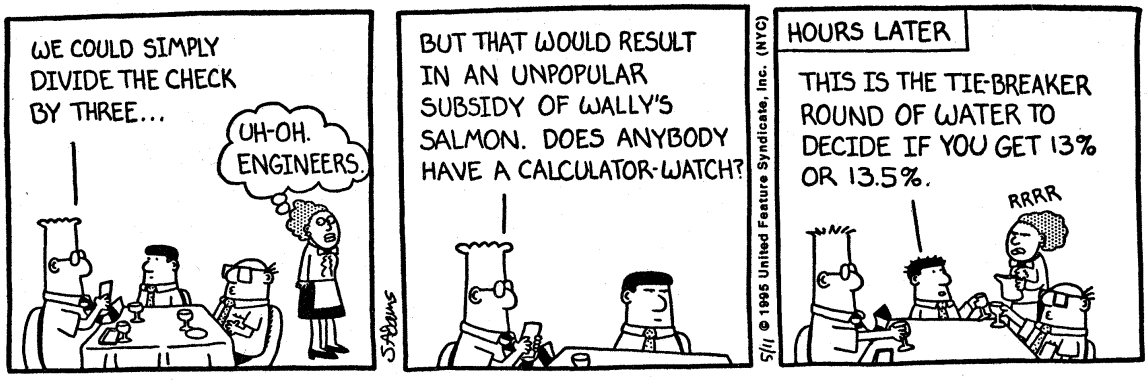
Second Session

High Beam Currents
In-Target Production of Precursors

Co-Chairs: Jean-Luc Morelle
Robert J. Nickles



Karl Erdman, Ebco Technologies, Inc.: Ebco Target Design and Experience

I would like to make a few comments and then give a short description of our experience with targets. The first one is that there are two problems with these small machines. The first one is the dE/dx problem which is fierce for targets. If you consider a water target for example, assume that we use O-18 water to produce F-18, the range of a proton in that target is not very large, it's less than 2 mm. If you 're putting in 50 µA for example into such a target, you're dumping 500 watts into something like 0.2 mL of water. The temperature is going up at a rate off 500 deg/second in the region in which you are dumping in the power. So the real question is, how do you get the heat out ? There have been some solutions proposed in which you just make the target bigger so you distribute the bubbles that you are forming over a bigger volume. But there is no way of getting the heat out by conductivity because the conductivity of the water is very poor. If you wanted to get the heat out by conductivity, the temperature would have to be 2,500 °C. So there' a real problem of getting the heat out of targets. As soon as you make solid targets that also have lousy conductivity, you're faced with the same problem. You get fracturing, spallation, all kinds of things happen. So there's a real limit at low energies as to what kind of beam currents you can put on targets. On our small cyclotrons, we use a titanium target because titanium somehow or other it doesn't get contaminated. We have a target in Korea that has been operated now for two years. It has not been cleaned and the production rate has not fallen off. It's a solid titanium target, but we can't put more than 25 µA onto it because the temperature rise is too great if you do that. Remember, at 250 °C you are already at 500 psi inside that target, that's the vapor pressure, so unless you've designed this thing properly you are in real trouble. We can illuminate this target pretty uniformly because the beam from our machine is in fact pretty nice. If you have a beam which is a line or something it's not so easy. This actually shows the profile of the beam out of our machine in the two directions. So we have a very nice Gaussian profile which you can actually clip with collimators. What you really want to have is a fairly big beam, as soon as you increase the current you increase the target area. You also have to figure out how you get the heat out. That depends on the kind of target you have. If you have a metallic target that has reasonable conductivity, you don't have this problem.

Small machines are in fact able to produce such things as palladium if you can make a metallic target which is conductive because in fact you can get big currents out of the machine. This is typically the current we can get out of our small cyclotron, extracted current at 19 MeV, you can get some increase in current by bunching in here, but essentially if you go way up here you start to get space charge limited, you can't get any more beam out of these machines by doing bunching. However, we have been able to get over 3 mA of beam extracted from the system. So this does mean that these small machines can be used for doing such things as palladium. They could run over night as a palladium producer and still produce your PET in that system because a negative ion cyclotron with an external ion source is capable of producing more beam power than you can ever absorb in a target. We are no longer limited by currents from cyclotrons, the problem is in the targets. And this is where the work has to be done on the systems.

Discussion:

Q: J. Clark: Is anybody trying to move the water through these targets?

A: It turns out you can't get the velocity up high enough . If you remember, some years ago Yves Jongen tried to pump water through. He was using 10 mL and tried to circulate it. But if you calculate anything having to do with velocity it's impossible to move it fast enough. 500 degrees per second for this little volume that you have inside is difficult. It's not to say you

can't do it but usually you contaminate it if you start to put it through a pump. It's highly radioactive.
The other thing that happens is you are pouring in, let's assume you're putting in 50 µA into the system, you're dumping an awful lot of hydrogen into there. Hydrogen ions enter the solution which is stopping in the middle of the solution. So the pH drops phantastically, so you have a highly acid solution which is attacking the target under those conditions. And it's extremely difficult to circulate from the outside.

J.-L. Morelle: Heat Transfer Problems

What I had tried, I actually never went to making a commercial one. The problem is that when you want to recirculate a very small amount of water because you want to reduce the volume anyway for ammonia, for F-18 for instance, the problem is that you have to calculate on your heat exchanger somewhere. And your heat exchanger, the simplest one you can imagine, is having a little tube which will circulate the water across. But if you want to minimize the volumes, you see that you have the right exchange, you need high velocities to have correct exchange between the water temperature and the wall of the tube. Which means that you finally need quite a water pressure drop across the system..

And the second thing that has to be calculated is the optimal wall thickness of the tubing and the heat conductivity of the metal you use for your heat exchanger tubing. Actually there is an optimum, I make a little drawing, although I have no figures calculated. When you are working on a heat exchanger (explains drawing). So you see that this area is much larger than this one. So you like to increase the thickness of the tube of your heat exchanger but at the same time you increase the distance, that means you reduce the gradient. So for a given material there is an optimum for the ratio of internal and external diameter. And that's something that can be calculated. The problem is that ...(inaudible, background noise) there are many constraints that are difficult to optimize.

