ,  $^{178}\text{W}$  ( $^{178}\text{Ta}$ ),  $^{235,236}\text{Np}$ ,  $^{236,237}\text{Pu}$  were produced through p, d,  $^4\text{He}$  and photonuclear reactions. The irradiations of stable and radioactive targets were conducted with JINR accelerators. Processes for preparation of high-pure carrier-free isotopes were developed, including target and chemistry designs, radiochemical methods, electromagnetic isotopes separation.

a) Gamma-emitters  $^{97}\text{Ru}$  ( $T_{1/2} = 2.9$  d, EC;  $E_\gamma$  215 and 324 keV) and  $^{178}\text{Ta}$  ( $T_{1/2} = 9.3$  m, EC;  $E_\gamma$  93.2 keV) are suitable for application in diagnostic and therapeutic ( $^{97}\text{Ru}$ ) purposes. The nuclear reactions forming these nuclides are  $^{99}\text{Tc}(p,3n)^{97}\text{Ru}$  ( $\sigma_{\text{max}} = 440$  mb; 32 MeV) and  $^{181}\text{Ta}(p,4n)^{178}\text{W} \xrightarrow[EC]{22\text{ d}} ^{178}\text{Ta}$  ( $\sigma_{\text{max}} = 495$  mb; 40 MeV) [1].

An efficient method to separation of  $\text{Tc}_{\text{met}}$  macroquantities (grams) and carrier-free  $^{97}\text{Ru}$  has been developed [2]. It is based on  $^{97}\text{RuO}_4$  distillation from  $\text{HNO}_3$  or  $\text{H}_2\text{SO}_4$  solution ( $\text{HTcO}_4$ ) at  $90^\circ\text{C}$  with following absorption in a  $\text{HCl-H}_2\text{O}_2$  trap. Gamma-spectroscopic analysis of final  $^{97}\text{Ru}$  solution revealed absence of Tc and other impurities. The chemical yield of  $^{97}\text{Ru}$  is 95-98 %.

The method of  $^{178}\text{W}$  separation has been developed on the basis of the data high temperature ( $1100\text{-}1200^\circ\text{C}$ ) sublimation and gas-thermochromatographic behavior carrier-free W [3]. The high W volatilization yield from heated  $\text{Ta}_{\text{met}}$  in the  $\text{O}_2\text{-H}_2\text{O}$ -vapor atmosphere ( $T \sim 1160^\circ\text{C}$ ,  $P_{\text{H}_2\text{O}} \geq 400$  torr,  $V_{\text{gas carrier}} = 15$  cm<sup>3</sup>/min) and its distribution along the thermochromatographic column ( $T_{\text{absorb}} \sim 560^\circ\text{C}$ ) were selected as conditions for radiochemically pure  $^{178}\text{W}$  production.

b)  $^{26}\text{Al}$  ( $T_{1/2} = 7.2 \times 10^5$  y;  $\beta^+$ , EC;  $E_\gamma$  1808 keV) and  $^{237}\text{Pu}$  ( $T_{1/2} = 45.6$  d; EC;  $E_\gamma$  97, 101, 114 keV) are of interest for studies of metabolism of Al and Pu tracers in the human body. The nuclear reactions  $^{\text{nat}}\text{Mg}(^4\text{He},pxn)^{26}\text{Al}$  and  $^{235}\text{U}(^4\text{He},2n)^{237}\text{Pu}$  were used [1].

Radiochemical separation of  $^{26}\text{Al}$  from irradiated  $\text{Mg}_{\text{met}}$  was realized after one month storage. The different procedures of  $^{26}\text{Al}$  purification from other nuclidic impurities were used: sorption of cobalt, zinc and gallium on the anion exchange column, coprecipitation of Fe-hydroxide (at this stage magnesium and sodium remained in the solution), further cation exchange procedure with hydrochloric-acetone mixtures. The chemical yield of high pure  $^{26}\text{Al}$  was estimated as not less than 95 % [4].

Plutonium-237 isolation from the irradiated enriched  $^{235}\text{U}_3\text{O}_8$  target was carried out using the lanthanum-fluoride method, anion exchange chromatography and following additional isotopic enrichment with electromagnetic mass-separation. The obtained purity of  $^{237}\text{Pu}$  ( $^{236}\text{Pu} : ^{237}\text{Pu} : ^{238}\text{Pu} = 3 \times 10^{-8} : 1 : 3 \times 10^{-8}$ ) is the highest reported to date [5].

c)  $^{235}\text{Np}$  ( $T_{1/2} = 396.1$  d; EC;  $E_\gamma$ , X),  $^{236}\text{Np}$  ( $T_{1/2} = 1.54 \times 10^5$  y; EC;  $\beta^-$ ;  $E_\gamma$  160 keV),  $^{236}\text{Pu}$  ( $T_{1/2} = 2.86$  y;  $\alpha$ ) are neptunium and plutonium tracers in the investigations of distribution, transfer and transport processes of these elements in the environment. These isotopes have been



produced by means of the following nuclear reactions on  $^{235}\text{U}$  (highly enriched  $^{235}\text{U}_3\text{O}_8$ ) and  $^{237}\text{Np}$  (preliminary purified  $^{237}\text{NpO}_2$ ) [6]:



The radiochemical separation procedures were as one for  $^{237}\text{Pu}$ . The  $^{236}\text{Pu}$  purity (relative activity) was  $7 \times 10^{-2}$   $^{237}\text{Pu}$ ,  $< 10^{-5}$   $^{238}\text{Pu}$  and  $\sim 10^{-9}$   $^{239}\text{Pu}$  from  $^{235}\text{U}$  target (after chemistry and mass separation) and  $7 \times 10^{-5}$   $^{238}\text{Pu}$  and  $6 \times 10^{-6}$   $^{239}\text{Pu}$  from  $^{237}\text{Np}$  target (only after chemistry).

d)  $^{97}\text{Tc}$  ( $T_{1/2} = 3.5 \times 10^6$  y; EC 100 %, no gammas) is convenient tracer for mass-spectrometric determination of  $^{99}\text{Tc}$ . The  $^{95}\text{Mo}(^4\text{He},2\text{n})^{97}\text{Ru} \xrightarrow[\text{EC}]{2.9 \text{ d}} ^{97}\text{Tc}$  reaction was used. After cooling for 100 days  $^{97}\text{Tc}$  was separate from enriched  $^{95}\text{Mo}_{\text{met}}$  by distillation procedure with 98 % chemical yield and high purity (purification from Mo was  $2 \times 10^{-6}$ ).

All obtained results are shown that the developed methods (target material, construction, conditions of irradiation, radiochemical separation and purification) allow to prepare very isotopically pure radionuclides.

#### References:

- [1] S.N. Dmitriev, N.G. Zaitseva, Phys. Part. Nucl. **27**(4): 403 (1996)
- [2] N.G. Zaitseva, V.I. Stegailov, V.A. Khalkin et al., Appl. Radiat. Isot. **47**: 145 (1996)
- [3] B. Bayar, A.F. Novgorodov, I. Vocilka, Radiochem. Radioanal. Lett. **22**: 53 (1975)
- [4] S.N. Dmitriev, Y.T. Oganessian, J. Radioanal. Nucl. Chem., Art's. **205**: 101 (1996)
- [5] S.N. Dmitriev, Y.T. Oganessian, G.Y. Starodub et al. Appl. Radiat. Isot. **46**: 307 (1995)
- [6] S.N. Dmitriev, JINR Sci.Report 7-96-361, p. 64 (1996)



## The Target for Production of High-Purity $^{236}\text{Pu}$ in $^{237}\text{Np}(\text{gn})$ Reaction

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As a result of nuclear tests, nuclear industry accidents, but also in connection with the accumulation of nuclear waste, the concentrations of various long-lived radionuclides in the environment have increased. Some of the most environmentally dangerous ones, because of their effects on the biosphere, are the plutonium isotopes. To address the urgent problems related to plutonium contamination, ultra sensitive methods of analysis must be developed for the measurement of Pu concentrations in biological samples (food products, plants, animal tissue, etc.), as well as other environmental samples (soil, water, etc.). Particular interest have attracted methods which permit the determination of the isotopic ratio  $^{239,240}\text{Pu}/^{238}\text{Pu}$ , which enables identification of the source of contamination (i.e. nuclear tests, nuclear plant accidents, etc.). One of the best suited radiotracers for this purpose is  $^{236}\text{Pu}$ .

However, in order to be used as a tracer,  $^{236}\text{Pu}$  must meet stringent purity requirements. In particular, the concentrations of  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  should not exceed (Bq/Bq) of  $10^{-3}$ - $10^{-4}$  and  $10^{-6}$ - $10^{-7}$ , respectively. Existing methods of  $^{236}\text{Pu}$  which satisfy these requirements are of low efficiency and expensive due to the need for an additional purification step of one or two mass separation procedures.

Research performed in our laboratory indicates that the  $^{237}\text{Np}(\gamma, n)^{236\text{m}}\text{Np} \rightarrow ^{236}\text{Pu}$  reaction is more promising for the production of high-purity  $^{236}\text{Pu}$  [1]. This is because in the interaction of photons with  $^{237}\text{Np}$  there are no channels open resulting in  $^{238-240}\text{Pu}$ . Under this scenario, the isotopic purity of the  $^{236}\text{Pu}$  preparation is determined only by the radiochemical purity of the initial  $^{237}\text{Np}$ . In addition, the fission cross section is comparable to the cross section for the main reaction.

As a source of gamma-quanta, the circular small footprint "microtron" electron accelerator MT-25 of the FLNR JINR was used. The accelerated were extracted from the microtron through an 8 mm collimator and impeded on a stopping tungsten target of 2 mm thickness in order to produce gamma-rays.