...rebuilt a main core transformer for cyclotrons with big copper bars around it so that I could heat up a little sample, it was in half currents up to 5000 A under very low voltage. So I built little test tubes and put some nickel around because it has a good electrical resistance and a rather high heat conductivity with something like 6 mm internal diameter and almost the same external diameter and put the current through there so you could have a rather homogenous source of heating on a tube with a perfect knowledge of the amount of heat that we pumped in that small piece of tube. And then what we did was to measure the difference of temperature to have confirmation also that the power was matched, and measured the external temperature of the little nickel tubing by putting clamps on the tube so that we can have a good measurement of the external temperature of he tube. Then using the heat conductivity equations but in the cylindrical coordinate changes you can calculate internal surface temperature of the tube and knowing the temperature of the water and that inner temperature of the metal. We could define curves that could be used to design the target itself. And my idea was to try to work in a regime which is not the laminar or usual flow. Under boiling conditions this can increase tremendously the amount of energy you get out of the target per cm^2. And this is roughly the figures we got, you have here the temperature in degrees centigrade and you have here the power in kW per cm^2. And that's the way the current increases, at a given moment you have a sharp bend in the curve and the curve goes up, which means that for a very small increase in temperature we have a very high increase, practically 1 kW per cm^2 increase in heat removed. I have looked at reactor experiments and that data on the subject and the funny thing was that I couldn't find a very good matching between those experiments that had been done. Here I had working at up to 10,15 atmospheres at high velocities and I was not able to really put in between the 5 and 15 bar. You see a variation of this curve but in the curve of the reactor experiments they had

pressures from 50 bars to 300 bars. So that's very different conditions. And actually what hides behind that is that on the heated surface, where there was rather fast flow of water, we had little bubbles of vapor that appear. And of course at high pressure the idea is that the vapor can be contained, evaporated water can be contained in a smaller volume. But especially in the nuclear boiling phenonema what happens is that the bubbles appear at the surface and than they collaps and they create an increase of turbulence near the surface which also increases the heat exchange. And what we notice as the most important factor was the velocity. So this is the curve for 27 m/s it's a tremendous velocity for water in a channel. So we chose it because the pressure drop was corresponding to the pumps we could find. So that gives you an idea. The point in the curve here was the burnout point, it means that point where we have a thin volume and then the thing collapses at once. Not in the cyclotron, just heated up electrically.

So that was the first point in having some data and what was really the degree of changes you can get. The other thing was to see what was happening with the target. So the target was at a very shallow angle. We tried to spread out the beam as much as we could give it to the dimensions of the machine. This is a cross section view of the target, with the target is actually 20 cm long altogether, of which 15 is used for the beam. The beam current is routinely 1.2 mA measured on the target, but actually the real current is much higher because there is some scattering of the target. And that can even be measured by temperature measurements between in and out, and also by RF consumption measurements. However, the shape of the target, this is a water channel, this is the target, a copper plate and on the copper plate there is an electroplated layer of Rhodium. This represents the beam current profile and the idea was to have a profile which matched the profile of the isothermal lines, because as you saw on the previous curve what you should avoid when you got to the very limits of heat exchange between the water and the solid, it's to avoid that you have one point where thin boiling starts up because at that point the cooling becomes worse, so the temperature increases near by and so the thin boiling extends in a few seconds up to the whole piece. So you should try to have the heat exchange along an isothermal line, so you can calculate for a given distribution. That means you have a different distribution of power for each point across the target. You can calculate rather easily by even superposition what would be isothermal lines and there we have a water channel that follows this line. Of course this would be done because we are speaking, we are working with a very high melting point product, this is the rhodium. Of course that way of reason can not be applied for instance to targets for thallium and things like that, there it's another problem.

Here you can get a rate at the highest amount of power generated in a very small area by diffusing that power on a bigger water surface. We're trying to keep that surface as cold as possible. Now here you have ideas according to calculations ...(inaudible).. 600 °C. This ...(inaudible).. it was much too wide actually, the beam was really concentrated on a line in the middle. Now we have been testing that up to 1.8 mA of measured beam current. It should have corresponded to something close to 3 mA of current actually hitting the target. Now another interesting figure was the back scattering. Here it's roughly 30 %, 20 %, 10 %. This is the back scattered current depending on the number we gridded for 14 MeV protons hitting rhodium. And as you see when the ... (inaudible)...goes up you have a large amount, a large percentage and in this case the percentage was actually beyond 50 %. But in the case of an internal beam target as we have it, there is another figure, I don't have it unfortunately any written data available now. It's seeing what is the distribution once you choose a given angle, what is the distribution of the angle that are knocked out? So you have the beam that comes in your very grazing angle target, and then a great amount of that percentage of lost beam goes away at a very shallow angle also. The curves can show that. And then the cyclotrons which were running what was happening is that the part of the beam was knocked off, it was in the magnetic field, was coming back after an extra turn but what had happened in the average is that the center of the circle had been slightly shifted and it was hitting the leading

edge of the target. So what we did, we already had the very angular shape matching the beam and then we tilted the target down so that the beam, that the second turn could come back and the target could recuperate a bit more current, and also which was a matter of reducing the activation of the machine. Now in case of the machine we reduced mA amounts of current. The problem of the shielding becomes very important and although it's on a totally settled program what we had done was to shield all the parts of the machine with a ¼ inch pure aluminum sheets that were cooled by channels, so it's large area to dissipate the power. And these shields were actually clamped on the machine which could rather easily be removed in such a way that when those shields were removed the machine was not too badly active. It still was very high activation, but after two weeks of stop you could work in the machine to make repairs if they were needed. Now that thing has been operating for five years and the company that produces Pd-103 commercially is Theragenix. I know they are operating five cyclotrons now and they still have a back order of three. So that means within a few years eight machines working day in day out at a current of 1.2 mA.

Q: T. Ruth: Jean-Luc, looking through these modern machines H minus where you get the beam out - why would you put a target inside rather than outside?

A: As I told you, when the decision had to be taken at IBA, that company came and said we are relying on the reactor now. The reactor will be shut down fifteen months from now and we need a solution. And at that time, it was the end of '91, we were not in a position of being able to guarantee over 1 mA of extracted beam for the negative ion machine. Although there were some experiments, there was a program doing it. And we had been a bit taken short by the demand, because the cyclotron companies were not really eager to show that they could produce 1 or more mA with a negative ion machine because it would have killed part of their market, on the radiopharmaceutical market. So when they came with that question, they also came with requirements for low costs etc and we shouldn't forget that the 18-9 was practically...the magnetic structure that was such that it was not very stable from the acceleration point of view. Nowadays it would be worthwhile considering to extract the beam.

C: J. Nickles: Just to follow up on that. To give you an idea of the scale of this thing. I'm told by radiologists that I work with that while there are 7000 of these procedures done a year now it's projected in the next ten years to go up to 200 000 procedures a year. And each of these procedures involve 100-150 seeds each of which is 2 mCi. So each patient is carrying 300mCi of material, 200,000 of these patients a year, that comes up to 60 kCi of Pd-103 that's got to be made every year. That's going to take dozens of 1 mA machines making this material all the time. And so the economics of it, it's a tight market because it's held down by I-125 to about $10 per mCi. But that's still enough to provide a million-dollar budget to a cyclotron that wants to work at night. And that can make a mA of beam. The funny thing is, we all have that machine. You turn the pole phases around, we run a RDS positive and with the rhodium target on port 4 sliding it in at night and I think we probably could be making a dent. There's four dozens, five dozens out there.

C: J.-L. Morelle: What you need is an external line source.

C: J. Nickles: Maybe in a year or two years I can tell you 'No'. The economics are hard, because of the I-125 problem, but it's something you might want to consider, certainly want to keep you eye on, because it's an infinite sink of beam.

C: K. Erdman: I just want to make a comment about that heat transfer. The data on heat transfer comes from power transmitting tubes which work preferably in the same pressure range that you are talking about. In terms of power dissipated on the anodes, there's all kinds of data from tube manufacturers ... like RCA, General Electric with these kinds of curves fit.

So people who are interested in getting data over various pressure ranges and so you get it from the radiotube manufacturers, waterproof tubes.

C: J.-L. Morelle: It's a good point. Actually when I did that I had not realized that and when I got my results I was discussing with people that have run a little company nearby in Belgium that produces X-ray inspection systems and we were talking over that's interesting data for us because we have to try to make X-ray tubes smaller and we have to work at those problems. Actually that's right. That data existed, but I didn't know where to actually find it.

Q: Before we really considered this optimum economic running of positive ions in our beautiful machines at night: Can you tell me, do you get any kind of early warning if this cooling is failing? I mean if you get evaporation, if you get gas, water vapor in that, and the cooling fails, do you fill the cyclotron, the ion source with water ?
Nickles: Jean-Luc, is there any shrieking or any acoustic thing?

C:J.-L. Morelle: Yes, first of all I come back to the curve, because that curve shows something. We are working roughly at this point. That shows you have a very strong reserve here for a very small increase in temperature of your parts. It's a very stable operation regime. Now as far as the testing we did during the early trials before we got confident in the system. We had various mirrors for direct view and a camera and were looking at the target constantly. That was during the first weeks of operation, because there's many cameras, because of the neutron flux in the room. That's how we did it and once we trusted the system we didn't worry about that any more.

Q: Alignment of the target.... ?

A: Extremely sensitive. It has been tried of course, when I mentioned the fact we were at 0.6, 0.7 degrees. It's of course extremely sensitive and by having that camera system that you can use to validate the shape because what we noticed is for one given cyclotron once you have the shape, it remains that shape. But to validate it we have of course a numeric machine calculated curve which we modified little by little and then we had the right curvature. And that we could do using the camera. What we did was to take copper backings, sputter aluminum oxide over them, so that they would make light when there was a beam on it. And then to simulate high beam current conditions we were bunching the beam at 1% time 10 ms then off and then 10 ms, so that we could have after 10 ms we could show we were at steady state as far as the RF system was concerned. And using that bunching system we could simulate high current although operating at low average power. And we could do various experimental targets at various angles until we had the right angle. That's how we proceeded, experimentally, one by one. Actually the first target we designed was a target mounted on a shaft and we had this screw and we could turn the screw and each time we took a picture. I even have bad photocopies of the experiments here. And once we had the right shape, we were happy with it. We also noticed that we could place slightly on the shape by tuning the dee voltage. So we had some fine tuning with the dee voltage. And that was what we tried.

C: M. Nortier: Maybe I should just make a comment on the nuclear boiling regime, the cooling method. The point at which the normal forced convection regime goes over into the other regime is very dependent on the surface, the quality of the cooling channel. So you are going to make a lot of targets, it's important that you can control that actually.

Q: You were killing cameras. What kind of cameras are you using? CCD?

A: Morelle: Yes, we used CCDs because the magnetic field in the vault is such that you can not use other cameras than CCDs.

Experience with Curved Natural Bismuth Internal Targets for Production of Astatine-211 via the $^{209}Bi(\alpha,2n)^{211}At$ Reaction

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Recent preclinical and clinical [1] work suggests that the 7.21 hour α-emitter ^{211}At may soon be evaluated clinically in combination with a variety of tumor-specific carrier molecules. A novel high-performance internal beam target system (Model MIT-1, Cyclotron Inc., Napa, CA) was installed on the Duke CS-30 cyclotron in 1994 to produce sufficient ^{211}At for preclinical and clinical studies in parallel with the growing radionuclide production requirements for clinical positron emission tomography. First-generation targetry and distillation equipment [2,3] has been steadily modified during a series of 128 production runs of 60-90 minute duration at beam currents between 30 and 85 μA. This experience has improved production and recovery through evaluation of single and two-piece target assemblies, Cu and Al target substrates, and four Bi target material coating methods (brush electrode plating, jet vapor deposition, hand-applied molten Bi, and computer controlled milling).

The internal target system is shown in Fig 1 where it is installed on the cyclotron in the optically favorable mid-valley position formerly occupied by the relocated beam probe. The preexisting external beam system indicates the radionuclides produced on the seven target stations. The target is installed and removed manually through vacuum lock ports which are controlled by a panel on the cabinet supporting the radial and angle drives at the end of the ram carriage. Water cooling, blow-down, venting, and roughing pump switches are also on this panel, as well as ram radial and angle drive controls for testing and alignment.

The most recent target face design is shown in Fig. 2 with carbon edge monitors for measuring leading and trailing beam current. The Al alloy target face ($75 US machining cost) has a 1.5 by 10.2 cm beam strike with 81 cm radius to spread the incident alpha beam between the monitors. The angle between the face and the beam can be controlled by moving the ram carriage about the carriage pivot point. An Al clamping block presses the face against an O-ring on the back side where six 0.7 mm slots channel high velocity cooling water. Heat transfer has proven adequate to prevent melting Bi at the maximum available 95 μA beam current.

Bi coatings ranging from 7 to 130 μm have been tested. Two promising Bi coating methods are machined hand-coating and jet vapor deposition (JVD). They are being evaluated using scanning electron microscopy (SEM) and quantitative energy dispersive x-ray analysis (EDX). The hand method consists of melting Bi onto the surface of a heated horizontal Al target substrate and scraping the surface to remove the Al_2O_3 and form a Bi-Al alloy at the interface, resulting in a high melting point (500-800°C) bond with good mechanical and thermal characteristics. Some solid slag is produced in this process, which raises a concern that there may be some alloy present in the coating above the interface. EDX has identified Bi and Al in the slag and at the interface surface exposed by chipping off small areas of coating. Recently we have begun to investigate alloy in the coating after observing more difficult melting and decreased distillation yields in thin (10 μm) coatings. SEM, EDX, and beam testing are scheduled for several thicknesses of uniformly milled target faces of this type.