The intensity of the gamma-ray beam was measured along its axis up to a distance of 140 mm and was found to decrease linearly with distance. A 20 % decrease of photon intensity was observed at a radial distance of 8 mm from the beam axis at a distance of 13 mm from the stopping target along the beam axis. The gamma-ray beam aperture corner is equal to  $7.4 \pm 0.1^\circ$ .

A cylindrical aluminum container of 8 mm inner diameter and 5 mm height, filled with the neptunium compound, was positioned in the center of the irradiation target unit. The container was closed with an aluminum cover of 0.1 mm thickness. It was then wrapped in 50  $\mu\text{m}$  thick aluminum foil and placed snugly in the bottom of a cylindrical aluminum holder open on one end. An aluminum foil seal of 50  $\mu\text{m}$  thickness was glued to the open end of the holder to protect against accidental spills. The holder was then tightly attached to the stopping target holder, which was water cooled, with its axis coinciding with the gamma beam axis. The distance between the stopping target and the irradiated target was 8 mm. As beam position controls, Au and Ag monitors of 5 mm diameter were positioned externally at the non-beam facing base of the cylindrical holder.



The temperature of the sample during irradiation did not exceed 100 °C. This allows the use of not only neptunium oxide, but also of other neptunium compounds after preliminary processing in 100 °C.

The concentrations of  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  in the initial  $^{237}\text{Np}$  were 0.7 and 0.06 Bq/mg, respectively. The target contained 10 mg  $^{237}\text{Np}$  and was irradiated for 100 hours with maximum photons energy of 25 MeV. The average electron beam current extracted from the MT-25 microtron was  $\sim 15 \mu\text{A}$ .

The irradiated target was allowed to "cool" for one month in order to allow a complete decay of  $^{236\text{m}}\text{Np}$  and short lived  $\gamma$ - and  $\beta$ - emitters. The target was subsequently dissolved in 8 M  $\text{HNO}_3$ . The nuclear reaction products were separated with by anion exchange in a column containing the strong acid anion exchanger Wofatit SWB (200 mesh). The isolation of the plutonium fraction from the  $^{237}\text{Np}$  was carried out by sorption of the latter from a 9 M  $\text{HCl}$  + 0.1 M  $\text{NH}_4\text{I}$  solution as  $\text{Np(IV)}$ .  $\text{Pu(IV)}$  was loaded on to a column from a 8 M  $\text{HNO}_3$  +  $\text{NaNO}_2$  solution and was eluted using 9 M  $\text{HCl}$  + 0.1 M  $\text{NH}_4\text{I}$ . The  $^{236}\text{Pu}$  solution was examined with a  $\text{Si(Au)}$  surface-barrier detector ( $0.7 \text{ cm}^2$ ) with a resolution of 16 keV. The  $^{236}\text{Pu}$  yield was estimated to be as much as 50 Bq/ $\mu\text{Ah}$ . Determination of the  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  concentrations in the  $^{236}\text{Pu}$  preparation was carried out after a preliminary separation using the electromagnetic mass separator of FLNR JINR. The ratio  $^{236}\text{Pu}/^{238}\text{Pu}/^{239}\text{Pu}$  was estimated to be  $1/7 \times 10^{-5}/6 \times 10^{-6}$  Bq/Bq.

The isotopic purity achieved this way meets the requirements of the majority of radioecological investigations. It can be enhanced by 1 to 2 orders of magnitude by an additional purification of the target material and by increasing the irradiation time. Expenditures for the production of  $^{236}\text{Pu}$  using the  $^{237}\text{Np}(\gamma, n)^{236\text{m}}\text{Np} \rightarrow ^{236}\text{Pu}$  reaction are 10 lower compared to methods utilizing the  $^{235}\text{U}(\alpha, 3n)$  reaction (followed by mass separation) and  $^{235}\text{U}(d, n)$  one. Therefore we can conclude that the  $^{237}\text{Np}(\gamma, n)$  reaction is the most optimal one for the production of  $^{236}\text{Pu}$  with isotopic purity of  $10^{-6}$ - $10^{-7}$  Bq/Bq.

#### Reference:

[1] S.N. Dmitriev et al., Abstract of Papers Sixth Int. Conf. Low Level Measurements of Actinides and Long-Lived Radionuclides in Biological and Environmental Samples, Cherbourg, France, 1996, p.52

## Production of $^{139}\text{Ce}$ by Bombardment of a Praseodymium Target with 66 MeV Protons

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Long-lived ( $T_{1/2} = 137.64$  d)  $^{139}\text{Ce}$  is well-known as a laboratory standard for the calibration of  $\gamma$ -ray detectors. Since it emits only one  $\gamma$ -ray (165.857 keV) in the optimum energy range for detection with a gamma camera this radioisotope also has a high potential for use in calibration sources for SPET. In order to explore this possibility a production procedure for  $^{139}\text{Ce}$  was developed at the NAC and an experimental  $^{139}\text{Ce}$  line source was prepared for one of the South African hospitals.

Surprisingly, no suitable cross-section data for the cyclotron production of  $^{139}\text{Ce}$  could be found in the literature. The excitation function for the production of  $^{139}\text{Ce}$  in the bombardment of praseodymium with protons up to 100 MeV was therefore measured in this work and the thick-target yield calculated from these data is compared with yield measurements obtained with a Pr-metal target.

Finally, several high-current production runs were executed with the 66 MeV proton beam which is available on a regular basis for the production of radioisotopes, employing Pr-metal as target material rather than  $\text{Pr}_6\text{O}_{11}$  as originally envisaged [1].

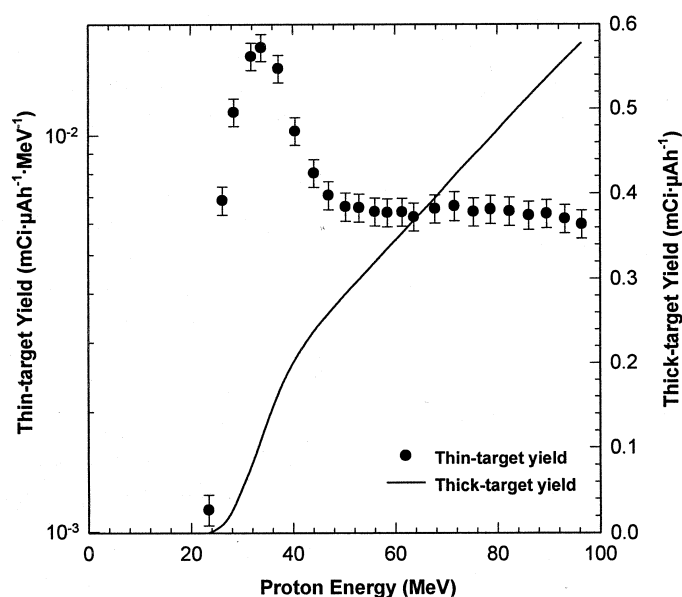


Fig. 1:  $^{139}\text{Ce}$  yield in the bombardment of praseodymium with protons

### Reference:

[1] F.J. Haasbroek, P. Andersen, J.H. Langenhoven, F.M. Nortier, D. Du T. Rossouw and T.N. Van der Walt, Production of radionuclides at the NAC cyclotron facility. Proc. 14th Int. Conf: on Cyclotrons and their Applications, Cape Town, 1995, ed. J.C. Cornell (World Scientific, 1996) p. 559

## **Automated Solid Targets and Processing Systems for the Production of Intermediate-Lived Radionuclides**

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An automated target and radiochemical processing system have been developed for the production and separation of intermediate-lived positron-emitting radionuclides using small biomedical cyclotrons. The target utilizes a remotely actuated air cylinder to position a target substrate disk in the cyclotron beam strike. During irradiation, cooling is provided by water flowing in direct contact with the back surface of the substrate disk. The target has operated reliably for 2-3 hour irradiations at proton beam powers of up to 700 Watts with a coolant flow rate of approximately 1 liter per minute. After irradiation, the disk is ejected under remote control and transported via a dedicated pneumatic line to a hot cell located above the cyclotron vault. The target has been tested extensively at Washington University School of Medicine for the production of high specific activity  $^{64}\text{Cu}$  and other copper radioisotopes on layers of enriched nickel electroplated on gold (see: D.W. McCarthy et. al., these proceedings). Production of other intermediate-lived radioisotopes, including  $^{66}\text{Ga}$ ,  $^{76}\text{Br}$ ,  $^{77}\text{Br}$ ,  $^{86}\text{Y}$ ,  $^{94\text{m}}\text{Tc}$  and  $^{124}\text{I}$ , using the automated solid target holder is under investigation. Calculated yields and solid target designs for these radioisotopes will be presented. Because of their relatively long half-lives, many of these radioisotopes are suitable for distribution. Shipment of  $^{64}\text{Cu}$  produced at Washington University is currently being investigated, and plans for making this radioisotope available to other researchers in the near future will be described.

An automated processing system has been designed and constructed for the separation of copper radioisotopes from nickel target material. A heated cell for dissolution of the irradiated nickel from the target substrate accepts targets from the pneumatic line with no operator intervention. The target solute is transferred under computer control to an anion exchange column for separation of the dissolved copper fraction from the nickel target material and other radionuclides. Provisions are included for recovery of the enriched nickel in a form suitable for reuse in electroplating. All system components in contact with the target solute are pre-sterilized and disposable, thereby minimizing set-up and clean-up time. Testing of the automated system is currently underway and preliminary results of automated copper separation will be reported. Extension of the capabilities of this system to the separation of the other radioisotopes listed above from solid targets will also be described.

### Discussion:

Q: J. Link: Is that essentially a peristaltic pump that you are using?

A: Yes, it's similar to the mechanism in a peristaltic pump. These are very small, this is less than 1" across.

Q: J. Link: What kind of plastic is it? The tubing.

A: The tubing is C-flex, I don't remember what the polymer is. (editors note: C flex is obtained from Cole Parmer)



Q: N. Stevenson: I have a question about the pneumatic transfer. When the targets drop into the tube, do you get any contamination? It doesn't use rabbits, so what prevents transfer of the radioactivity into the pneumatic system.

A: The contamination of the tube, that hasn't been looked at carefully. This tube is only used for that target holder. And there's no obvious damage to the Ni-layer when the target comes out the other end. That has been checked out carefully.

Q: J. Link: Does the Ni/Cu look as white as the picture? Does it look like that before it is irradiated?

A: The Ni coating is sort of silvery white, the electrodeposited coating. It looks like that before and after irradiation.

Q: R. Hichwa: This pinch valve that you designed, it's not obvious to me from the picture. Is there another tube around the outside? Circling that valve?

A: (pointing to slide) You mean there? This is a bearing.

Q: R. Hichwa: So that's actually metal there?

A: Yes, it's metal, it's a bearing. So this rotates independently of the cam.

Q: I have a question on the lifetime of the plastics. The tubing and the dissolution vessel, have you had to replace it because of radiation damage?

A: We don't know yet. We've only started testing the system with cold Cu recently. However, our intention is that this whole backing with the tubing will be disposable, be single use. So we are not very concerned about the lifetime of the plastics.

Q: And the dissolution vessel?

A: The dissolution vessel could be used for more than one dissolution, but we don't know, what the lifetime will be.

Q: J. Clark: Do I understand that you have a Cu backing, Ni front cover? Ni on Cu?

A: Ni on Au.

C: J. Clark: It didn't look like Au.

A: No it is. That's very important actually, because we were concerned about specific activity with all the Cu around.

C: K. Erdman: I just want to make a comment about pinch valves. We did an extensive development on pinch valves for automating of our FDG system and we have now got a faultless pinch valve. You can see absolutely no visible damage to the tube after over a thousand cycles of operation. You can use different tubes. And we have a small model out in front, where you can actually see how this pinch valve works. You can push a button and operate it if you want. And it's a rotating pinch valve. Pinch valves work very well, if they are designed properly

Q: J. Comor: I couldn't figure out from the slide you showed us, how you are inserting the target before irradiation?





A: We are actually designing a turret now, which will hold several disks and will position each disk sequentially into the position. But right now it's inserted by hand. That's the only part that's done manually.

Q: J. Comor: And the other point, are you reusing these gold backings?

A: Yes.

C: M. Welch: Ruth, you might want to point out that we are not using only the gold disk with this target system.

A: Yes, I just want to show you one transparency from Tim McCarthy who has been using this target to irradiate graphite to make  $^{13}\text{N}$ , to make NO and  $\text{N}_2$ . And the graphite in this case is press fitted into a high temperature ceramic piece, which is then inserted, right where the gold disk would be inserted. And that withstood up to 100 W of beam power. One thing that I didn't mention before is that we irradiate our gold disks up to about 700 W on target. So this is another type of target, that can be used in this holder. We are also looking at the production of other radioisotopes on packed powders and other materials by adapting this holder.

## The Efficient Production of Various Positron Copper Radionuclides Using a Biomedical Cyclotron

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We have previously described a target system that has been routinely used for the production of high specific activity  $^{64}\text{Cu}$  via the  $^{64}\text{Ni}(p,n)^{64}\text{Cu}$  reaction [1].  $^{64}\text{Cu}$  produced this way has been used successfully for chemistry studies and expanded to imaging studies in both animals and patients [2]. Using a 15 MeV proton beam, yields of up to  $\sim 1$  Ci (at EOB) have been achieved with this system (specific activities up to 310 mCi/ $\mu\text{g}$  Cu). We have recently adapted this system for the production of  $^{60}\text{Cu}$  and  $^{61}\text{Cu}$ . Both isotopes have potential use as PET tracers.  $^{60}\text{Cu}$  is a 93 % positron emitter with a 23.4 min half-life.  $^{61}\text{Cu}$  is a 60 % positron emitter with 3.3 hr half-life.  $^{60}\text{Cu}$  has been produced by the  $^{60}\text{Ni}(p,n)^{60}\text{Cu}$  nuclear reaction and  $^{61}\text{Cu}$  by both the  $^{60}\text{Ni}(d,n)^{61}\text{Cu}$  (8 MeV deuteron beam) and  $^{61}\text{Ni}(p,n)^{61}\text{Cu}$  nuclear reactions. Yields of up to 865 mCi of  $^{60}\text{Cu}$  have been achieved using enriched  $^{60}\text{Ni}$  (major isotopic impurities: 0.05 %  $^{61}\text{Cu}$ , 0.025 %  $^{57}\text{Co}$ ).  $^{61}\text{Cu}$  has been produced with a maximum yield of 144 mCi using enriched  $^{61}\text{Ni}$  and a yield of 72 mCi using enriched  $^{60}\text{Ni}$  (major isotopic impurity is 0.04 %  $^{58}\text{Co}$ ).

**Tab. 1:** Production Parameters (15 MeV protons, 8 MeV deuterons, 99+ % enrichment)

Isotope	Nuclear Reaction	Target Thickness [microns]	Max. Yield [mCi]	Yield [mCi/ $\mu\text{A}$ hr]	Major Isotopic Impurities
$^{60}\text{Cu}$	$^{60}\text{Ni}(p,n)^{60}\text{Cu}$	225	865	58	0.05 % $^{61}\text{Cu}$ , 0.025 % $^{57}\text{Co}$
$^{61}\text{Cu}$	$^{61}\text{Ni}(p,n)^{61}\text{Cu}$	118	144	7.6	0.04 % $^{58}\text{Co}$
$^{61}\text{Cu}$	$^{60}\text{Ni}(d,n)^{61}\text{Cu}$	122	72	2.44	0.02 % $^{58}\text{Co}$
$^{64}\text{Cu}$	$^{64}\text{Ni}(p,n)^{64}\text{Cu}$	215	921	8.0	0.012 % $^{55}\text{Co}$

In summary, the target system that we have developed is a reliable method for the routine production of  $^{60}\text{Cu}$ ,  $^{61}\text{Cu}$  and  $^{64}\text{Cu}$ .

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### References:

- [1] D.W. McCarthy, R.E. Shefer, R.E. Klinkowstein, L.A. Bass, W.H. Margenau, C.C. Cutler, C.J. Anderson, M.J. Welch, J. Nucl. Med. Biol. **24**: 35-43 (1997)
- [2] F. Dehdashti, C.J. Anderson, D.D. Trask, L.A. Bass, S.W. Schwarz, P.D. Cutler, D.W. McCarthy, M.V. Lanahan, J. Nucl. Med. (abstract in press) (1997)



Discussion:

Q: How much  $^{61}\text{Cu}$  do you use for an average study or a typical study?

A: Depends on what you are going to use it for. We use it for doing imaging of tumors and we used  $\mu\text{Ci}$  amounts, 100  $\mu\text{Ci}$  in a rat.

C: You can use natural Ni and use deuterons. You will produce plenty of  $^{61}\text{Cu}$ . The only impurity will be  $^{62}\text{Cu}$ .

## **Evaluation of a Commercially Available Inductive Heater for the Rapid Combustion of Graphite Disks Used in the Production of $[^{13}\text{N}]\text{NO}$ and $[^{13}\text{N}]\text{NO}_2$**

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The production of nitrogen-13 by the  $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$  reaction on graphite and subsequent combustion has been demonstrated to be of use in the production of oxides of nitrogen-13. As a part of our ongoing interest in the role of inhaled nitric oxide [1], we are interested in developing a portable device that can be used to produce and deliver  $[^{13}\text{N}]\text{nitric oxide}$  of varying, but controlled, specific activity. Since it must be portable, our strategy is to deliver an irradiated graphite disk contained in a suitable holder directly to the device for combustion.

We have purchased a suitable induction heater from Ameritherm, Inc. The heater consists of a 1 KW Nova 1 power supply and a custom designed heating station which can be situated up to 10 feet from the power supply. The heating coils of the inductive heater were designed to rapidly heat the target (high density graphite; 0.5" diam. x 0.03" thick) and holder. Using an infra-red temperature transducer to monitor the combustion process, we determined that the target can reach temperatures in excess of 1000 °C in less than 30 sec. These results will allow us to significantly shorten the synthesis and delivery time of  $[^{13}\text{N}]\text{nitric oxide}$ . We are currently integrating this heating method with the rest of the device for further evaluation.

### **Acknowledgment:**

The authors are grateful to the Whitaker Foundation for supporting this research.

### **Reference:**

[1] T.J. McCarthy, C.S. Dence, S.W. Holmberg, J. Markham, D.P. Schuster and M.J. Welch, Inhaled  $[^{13}\text{N}]\text{nitric oxide}$ : a positron emission tomography study, Nucl. Med. Biol. **23**: 773-777 (1996)

## Co-Production of High Specific Activity $^{64}\text{Cu}$ from $^{67}\text{Ga}$ Production Waste

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$\text{Cu-64}$  has been reported to have attractive physical properties ( $t_{1/2} = 12.7$  hr,  $E_{\beta} = 0.57$  MeV,  $E_{\beta+} = 0.66$  MeV) relevant to the diagnosis and therapy of disease [1,4]. Its production is readily achieved in either a cyclotron or a reactor and considerable information exists regarding its purification. Boothe et al. was the first to report high activity levels of  $^{64}\text{Cu}$  formed during the production of  $^{67}\text{Ga}$  [5]. Unfortunately the product they isolated was contaminated with inactive copper, presumably from the target backing. At the National Medical Cyclotron, (NMC) Sydney, the same reaction is used to produce  $^{67}\text{Ga}$  from enriched  $^{68}\text{Zn}$ , on a nickel coated copper target plate. Analysis of the  $^{67}\text{Ga}$  waste showed that there was  $^{64}\text{Cu}$  present and it appeared to be included by the proton activation of nickel; to attempt to gain access to this potentially useful radionuclide a method of extraction that would avoid contamination of inactive copper was investigated [6].