The JVD coating method [4] offers advantages of very uniform coating thickness, automated fabrication, and the possibility of good bonding to Al through removal of oxide by ion

bombardment prior to application of Bi. Initial JVD coatings were found to have a columnar structure of about 75 % theoretical density suggesting porosity with opportunity for oxidation of Bi through increased surface area. These coatings were observed to chip easily indicating a weaker bond than the hand-coated method, and EDX indicated some presence of oxygen. The process was improved by correcting vacuum leaks in the JVD system, improving the Bi vapor source, and heating the substrate during deposition to improve density and bonding. Several improved target faces were recently received and are scheduled for SEM, EDX, and beam testing. An initial thin JVD Bi layer applied above the melting point may result in an alloy bond.

Beam distribution autoradiographs of target faces on Polaroid Type 55 sheet film are shown in Fig. 3. ^{211}At x-rays penetrate the film cover which is thick enough to stop the alpha emission (6.8 MeV average). Calculated average Bi thickness for the three images (top to bottom) are 20, 25, and 36 μm. The top line of the numbers to the right of the images indicates the lead/target/trail currents. The lower line indicates probe current and bombardment time. Observe the uniformity of the JVD coating as indicated by a smooth broad image, compared with the slight waviness in the other two images. Although the Al substrate radius is the same in the top two images, the radius of the Bi surface in the middle image is significantly greater because of gravity pooling causing a thicker coating in the center. We expect the uniform thickness of milled hand-coated targets to give images like the JVD example. The lower flat target face clearly demonstrates the advantage of the curved target face. Using the angle drive, it was possible to scan the beam strike from one monitor to the other or find a position in the middle where no current appeared on either monitor. Target radii from 71 through 162 cm and a 162/81 cm compound radius have been fabricated and are scheduled for testing.

Tuning for bombardment starts by efficient external extraction of a 30 MeV internal 30 μA beam at 19 kV dee voltage. Harmonic coils are not adjusted from this point on. The probe is inserted 1.25 cm from extraction radius to obtain 28 MeV, dee voltage is increased to 22 kV to diffuse the beam, and arc current is increased to produce the 50 μA now used routinely. Using the remote controls and beam meters at the cyclotron console, the internal target is then inserted until beam appears. The probe is withdrawn and the angle drive is adjusted for peak target current with both monitors showing beam. Digital displays of radial and angle drive are reproducible and accurate to within 0.5 mm. Note in Fig. 3 that the sum of the target and monitor currents is about 75 % of the probe current due to multiple scattering of alphas out of the target face, confirmed by Monte Carlo computer modeling of grazing angle flat targets (exiting α energies observed to be less than 20 MeV). Computed scatter loss is significant for Bi but not for Al, which was confirmed experimentally by running bare Al target faces.

^{211}At yields range from 30-40 MBq/μAh due to the variation of the many parameters discussed above. Tuning variations result from the frequent daily particle changes between protons, deuterons, and alphas required by a busy clinical schedule. The multi-purpose ion source is removed for cathode replacement every two to three weeks, and reinstallation alters physical location which changes the beam platter and radial energy profile. We plan to initiate target assays for ^{210}At gamma rays as a function of radial ram position to use as an energy threshold measurement indicating 28 MeV, and to see if it varies with some of the parameters we suspect have an effect.

We routinely produce ^{211}At at 2 GBq/h, which is adequate to support initial clinical trials. Beam current and bombardment times can be increased. We will continue exploring concepts with potential for cost-effective commercial production and distribution of ^{211}At.

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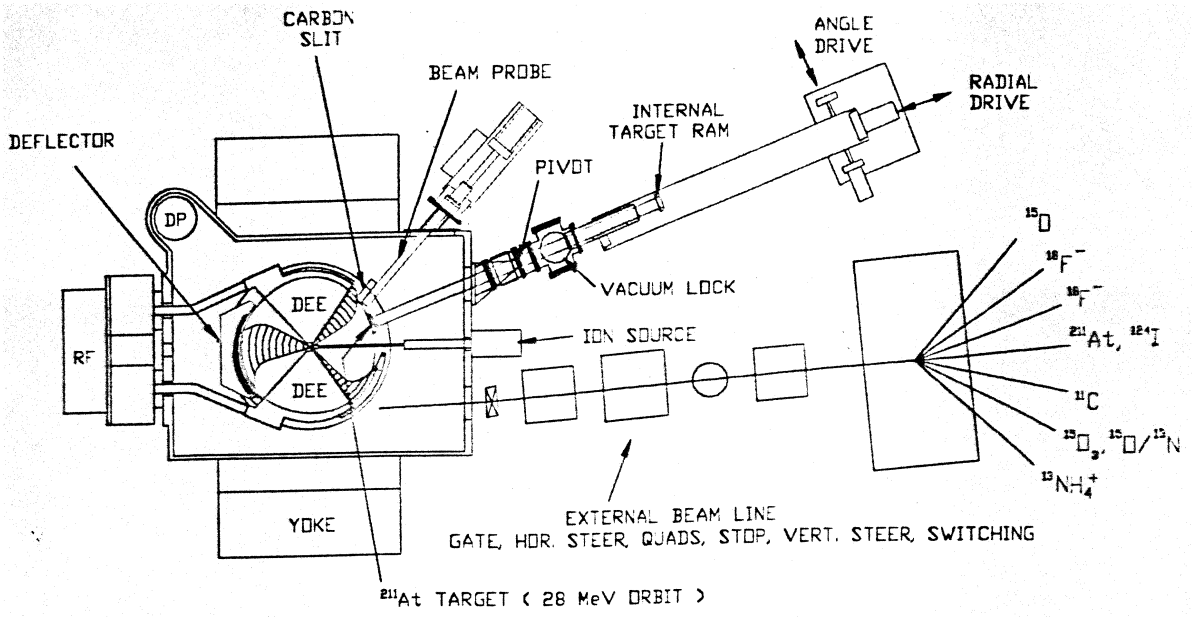

Fig. 1: Plan view of CS-30 cyclotron with internal target system and external beam lines.