The separation of metal ions using non-aqueous solvents and anion exchange resin was first reported in the 50's [7a,7b]. Fritz and Pietrzyk (1961) reported a number of distribution coefficients for the partition of a number of metal ions between anion exchange resins and various organic solvent-water mixtures [8]. The use of non-aqueous solvent systems was attractive for this study as they provide greater separation power than aqueous acid solution for the desired radionuclides. A process was developed at the NMC which provided a quick and efficient method for separating  $^{64}\text{Cu}$  from the  $^{67}\text{Ga}$  waste that used low acid aqueous/organic mixtures [6].

Proposed Reactions for the Formation of $^{67}\text{Cu}$ and $^{64}\text{Cu}$ during the Production of $^{67}\text{Ga}$	
$^{68}\text{Zn}(p,pn)^{67}\text{Cu}$	at 1.6 – 2.9 $\mu\text{Ci}/\mu\text{A hr}$
$^{68}\text{Zn}(p,pn)^{67}\text{Cu}$	and / or
$^{68}\text{Zn}(p,pn)^{67}\text{Cu}$	at 1.6 – 2.9 $\mu\text{Ci}/\mu\text{A hr}$
Production Yield of $^{64}\text{Cu}$ from Natural Nickel Foil	
$^{68}\text{Zn}(p,pn)^{67}\text{Cu}$	at 1.6 – 2.9 $\mu\text{Ci}/\mu\text{A hr}$

More than 50 experiments have been conducted and the separation method has been refined. High yields (up to 180 GBq at EOB) of  $^{64}\text{Cu}$  are routinely isolated from the  $^{67}\text{Ga}$  waste. The specific activity of the product is high,  $5 \pm 5 \times 10^{14}$  Bq/g, and is hence suited to the radiolabelling of proteins and peptides. The purity (i.e. radionuclidic, radiochemical and chemical) of the  $^{64}\text{Cu}$  is sufficiently high that it may also be used in pre-natal diagnosis testing for Menke's disease.

Other radionuclides such as,  $^{57}\text{Ni}$ ,  $^{57/55}\text{Co}$ , and  $^{67}\text{Cu}$  are co-produced during the  $^{67}\text{Ga}$  production [6] Distribution coefficients for up to 35 different acid/organic solvent mixtures with AGI-X8 anion exchange resin for each metal ion radionuclide have been determined. Highly efficient separation of mixtures of these elements, that display similar chemistry, is now readily achieved. The new separation method used at the NMC allows reliable separation of

$^{57}\text{Ni}$ ,  $^{57/55}\text{Co}$ , and  $^{67/64}\text{Cu}$ . The use of a single anion exchange column provides a simple, fast and high yielding process that also simplifies waste management of these radionuclides

The ability to separate radionuclides from a single column provides an opportunity to exploit parasitic activation which would otherwise only add to the problem of waste disposal. More importantly this advance has led to the possibility of designing targets from which multiple products can be routinely extracted, thereby broadening of the range of radionuclides that is now readily achievable.

#### References:

- [1] A. Smith, U. Zsigmondy-Wittke, R. Walble, T. Schenker, P.A. Schubiger, R.A. Stahel, *Int. J. Cancer (Suppl)* **8**: 43 (1994)
- [2] P. Smith-Jones, R. Fridrich, T. Kaden, I. Novak-Hofer, K. Siebold, D. Tschudin, H. Mack, *Bioconjugate Chem.* **2**: 415 (1991)
- [3] A. Smith, R. Alberto, P. Blaustein, I. Novak-Hofer, R.H. Macke, P.A. Schubiger, *Cancer Res.* **53**: 5727 (1993)
- [4] C.J. Anderson, J.M. Connett, S.W. Schwarz et al., *J. Nucl. Med.* **13**: 1685 (1992)
- [5] T.E. Boothe, E. Tavano, J. Mucoz, S. Carrol, *J. Label. Compd. Radiopharm.* **30**: 108 (1991)
- [6] S.V. Smith, D.J. Waters, N. Di Bartolo, *Radiochimica Acta* **75**: 65 (1996)
- [7a] E.W. Berg, J.T. Truemper, *Analy. Chem.* **30**: 1827 (1958)
- [7b] Y. Yoshima, Y. Kurimura, *Bull. Chem. Soc. Japan* **30**: 563 (1957)
- [8] J.S. Fritz, D.J. Pietrzyk, *Talanta* **8**: 143 (1961)

## Production of $^{72}\text{Se}$ to Support a Clinical $^{72}\text{As}$ Generator for PET

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Arsenic-72 (half-life 1.1 days) is a radioisotope with significant potential to support medical imaging by Positron Emission Tomography (PET). Its physical decay characteristics and the versatile chemistry of arsenic make this nuclide very attractive for the synthesis of a variety of PET radiopharmaceuticals. Since  $^{72}\text{As}$  is the daughter of the longer-lived  $^{72}\text{Se}$  (half-life 8.5 days), a radiochemical generator can be used on-site at a clinical imaging center or regional radiopharmacy to provide the  $^{72}\text{As}$  radiopharmaceuticals in high specific activity. A generator system based upon precipitation of elemental selenium followed by filtration to yield the arsenic-containing labeling solution has been developed [1]. The applicability of  $^{72}\text{As}$  for PET imaging in comparison to  $^{18}\text{F}$  has been demonstrated [2]. Due to these developments there is growing interest in establishing a clinical generator system to provide  $^{72}\text{As}$  for PET. A critical element to the success of such a generator will be the reliable continuous supply of the  $^{72}\text{Se}$  parent. We will review past work and our current investigations into proton accelerator production and recovery of this radioisotope.

At Los Alamos  $^{72}\text{Se}$  is produced by spallation reactions on natural RbBr targets in an 800 MeV, 1 mA proton current [3]. Numerous radionuclides of many elements are produced in these targets necessitating a careful chemistry for recovery of the selenium fraction. Furthermore several radioisotopes of selenium are produced including Se-72 (12 mb cross-section), Se-73 (22 mb cross-section), and Se-75 (67 mb cross-section). The ratio of  $^{72}\text{Se}/^{75}\text{Se}$  is extremely important for future medical use of the  $^{72}\text{As}$  generator. The best  $^{72}\text{Se}/^{75}\text{Se}$  ratios (1.0-1.2) are obtained using short irradiations (25,000-80,000 mA-hrs) yielding a total of 200-300 mCi  $^{72}\text{Se}$  on 30-35 grams of RbBr [4]. Much higher total yields (2 Ci) are obtained with longer irradiations (400,000-500,000 mA-hrs) on targets of 160-180 grams, but the ratio of  $^{72}\text{Se}/^{75}\text{Se}$  is decreased by a factor of 2 or more [5]. Selenium isotopes are recovered from the target by dissolving the RbBr into a 6 M HCl matrix, adding 2 mg of selenic acid carrier, and hydrazinium hydrochloride. The solution is boiled to distill and recover arsenic and germanium isotopes in the form of volatile hydrides. During the distillation, elemental selenium is selectively precipitated from the target solution by reduction of the selenic acid by the hydrazinium ion. The selenium is recovered by filtration, and dissolved by oxidation back to selenic acid by hydrogen peroxide into a hydrochloric acid solution.

At the Institute for Nuclear Research (INR) production yields of  $^{72}\text{Se}$  as well as  $^{71,73,74}\text{As}$ ,  $^{73,75}\text{Se}$ ,  $^{68,69}\text{Ge}$  were measured on GaAs thin plate targets bombarded with 40-140 MeV protons. The measured excitation function for the reaction  $^{75}\text{As}(p, 4n)^{72}\text{Se}$  has been compared with those calculated by the cascade-evaporation model and ALICE code. It is anticipated that the product from this mode of production would have a much more favorable  $^{72}\text{Se}/^{75}\text{Se}$  ratio (about 15:1) over the peak production energy range than that of the spallation production method. A design for a GaAs target for high grade  $^{72}\text{Se}$  production is being developed and a high temperature gas chemical method is being developed to recover selenium isotopes from the irradiated GaAs. In this method GaAs is inserted into a graphite container and heated in pure helium and hydrogen flow. Arsenic is removed from the sublimed products by high temperature absorption with a liquid aluminum filter in helium flow and by a CaO chemical filter in air flow while the selenium is passed through the filter and



collected on copper catcher foils [6,7]. The selenium would be eluted from the catcher foils by an appropriate dilute acid solution to provide the feedstock for the existing solution based generator system.