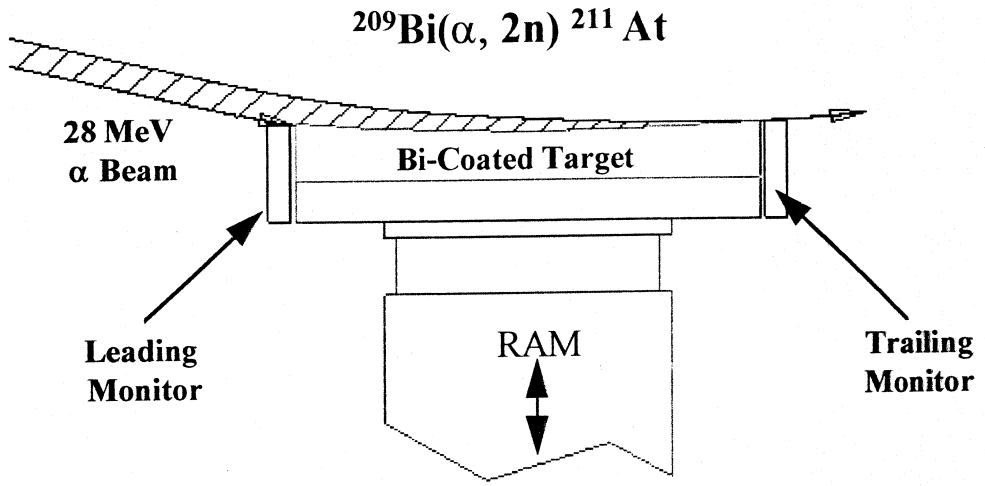

Fig. 2: Top view of 81.3 cm radius target face mounted on end of ram with edge monitors.

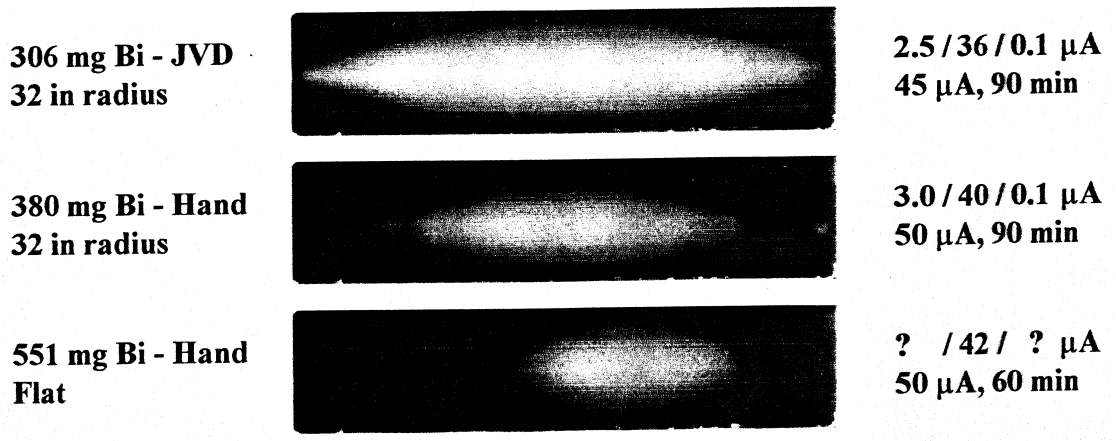

Fig. 3: ^{211}At autoradiographs of vertical target faces as viewed from center of cyclotron.

Discussion:

Q: Bruce, two quick questions. Do you have any volatilization of the astatine?

A: The only time we have seen any melting at all was when a cyclotron operator got a little carried away and violated our rule of not changing the coils. There are two carbon slits which let you ... protect the dees. And I thought there is a little bit too much current on one of the carbon slits, so we adjusted the coils to remedy that and ended up getting a melted area on the target. But other than that we haven't as long as you stick to our procedure of tuning for external beam, then turning up the dee voltage to fuzz out the beam, we haven't any problems with melting.

Q: You could go right into the machine to fix it if you had to, after having made a batch of astatine ?

A: Well there's nothing to fix because the target, it couples out into the vacuum vault. We have tried at the inside of our machine, if that's what you mean.

Q: Michael?: You don't sputter astatine out from the tank at all, I mean an alpha-emitter in your cyclotron tank?

A: There are a lot of nasty things in our cyclotron tank, which completely mask anything we do with astatine. But at low current and high current we still get about the same target yield. And the melting point of bismuth is 270 degrees centigrade, which is of course quite a constraint in terms of keeping the target cool. There was actually the question about sputtering: our target surface looks very clean and shiny before the run and it looks very clean and shiny after the run, so that's no evidence that you sputter.

Q: The point is, you don't get any other alpha-emitters. If you got alpha-emitters, it must be in the astatine.

A: Right. In other words, do we do wipes? No, we haven't.

Q: N. Stevenson: There is an alternate way to make astatine. At TRIUMF we tried to do it and we were actually successful. You need to do a spallation, so we did a spallation on a thorium target. And we did that experiment, it's at 70 MeV. We measured the cross section, figured out we could make several Ci of it, not of the astatine, of the generator, radon-211, which has something like a 16 hour half-life, I think, it's rather a short half-life generator, but a generator it is. It would be transportable as the gas, with the astatine depositing around the outside of the walls, maybe it has to be cooled with nitrogen. The trouble with that setup is that we process alpha emitters.

Q: When you start the humanized trials, what level of activity will one administered dose be roughly?

A: Five mCi I think is what is planned.

C: R. Ferrieri: Just a comment on the sputtering of the bismuth. I think sputtering is not dependent on thermal stability of the matrix. It's a high energy process. I've done sputtering on metal oxides, which are thermally stable to 1200, 1500 degrees C, and with a lot of material. So that's a big awarenes now, a big concern.

Methane Production in Small Volume High Pressure Gas Targets

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Recent developments in the production of ^{11}C methyl iodide via a gas phase reaction reported by both Larsen et al. [1] and Link et al. [2] at the last ISRP and WTTC meetings prompted us to look at the production of methane, CH_4, from the irradiation of a gas mixture of N_2 and H_2. We felt the direct production of ^{11}C-CH_4 in target was the most elegant implementation of the gas phase reaction for routine use of ^{11}C-CH_3I. However unexpectedly poor yields of ^{11}C-CH_4 from our small volume high pressure gas targets lead us to further investigate the conditions required for efficient production of methane. We routinely produced several hundred millicuries of ^{11}C-CO_2 in a 10 mL STP target, open to a 20 mL expansion volume, operated with an initial pressure of 400 psi at 15 μA, 13 MeV protons on our TR13 cyclotron. Similar operating conditions with a gas mixture of N_2/5%H_2 yielded only 10's of millicuries of ^{11}C-CH_4.

Investigation of other targets reported in the literature used to produce methane indicated that they tended to be large volume targets (flow through design in some cases) operated at comparatively low pressures with 1 to 5 % hydrogen content. Christman et al. [3] reported on a flow through design employing 5 % H_2 where they noted that 1 % H_2 resulted in 50 % of the methane production compared to 5 % H_2 and that at least 0.1 eV/molecule was necessary for the decomposition of formed products to CH_4. They also noted the production of ammonia in target.

The first target we used for the production of methane was a 1.0 cm diameter cylinder, 10 cm long. Target valves mounted on the target body restricted the gas volume to 10 mL STP, virtually all encompassed in the beam strike region. This target configuration, when filled with 300 psi N_2/5%H_2 would deliver 70 mCi at EOB following a 15 μA/ 5 min irradiation. Longer irradiations would result in a lower saturated yield. Running with one target valve open to allow an expansion volume of ~30 mL decreased saturated yields for short runs but did not change saturation yields for longer irradiations. Examination of saturated yields as a function of irradiation time suggest a competing process.

We believe that our target conditions lead to excessive production of ammonia, NH_3, which consumes the H_2 and limits the formation of CH_4 in the target. Measurements of the decay of the radiation field in the vicinity of the production target indicate that ^{11}C is formed in the target but not carried out in the gas stream when the target is emptied. Saturation yields for $^{11}CO_2$ production are well in excess of those achieved for $^{11}CH_4$ production. Modifications of the target to a conical design with an expansion volume outside the beam strike region, and an increase of the H_2 concentration to 10 % have resulted in the achievement of acceptable saturation yields.

Table of Saturated Yields:

Prod.	Target Configuration	Gas Compo-sition	Beam Current [μA]	Irradiation Time [min]	Initial Pressure [psi]	Beam on Pressure [psi]	Y_{SAT} [mCi/μA]
$^{11}CO_2$	1 cm Ø cyl. 10 mL + 20 mL exp.	99.5 % N_2/ 0.5 % O_2	15	5	340	407	89
			25	5	340	416	67
$^{11}CH_4$	1 cm Ø cyl. 10 mL	95 % N_2/ 5 % H_2	15	5	304	548	27
			15	15	310	530	17
$^{11}CH_4$	1 cm Ø cyl. 10 mL + ~30 mL exp.		15	5	300	322	19
			15	15	310	330	17
$^{11}CH_4$	1-2 cm Ø cone. 24 mL + 20 mL exp.	95 % N_2/ 5 % H_2	10	5	297	345	60
			15	5	304	365	40
			15	10	302	363	32
			15	15	298	357	30
$^{11}CH_4$		90 % N_2/ 10 % H_2	25	5	302	372	49
			25	15	302	372	48
$^{11}CH_4$		85 % N_2/ 15 % H_2	25	15	299	340	53
$^{11}CH_4$		80 % N_2/ 20 % H_2	25	15	297	328	43

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[1] P. Larsen, J. Ulin, and K. Dahlstrom, A New Method for Production of ^{11}C-labelled Methyl Iodide from ^{11}C-Methane, Journal of Labelled Compounds and Radiopharmaceuticals, Vol. XXXVII, pp. 73-75 (1995)

[2] J.M. Link, J.C. Clark, P. Larsen, K.A. Krohn, Production of [^{11}C]Methyl Iodide by Reaction of $^{11}CH_4$ with I_2, Journal of Labelled Compounds and Radiopharmaceuticals, Vol. XXXVII, pp.76-78 (1995)

[3] D.R. Christman et alt, The Production of Ultra High Activity ^{11}C-labeled Hydrogen Cyanide, Carbon Dioxide, Carbon Monoxide and Methane via the $^{14}N(p,\alpha)^{11}C$ Reaction, International Journal of Applied Radiation and Isotopes, Vol. 26, pp. 435-442 (1975)

Discussion:

C: R. Ferrieri: At PSI I reported on the effect of CO_2 specific activity as the specific activity went up, the target output on CO_2 went down. So when you had a dirty target the specific activity is in the order of 1 Ci per μmol. You got a hundred percent of theory out. As you increase to, say, 13 Ci per μmol which is the maximum we were able to shoot. This is with the conical target, very similar to what you mentioned your design there it dropped to about 45 % there, for whatever reason. And I actually use this as a diagnostic, when we opened the target to change the front target window, I use the output of the target from a series of one minute irradiations and as that decreases to a certain steady state level, I assume the target is now clean and putting out a high specific activity. But since according what you say you see the same phenomenon with a similar shape target with C-11 methane as well. I think you are right, there are some surface phenomena going on there, that seem to glum it up. It doesn't matter what the chemical state of the C-11 is. And it's probably just getting caught up in the aluminum surface.

C: T. Ruth: ... effect of some kind in the aluminum matrix.

Q: M. Welch: (hardly audible) ...reaction of C-11 with nitrogen to form cyanide, with methane to form cyanide, methyl amine and some imine, as I have it in my memory, did you see any of those ? Would they come off that GC column ?

A: But we don't see any radioactivity sticking anywhere on the column. We looked for activity elsewhere other than what was coming off. They could be formed and than not coming out in the gas phase.

Q: T. Tewson: Tom, what was the percentage conversion to ammonia, in other terms did you get the same value because you used all the hydrogen up?

A: No, I don't think we were using all the hydrogen up. I'm not quite sure why in the beginning we calculated the number of mmols of hydrogen left. We were still like an order of magnitude higher (hydrogen than methane) with the ammonia we were producing. We tried to look at the kinematics that way and that's not the answer. I don't know what the answer is because there's more hydrogen, even at five percent, than is necessary to do both the methane and ammonia. Because the ammonia just was produced and stays flat so it must be some kind of equilibrium reaction that's set up. But the hydrogen is tied up elsewhere. We don't understand that phenomenon at all.

Q: B. Mock: We had the same type of work at CTI target. It was the conical 10-15 mm bore, 10 cm target. I tried five percent hydrogen and had no good recovery at low beam currents and as soon as I got to about 10 μA or greater it fell off to virtually nothing. We did the same thing about two months ago with 10 % hydrogen, same exact results. Great recovery at 5 μA, and then 20 μA it dropped off, at 30 μA we had 4mCi/μ, at 40 μA then less then 1. So with the CTI aluminum body target, same conical design, with the collimator restricting, trying to keep the beam from scrubbing the walls or trying to make it scrubbing the walls, both ways, it does not come out of the target. The other bad side to that is, that after that attempt was tried, we did some nitrogen/oxygen runs for CO_2 production. It took 24 hours until I got my CO_2 back out of that target again. The CO_2 that was being produced was being trapped in the target. The oxygen coating was either destroyed on the target surface, I'm not sure, it took 24 hours to go to get reasonable CO_2 yields back out of having one fill of the 10 % hydrogen target. Have you got any similar observations after running with hydrogen, switching back and forth between hydrogen and oxygen ?

A: Well, we really haven't. We've done a few CO_2 - methane back and forth - Ken ?

C: K. Buckley: When we started we switched back and forth. We have done methane after doing CO_2 runs and vice versa, but I don't think our CO_2 production was terribly affected.

C: Ruth: One point about specific activity. We haven't measured the methane directly, but from our radiochemical reactions we've gotten a size of 5000 mCi/μmol of raclopride after 40 minutes or so. So we were up in the 10-20 Ci per μmol, at least as judged from the final product. And this is fairly consistent.