#### References:

- [1] D.R. Phillips, V.T. Hamilton, D.A. Nix, W.A. Taylor, D.J. Jamriska, R.C. Staroski, R.A. Lopez, A.M. Emran, Chemistry and Concept for an Automated  $^{72}\text{Se}/^{72}\text{As}$  Generator, in *New Trends in Radiopharmaceutical Synthesis, Quality Assurance and Regulatory Control*, A.M. Emran, ed.: Plenum Press, New York, 1991
- [2] D.R. Phillips, V.T. Hamilton, M.D. Taylor, J.E. Farnham, A.M. Emran, R.W. Rowe, D. Pattel, Generator-Produced Arsenic-72 in Positron Emission Tomography, *Radioactivity and Radiochemistry* **3**: 53-58 (1992)
- [3] P.M. Grant, D.A. Miller, J.S. Gilmore, H.A. O'Brien, Medium-Energy Spallation Cross Sections. 1. RbBr Irradiation with 800 MeV Protons, *Int. J. Appl. Radiat. Isot.* **33**: 415-417 (1982)
- [4] D.R. Phillips, D.C. Moody, W.A. Taylor, N.J. Segura, B.D. Pate, Electrolytic Separation of Selenium Isotopes from Proton Irradiated RbBr Targets, *Int. J. Radiat. Appl. Instrum. Part A: Appl. Radiat. Isoto.* **38**: 521-525 (1987)
- [5] D.R. Phillips, V.T. Hamilton, W.A. Taylor, D.J. Jamriska, R.C. Staroski, R.C. Heaton, Production of  $^{68}\text{Ge}$ ,  $^{73,74}\text{As}$ , and  $^{72,75}\text{Se}$ , published abstract from the 205<sup>th</sup> American Chemical Society National Meeting, Denver, Colorado, March 28-April 2, 1993
- [6] B.L. Zhuikov, Separation of Volatile Elements and Oxides: Thermochromatography and the Use of Chemical Filters. Report ORNL-TR-5125, Oakridge National Laboratory, 1984
- [7] B.L. Zhuikov, Report of Joint Institute for Nuclear Research, P12-82-64



## Production and Recovery of $^{109}\text{Cd}$ from Metallic Indium Targets

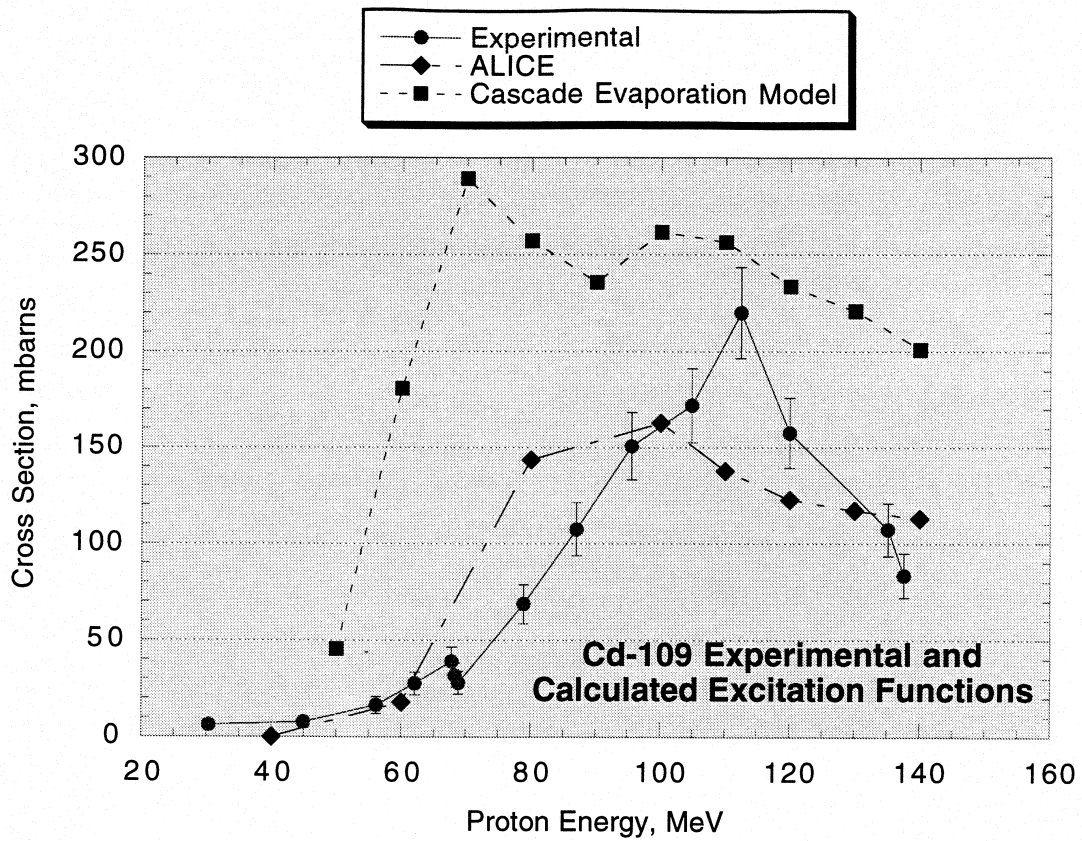
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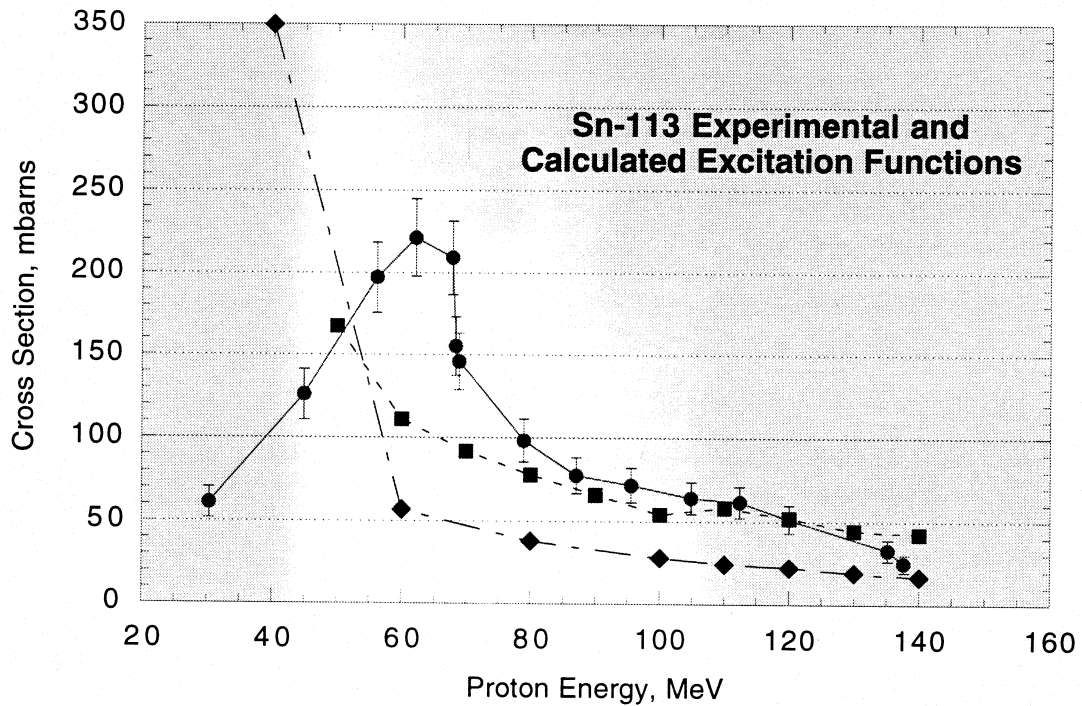
Indium metal targets are used to produce  $^{109}\text{Cd}$  by proton bombardment at the Isotope Production Facilities at both Los Alamos National Laboratory and the Institute for Nuclear Research. Both Institutions currently use traditional wet-chemical procedures (dissolution in concentrated hydrochloric acid followed by ion-exchange methods) to recover and purify the  $^{109}\text{Cd}$ . In this paper we report yield measurements for production of the cadmium and a new gas-chemical procedure for its recovery.

The excitation functions for several radionuclides produced by proton bombardment of indium metal were measured over the energy range 30 - 140 MeV at the INR Isotope Production Facility. These include  $^{109}\text{Cd}$ ,  $^{113}\text{Sn}$ ,  $^{111}\text{In}$ ,  $^{114\text{m}}\text{In}$ ,  $^{105}\text{Ag}$ , and  $^{106}\text{Ag}$ . Figures 1 and 2 show the experimental and calculated excitation functions for  $^{109}\text{Cd}$  and  $^{113}\text{Sn}$ , respectively. The data for  $^{113}\text{Sn}$  are shown due to the growing interest in  $^{113}\text{Sn}$  as the parent of  $^{113}\text{In}$  which has a variety of potential practical uses. The maximum cumulative cross section value for  $^{109}\text{Cd}$  (including  $^{109}\text{Sn}$  and  $^{109}\text{In}$ ) is  $290 \pm 60$  mbarn at about 120 MeV. Based upon the measured excitation function the total production yield of  $^{109}\text{Cd}$  from a thick target would be near 0.3 mCi/ $\mu\text{A}$  hr at EOB in the energy range 80 - 140 MeV. Production yields have also been measured using 600 - 800 MeV protons at the Los Alamos Isotope Production Facility and found to be much lower; near 0.01 mCi/ $\mu\text{A}$  hr.