Q: P. Larsen: Have you tried to change between CO_2 and methane? It seems like if you produced ammonia, and the ammonia stuck in the tubes the CO_2 will also stick in the tubes as a salt or something like that. This might be an explanation. You can see the tubes are getting radioactive.

A: We haven't noticed the tubes themselves are getting radioactive. But Bruce, what I would suggest, a simple experiment might be to put a collimator that's 8 mm hole, just to see if it is

the wall interaction. That would help a little bit, but what we are going to do next is to make a bigger target that the inside volume is slightly bigger.

Q: B. Mock: We've also done it with a target that has a smaller, 6 mm collimator, still the same effect.

A: It just may be that the reaction products are hitting the wall very quickly.

Q: What's the wall material made of?

A: It's aluminum.

Q: J. Nickles: Did you try changing the pressure in the target? Increasing the pressure?

A: Yes, that's what we did. We went from 250 up to 400 and it did not make a lot of difference.

Q: M. Welch (to J. Nickles): Somebody from your place told me that you are going to make methyl iodide from methane. And you want to make methane in the target because the specific activity is higher with methane than with CO_2. Is that true?

A: J. Nickles: We haven't installed the system yet, the system is still coming. But we are hoping to have a higher specific methyl iodide from that system than we do with the CO_2 system.

A: Well, Mike, this is anecdotal, with making raclopride with a CO_2 target using $LiAlH_4$ it's not clear where the problem is. We were struggling to get over 300 or 400 mCi per µmol, going through the gas phase, we're routinely for all of them well over a thousand mCi per µmol.

Welch: The reason I ask is when Joanna was talking to me about our methyl iodide system the reason she said you wanted to start with methane was because your methane in target was higher specific activity than CO_2.

C: That's certainly something that we found at Hammersmith. We've been running methane and getting about two or three times better specific activity than from CO_2. We are part way through trying the methane to methyl iodide process. Hopefully that will be ready for two years time.

C: But our C-11 activity was not different between CO_2 and methane when we tried making methyl iodide out of this.

C: M. Welch: It's interesting. We first got the GE methyl iodide box. Our specific activity wasn't very good. And we went back and looked at our CO_2 , and our CO_2 wasn't very good either, so we cleaned the system out.

C: So, you were just using reagent grade CO_2....

C: T. Ruth: You mean oxygen. That's a real issue too, when you do any of these things you have to be careful with the grade of your mixture, hydrogen or oxygen or whatever. They can really throw you back a long, long time, messing around with impure gases. That was Workshop number one, you are too young to know about that.

C: J. Clark: Can I just make a general comment? You never see a mass spectrometer made from aluminum. And the good reason is, aluminum is shit. Stainless steel, electropolished, but it's a hell of a target.

C: Ruth: We're satisfied with the specific activity as with the total yield.

C: But you are loosing the product, because the wall is so reactive.

A: Yes. It is aluminum oxide, right?!

Remote Controlled In-Target Production of [^{13}N]Ammonia Using a Circulating Target

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[^{13}N]Ammonia is a frequently used radiotracer for the measurement of myocardial blood flow by PET [1]. We report a reliable and reproducible method for the in-target production of [^{13}N]NH_3 by using a circulating target including trapping of the end product in a shielded syringe (bolus delivery system) to be applied for injection in human studies.

Irradiations were performed by the Scanditronix MC-17 with a fixed proton energy 17 MeV beam. The target body is from aluminum (3.5 mL) with 35 mL of circulating water (70 mL/min), the entrance foil is from Havar (0.025 mm). A typical irradiation (1.7 µAh/25 µA) on the circulating target with 1 mM ethanol produces 17 - 21 mCi [^{13}N]ammonia in 10 mL saline (EOS). The advantage of a circulating target over a small volume target is a more efficient cooling of the target so higher beam currents can be used (up to 45 µA). The filling of the target is carried out from outside the vault.

In the literature a concentration of 5 mM ethanol is the common choice to ensure a radiochemical purity of >99 % [2]. In our case 5 mM ethanol in combination with a circulating target led to malfunction of our target circulation pump (TCP) since 97 % of the ethanol is degraded to carbondioxide and small amounts of formaldehyde [3]. To reduce CO_2- production we used 1 mM ethanol. The irradiated 1 mM ethanol is passed through an ion exchange column (Maxi-Clean SAX, Alltech) to remove anionic impurities ([^{18}F]fluoride < 0.1 %, [^{13}N]nitrate/nitrite < 5 %) [2] resulting in a radiochemical purity of [^{13}N]ammonia of > 99 %.

To fill/refill the target system Nupro valves 3, 4, 5 and 6 (figure 1) are opened and the TCP is activated for 3 minutes to ensure removal of air from the circuit before irradiation. During irradiation the route of the circuit is changed by activating valve 2, while valve 3 and 4 are closed. At EOB a part of the ammonia in a fixed volume (loop between valves 5 and 6) is sent to the laboratory by compressed air. Valve 7 and valve 9 are opened while the other valves are closed and the pump stops to circulate. The other part of ammonia is transported to the waste.

The column effluent is collected into a sterile vial, containing 0.25 mL saline (30 %). After two minutes the vent is closed by a pinch valve. By compressed air the isotonic and colorless solution is filtered through a sterile 0.22 µm membrane filter directly filling a 10 mL sterile syringe in a shielded trapping device (weight 6.4 kg). The syringe in the bolus delivery system is shielded with 4 cm lead alloy around. To measure the activity a PIN silicon photodiode is integrated, which signal is translated in mCi on a LCD-display (accuracy > 90 %). The dose rate is < 50 µSv/h measured at the handgrip (10 cm above the shielded syringe).

The whole procedure, controlled by a Siemens PLC, including an irradiation of 4 minutes, is performed within 10 minutes, which allows rapid protocols for clinical use. More than 800 synthesis have been carried out for human studies with a reliability of more than 99 %.