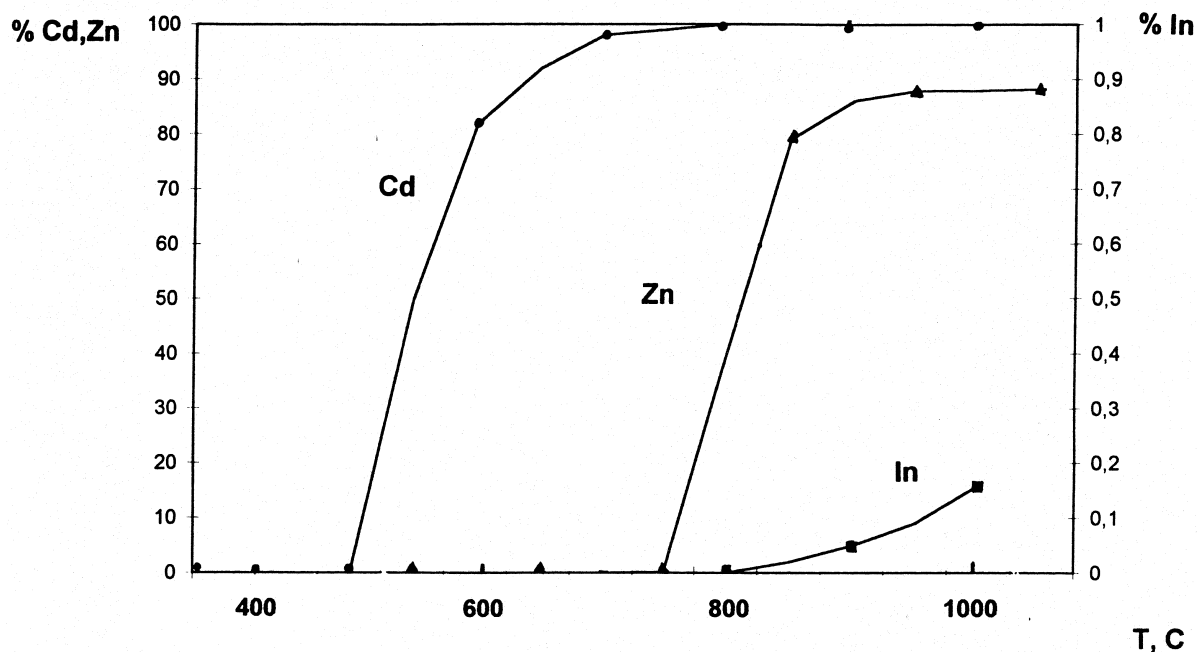
The currently used wet chemical methods used to recover and purify the  $^{109}\text{Cd}$  give rise to relatively large volumes of liquid radioactive waste. A new high temperature gas-chemical procedure for  $^{109}\text{Cd}$  recovery is being developed which would provide advantages over the wet-chemical approach. In this process cadmium is evaporated from metallic indium at high temperature in helium and hydrogen flow and is deposited on a quartz surface or on metallic catcher foils. Complete cadmium evaporation occurs at  $700 \pm 50^\circ\text{C}$  under the given conditions with no evaporation of other radionuclides observed in the targets (e.g.,  $^{114\text{m}}\text{In}$ ,  $^{113}\text{In}$ ,  $^{105}\text{Ag}$ ,  $^{110\text{m}}\text{Ag}$ ,  $^{102}\text{Rh}$ ). During evaluation of this technique it was important to understand the behavior of  $^{65}\text{Zn}$  which is a volatile impurity in the spallation production on indium targets at the higher proton energies. Therefore we explored isolation of zinc from gallium using the same high temperature method. Figure 3 shows the temperature-yield curves for  $^{109}\text{Cd}$  evaporated from indium and  $^{65}\text{Zn}$  evaporated from gallium. At  $750^\circ\text{C}$  greater than 98 % of the cadmium was evaporated from the molten indium. At this same temperature less than 0.2 % of the zinc evaporates from molten gallium. At  $900^\circ\text{C}$ , greater than 90 % of the zinc is volatilized. Based upon these experiments it appears probable that a clean separation of zinc and cadmium can be achieved from irradiated indium metal targets. In order to verify this the thermochromatographic separation of Cd and Zn on copper catcher foils was studied. Figure 4 shows the schematic experimental set-up and results. Zinc is deposited at  $700 - 500^\circ\text{C}$  on copper using a chemisorption process, while cadmium is deposited at  $360 - 120^\circ\text{C}$ . The adsorption heat is calculated from these data. Based upon these results and observations, we propose that the high temperature method is promising as a technology for high activity production of  $^{109}\text{Cd}$  for commercial distribution.



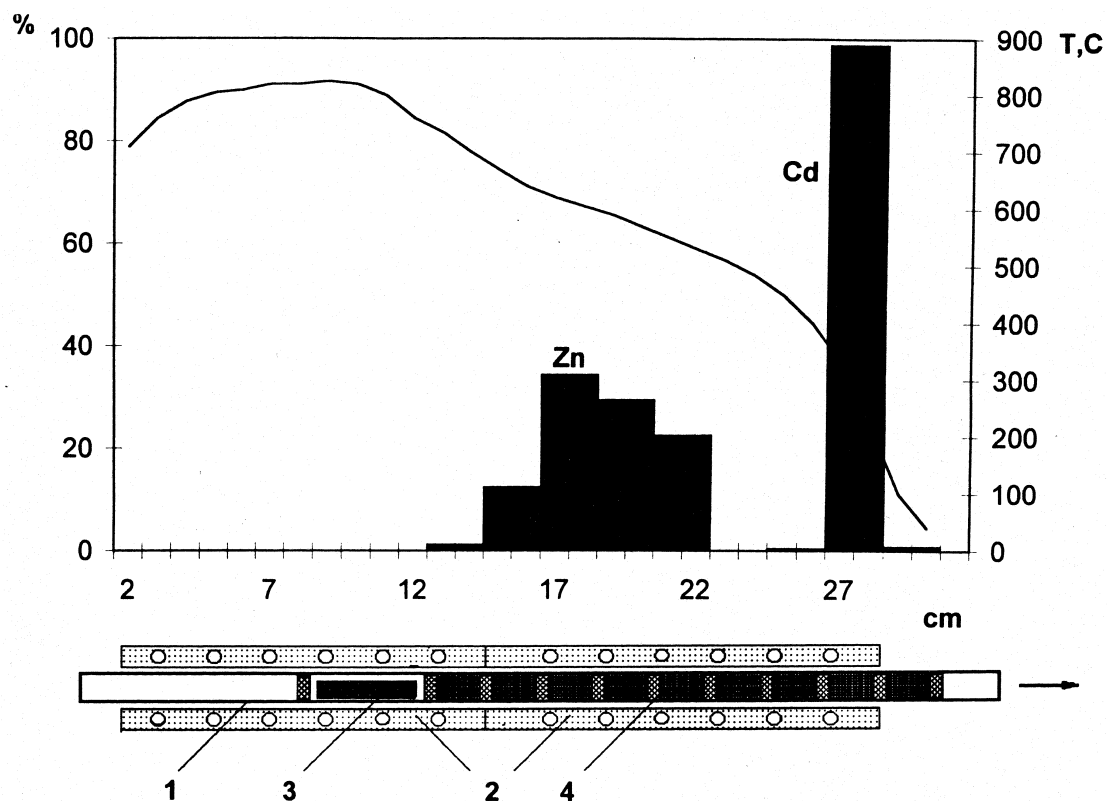
**Fig. 1:** Comparison of experimental and theoretical excitation functions for the production of  $^{109}\text{Cd}$  by bombardment of indium metal with 140 - 30 MeV protons.



**Fig. 2:** Comparison of experimental and theoretical excitation functions for the production of  $^{113}\text{Sn}$  by bombardment of indium metal with 140 - 30 MeV protons.



**Fig. 3:** Temperature dependence of integrated evaporation yield of  $^{109}\text{Cd}$  from Indium and  $^{65}\text{Zn}$  from gallium.



**Fig. 4:** Set-up and results for the thermochromatographic Separation of Cd and Zn from: Indium metal. 1) Quartz tube; 2) High temperature tube furnaces; 3) Boat with metallic Indium doped with  $^{109}\text{Cd}$  and  $^{65}\text{Zn}$  tracers; 4) Copper catcher foils on surface of column, fixed by alumina wool.



Discussion:

Q: J. Steinbach: How do you remove the separated radionuclides from the quartz tube without cross-contamination, when you take it out?

A: If it is a quartz tube, you may put just a catcher at the end. If it is quartz tube, it may be absorbed on the tube, then you may just add acid and remove it from the quartz without problem. It completely removes from quartz with acid.

C: T. Ruth: There's several things that you can play with, I separated rhenium from tungsten, and both rhenium and tungsten have very high melting points. But if you flow helium with a very small amount of oxygen over, you form rhenium oxide which has a very low melting point. And you can get quantitative removal of one from the other, even though the base metals could have very high or similar melting points.

## Routine Production of $^{76}\text{Br}$ at a Low-Energy Cyclotron

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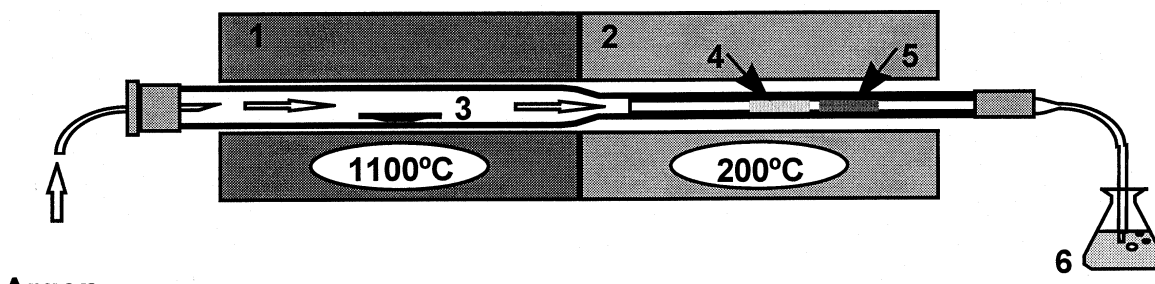
### Introduction

Positron Emission Tomography (PET) is a powerful diagnostic tool. So far it has mainly been associated with short-lived positron emitters with half-lives < 2 hours like  $^{11}\text{C}$  and  $^{18}\text{F}$ . However, many biomedical processes have to be monitored for several hours and days and thus positron emitters with longer half-lives are needed. To be able to meet such demands we are developing techniques for producing non-conventional positron emitters at a low energy cyclotron [1,2,3]. In this paper we present development of a routine production of  $^{76}\text{Br}$  aimed for clinical applications with e.g. monoclonal antibodies [4,5].

### Material and Methods

The cyclotron at the Uppsala PET center (proton energy 17 MeV) and the  $^{76}\text{Se}(p,n)^{76}\text{Br}$  reaction was used [6,7] with enriched  $\text{Cu}_2^{76}\text{Se}$  pellets as target material (Stabis Ltd, Moscow, Russia, enrichment 96.5 %). The  $\text{Cu}_2\text{Se}$  pellets (diameter 8 mm, thickness 180 mg/cm<sup>2</sup>) were placed on a water cooled copper backing and were irradiated for up to 2h with 16 MeV protons. Beam current was about 14  $\mu\text{A}$  or less.

In a simple set-up (Fig. 1) radiobromine was separated from the irradiated target material by dry distillation. Some amounts of selenium was distilled as well and  $^{76}\text{Br}$  was separated from the bulk of selenium traces by thermal chromatography. The target was placed in a platinum ship inside a quartz tube (i.d. 24 mm) positioned in the middle of a. In an adjacent auxiliary furnace (range 15-30 cm), a polytetrafluoroethylene (PTFE) tube (i.d. 2 mm) was inserted into the quartz tube. The end of the PTFE tube was immersed into a dilute NaOH solution.



**Fig. 6:** Schematic description of the  $^{76}\text{Br}$  separation equipment. 1) Furnace, 2) Auxiliary furnace, 3) Target in platinum ship, 4) Deposition area of selenium, 5) Deposition area of radiobromide, 6) Gas trap.

The separation procedure was initiated by raising the temperature in the furnace containing the target pellet to 1100 °C, and the second furnace to 200 °C under a constant 30 mL/min stream of argon gas through the tubes. The bromine leaving the target material was transferred to the PTFE tube by the carrier gas, and condensed on the wall of the tube.

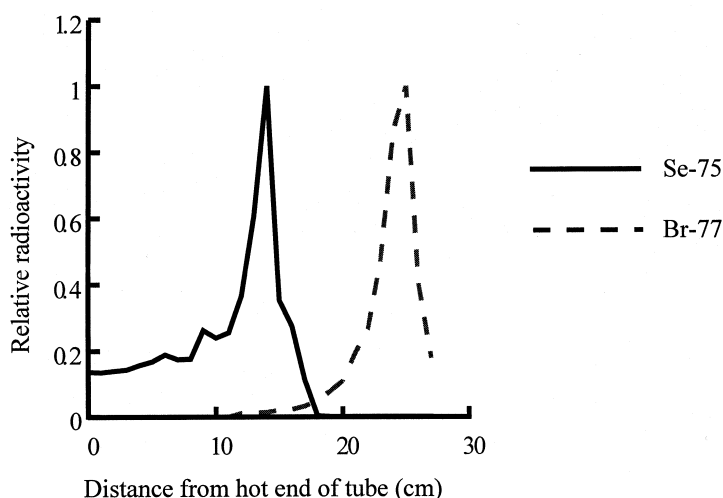


In order to evaluate the efficiency of the thermochromatographic separation, the deposition pattern of bromide and selenium was studied by utilizing  $^{77}\text{Br}$  and  $^{75}\text{Se}$ , formed via the  $\text{Se}(\text{p},\text{xn})^{77}\text{Br}$  and  $^{76}\text{Se}(\text{p},\text{pn})^{75}\text{Se}$  reactions after irradiating a copper selenide target of natural isotopic composition. The separation yield and loss of target material in the procedure was estimated by  $\gamma$ -spectroscopy and weighing of pellets before and after separation. Within a few hours after separation, the radiochemical purity of the  $^{76}\text{Br}$ -preparations was analyzed by thin-layer chromatography (TLC) and high performance liquid chromatography (HPLC).

## Results and Discussion

The target assembly could withstand beam currents up to 14  $\mu\text{A}$ . At higher beam currents the target material started to get somewhat affected. The production yield were found to be 65-70 MBq/ $\mu\text{Ah}$  (EOB). Usually production runs for 1-2 hours were made in the late evening and the separations in the morning in order to let coproduced  $^{63}\text{Zn}$  to decay. The targets were fetched manually and transported to an offline separation equipment.

Separation yields were typically 65-75 % and the separation time 1 hour or slightly higher. The final solution volume was in the order of 0.2 mL. Routine production twice a week (150-300 MBq/batch) has now been performed since September 1996. Typical radiation dose to personnel during re-load and processing of the target is 40  $\mu\text{Sv}$ . Losses in target material was found to be less than 1 % per run. Targets can be run up to twenty times before being refreshed. The pellet is then grind and target material replacement is added before repressing of target. Occasionally the target ma: be cracked and has to be refreshed earlier. Conditioning of the oven (rinsing with argon and heating up to working temperature) took



**Fig. 2:** Separation of selenium and bromide by thermal chromatography, showing the deposition areas of  $^{75}\text{Se}$  and  $^{77}\text{Br}$  along the PTFE tube. The radioactivity was normalized to maximum values.

most of the separation time. When at operation temperature there was a rapid release of radioactivity from the target. The differing condensation temperatures of selenium and bromine made possible the separation of these compounds by thermal chromatography. Using a copper selenide target of natural isotopic composition, an auxiliary furnace temperature of more than 150  $^{\circ}\text{C}$  separated the deposition areas of  $^{75}\text{Se}$  and radiobromine along the PTFE tube (Fig. 2).

Radiochemical analysis by TLC and ion-exchange HPLC showed that nearly all  $^{76}\text{Br}$ -activity (more than 97 %) was in the form of bromide. The only radionuclide impurity, detected was  $^{77}\text{Br}$  (< 2 %), probably originating from traces of  $^{77}\text{Se}$  in the enriched Se preparation.





## Conclusions

The  $^{76}\text{Se}(p, n)^{76}\text{Br}$  reaction using a  $^{76}\text{Se}$ -enriched  $\text{Cu}_2\text{Se}$  target enables a simple and cost-effective routine production of  $^{76}\text{Br}$  in a low-energy cyclotron. In the present study, the application of thermal chromatography in conjunction with dry distillation allowed the separation of  $^{76}\text{Br}$  from the bulk of the target material as well as traces of volatilized selenium.

## References:

- [1] H. Lundqvist, V. Tolmachev, A. Bruskin, L. Einarsson, P. Malmberg, Rapid separation of  $^{110}\text{In}$  from enriched Cd targets by thermal diffusion, *Appl. Radiat. Isot.* **46**: pp 859-863 (1995)
- [2] V. Tolmachev and H. Lundqvist, Rapid separation of Gallium from Zinc targets by thermal diffusion, *Appl. Radiat. Isot.*, **47**: 297-299 (1996)
- [3] V. Tolmachev, H. Lundqvist and L. Einarsson, Diffusion based separation methods: Dry distillation of zinc, cadmium and mercury isotopes from irradiated targets. *Appl. Radiat. Isot.*, In print
- [4] A. Lövgqvist, A. Sundin, A. Roberto, H. Ahlström, H. Carlsson and H. Lundqvist, Comparative PET imaging of Experimental Tumors with  $^{76}\text{Br}$ -Labeled Antibodies,  $^{18}\text{F}$ Fluorodeoxyglucose, and  $^{11}\text{C}$ Methionine, *J. Nucl. Med.*, Accepted 1996
- [5] A. Lövgqvist, A Sundin, H. Ahlström, J. Carlsson and H. Lundqvist, Pharmacokinetics and experimental PET imaging of a  $^{76}\text{Br}$ -labeled monoclonal anti-CEA antibody, *J. Nucl. Med.*, Accepted 1996
- [6] Z. Kovacs, G. Blessing, S.M. Quaim and G. Stöcklin, Production of  $^{75}\text{Br}$  reaction at a compact cyclotron, *Int. J. Appl. Radiat. Isot.*, **36**: 635-642 (1985)
- [7]. W. Vaalburg, M.J. Paans, J.W. Terpstra, T. Wiegman, K. Dekens, A. Rijskamp and M.G. Woldring, Fast recovery by dry distillation of  $^{75}\text{Br}$  induced in reusable metal selenide targets via the  $^{76}\text{Se}(p, 2n)^{75}\text{Br}$  reaction, *Int. J. Appl. Radiat. Isot.*, **36**: 961-964 (1985)

## Production and Applications of $^{55}\text{Co}$ in PET

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Positron emission tomography (PET) is an imaging technique originating from nuclear medicine, where it is mainly applied as an in-vivo, non-invasive medical research tool for investigations in the oncological, neurological and cardiological field. PET has the advantage over the other nuclear imaging technique (SPECT) that absolute quantification of activity (Bq/mL) is possible. The technique has been explored for the non-invasive in-situ dynamic imaging of 2 industrial model experiments: visualization and quantification of flooding with ( $^{66}\text{Ga}$ -EDTA) labeled water of a core (oil reservoir rock) and the separation of the same tracer molecule from a water/oil emulsion [1].

In a second study, in-situ exhaust catalysis studies using  $^{11}\text{C}$  - and  $^{15}\text{O}$ -labelled gasses ( $\text{O}_2$ ,  $\text{CO}$ ,  $\text{CO}_2$ ,  $\text{CH}_4$ ) provided useful information in the investigation of automotive exhaust catalysis kinetics and reactions [2].

Recently, the production of  $^{55}\text{Co}$  was developed and evaluated for the study of slow dynamic processes in stone (concrete and limestone).  $^{55}\text{Co}$  ( $t_{1/2} = 17.53$  h) is produced by irradiation of an Fe-foil with a proton beam (23 MeV) using a (p,2n) reaction. The details are summarized in Table 1.

**Table 1:** Irradiation conditions for the production of  $^{55}\text{Co}$

Target: Fe-foil (99.99+ % purity)	
	diameter: 15 mm
	thickness: 75 $\mu\text{m}$ - 250 $\mu\text{m}$ (113 - 355 mg)
Irradiation conditions:	
	particle: p
	energy interval: 23 - 18 MeV
	beam intensity: 5 $\mu\text{A}$
	duration: 2 h
	beam diameter: 12 mm
	yield: 0.3 GBq

$^{55}\text{Co}$  is separated from the Fe-matrix and radiochemical impurities ( $^{52}\text{Mn}$ ) in the technical set-up visualized in Fig. 1. The whole purification procedure is summarized in Scheme 1. The radionuclidic purity was checked with a Ge-Li detector.

After complexation with EDTA, the solution is ready for injection via a high pressure pump in a core flooding set up (Fig. 2a) placed in the Siemens 951/31 camera.

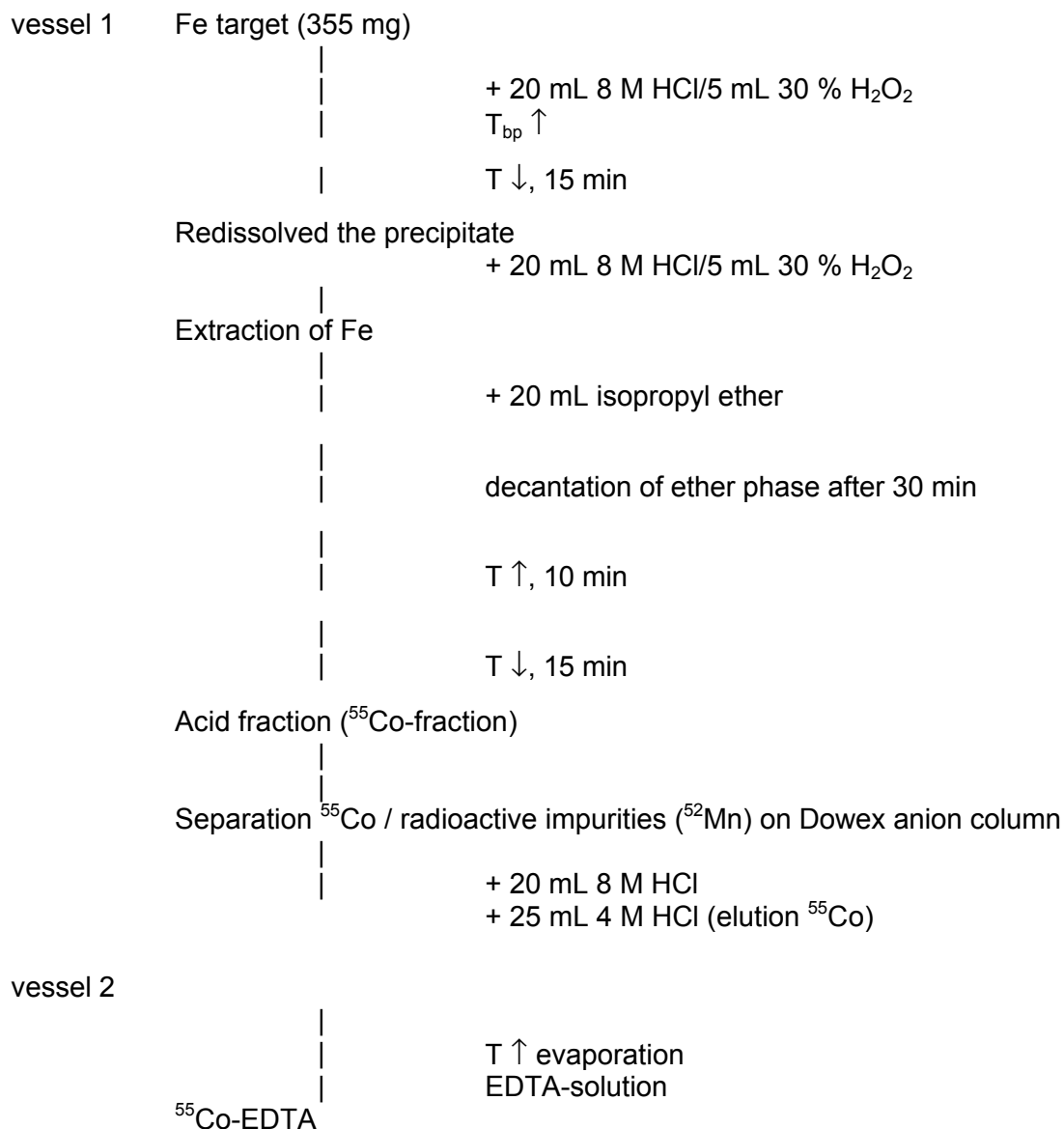
The PET study was performed as follows and the results are visualized in Fig. 2b to 2f

- transmission scan (Fig. 2b)
- consecutive emission scans of the dynamic experiments at intervals of ca. 1 h during the flooding of the core with  $^{55}\text{Co}$  EDTA labeled water (Fig. 2c to 2f).

In Fig. 3 the distributions showing the axial deviation of the tracer from its starting position after 4 time intervals. Those experiments are intensified and clearly show that PET allows in-situ monitoring of physical processes.

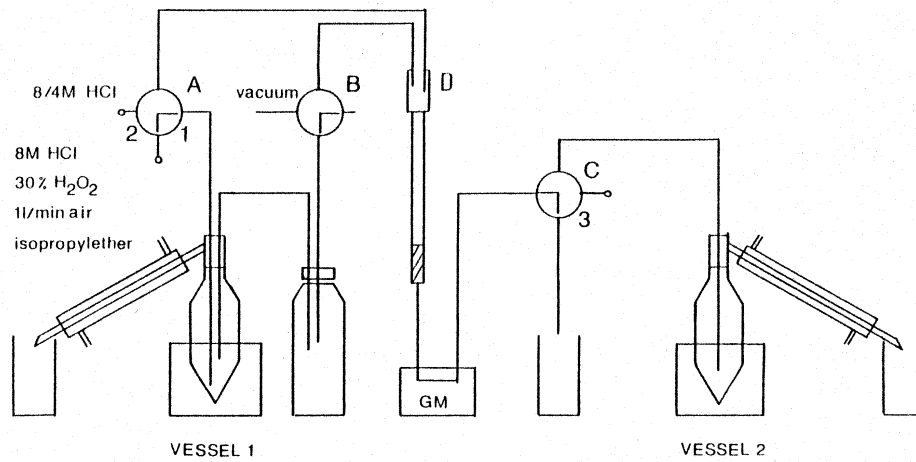


**Scheme 1: Purification of  $^{55}\text{Co}$**

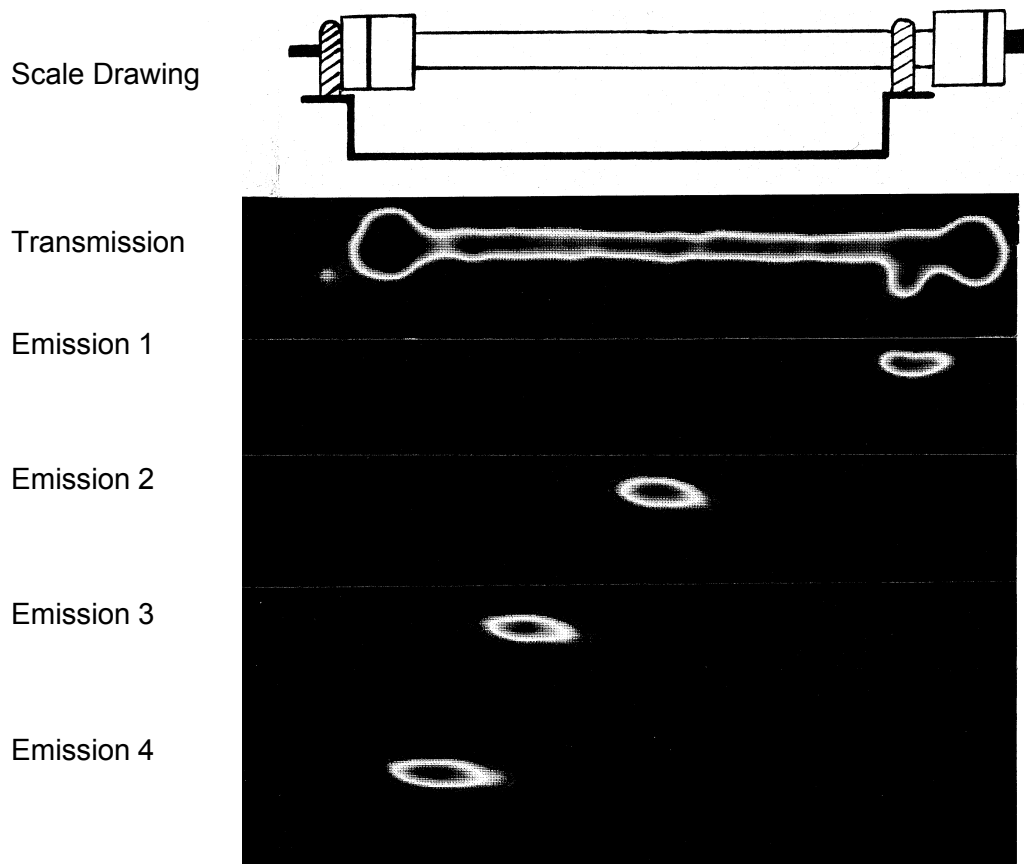


**References:**

- [1] E. Van Den Bergen, G. Jonkers, K. Strijckmans and P. Goethals, Industrial applications of positron emission tomography, Nucl. Geophy. **3**: 407-418 (1989)
- [2] G. Jonkers, K Vonkeman, S. van der Wal, R. van Santen, Surface catalysis studied by in-situ positron emission, Nature **355** (1992)



**Fig. 1:** Technical set-up for the Separation of  $^{55}\text{Co}$ ; A, B, C valves, D: chromatographic column filled with anion exchange resin.



**Fig. 2:** Positron Emission images of the displacement of  $^{55}\text{Co}$ -EDTA in sand core