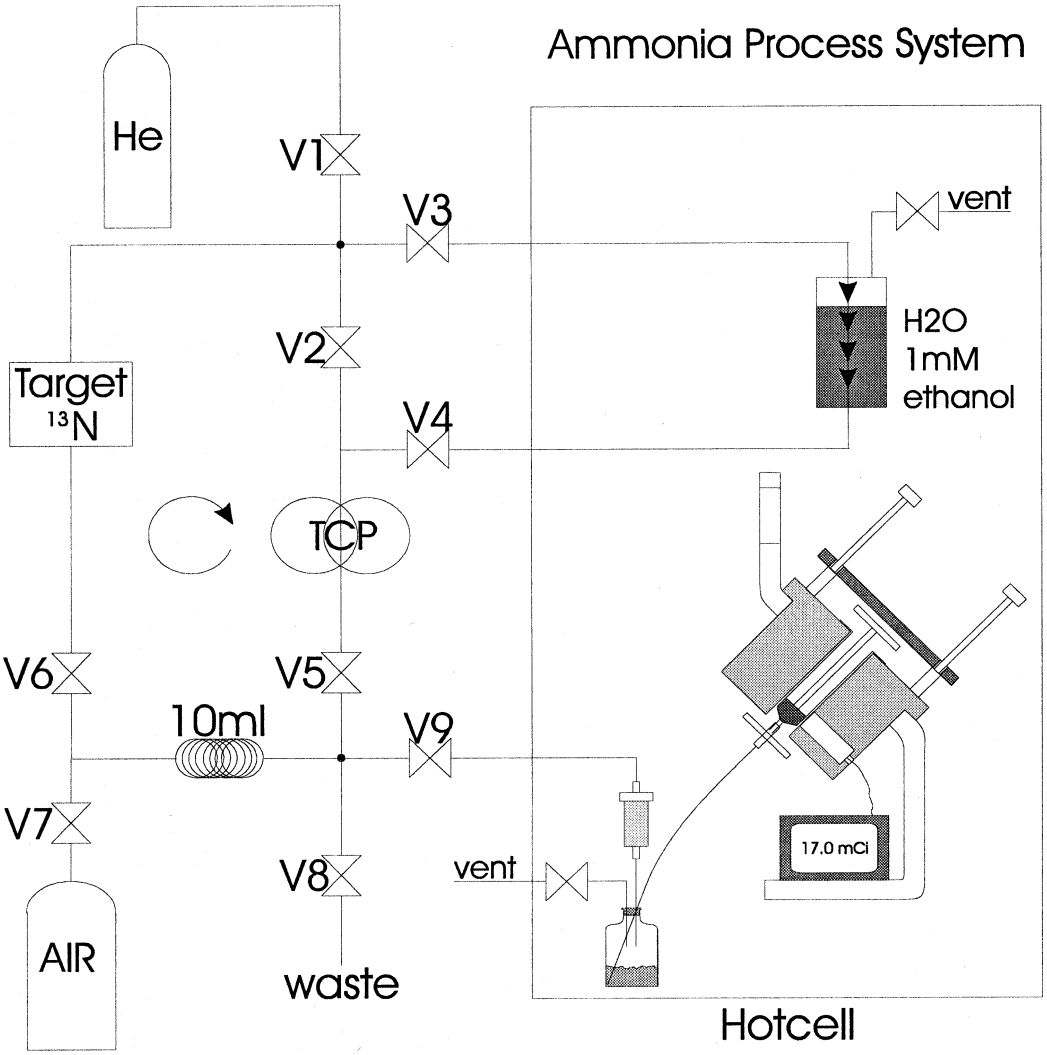

Table 1: Summarized quality control data of [^{13}N]NH_3

radiochemical purity (HPLC)	> 99 %
Al/Ag/Fe (AAS)	< 10 ppm
Ethanol (GC)	< 1 ppm
Bacterial endotoxins (LAL)	< 2.3 IU/mL
Formaldehyde (GC)	< 15 ppm
PH	6-8

References:

[1] H.R. Schelbert, M.E. Phelps, S.C. Huang, N.S. MacDonald, H.W. Hansen, C.F. Selin, Circulation **63**: 1259 (1981)
[2] B. Wieland, G. Bida, H. Padgett, G. Hendry, E. Zippi, G. Kabalka, J.L. Morelle, R. Verbruggen and M. Ghyoot, Appl. Radiot. Isot. **42**: 1095 (1991)
[3] Ferrier et. al., J. Lab. Compd. Radiopharm. **32**: 461 (1993)

Discussion:

Q: J. Clark: ...you left the filter on when you transported it?

A: We prepared the bolus system in a laminar flow hood, that's still our preparation, and then we take it to the hot cell. So the filter is disconnected, before the injection goes to the patient.

C: J. Nickles: We have a punctuation mark in our synthesis of ammonia, because we always got this two or three mile drive between the point of synthesis and the point of application, but in a hundred doses we never had a case where the dose they needed, when they said they would need it, even the minute before...I mean it's always: 'oh wait a minute, wait ten minutes, wait fifteen minutes, wait half an hour'. And under those conditions with one mmol ethanol we were always afraid we would burn it out, so we've gone to a one pass through. We simply pass the five mmol of ethanol through the target, then through a strong cation exchange resin (a Maxi-clean cartrigde), trapp all the ammonia back in the hot cell and then when we need it, when we *really* need it, not when they say they want, but when we really need it, we elute it with 150 millimolar base and then titrate it to neutrality to integrate or bunch or gather the ammonia, and that is independent of logistic snafus.

A: Yes, in our situation we must have a good contact with the camera of course, and we make about 90 or 70 mCi. So they are sure that after eight minutes they have 10 mCi. So then they can prepare the patient to give ... or whatever protocol they use.

Q: Why do you circulate the water ?

A: We had a system in the beginning of our PET center and this was our first infrastructure, so we used our target. And it's very simple if you can use the same infrastructure. The advantage of this target is that you have a good cooling, of course. The liquid is still circulating.

Q: Do you adjust for the isotonicity?

A: In that collecting file there is already saline of a high concentration, so we know that the same volume always before it goes over the filter, it's isotonic.

Q: I see. And otherwise, using the cation exchanger, it would ensure that we will not have any unwanted species, in the form of nitrate or nitrite. You don't get any anionic species ?

A: Yes the anion exchange column takes all nitrate, more than ninety nine percent. Normally in the literature they use 5 mmol ethanol. It couldn't be used because our target circulation pump had disfunctioned. Because when you bombard that liquid with 5 mmol ethanol, you make also CO_2. We had a plunger pump, and with the CO_2 we had a malfunction. It stops circulating. That's why we have a 1 millimolar.

Q: J. Clark: Can I just ask you, you have a very complicated plumbing system there. How do you ensure that it stays sterile?

A: It goes over a sterile system.

Clark: But if you got something growing in there, you've got pyrogens all over the place.

A: No, the tubing is from stainless steel, most of it.

C: That doesn't stop things growing.

A: No, ok but we always can .... the reservoir is upstairs in the hot cell, so we can circulate again. You can also circulate with ethanol.

Clark: I think if you had our medicine inspector breathing down your neck, you would have a big problem on your hands there. Things like growing in ethanol.

A: Till now, we don't have sterile products.

C: One solution to this sterility problem could be what we are doing. We use for flushing those lines, which could be critical, a solution of peracetic acid which can easily be cleared off with pure water afterwards and can easily be spotted with a simple spotting paper if it's all out. It's even better than hypochloride I think, because that could be problematic to find whether everything is gone afterwards. This is a method which is routinely used for cleaning equipment for dialysis. So this is very compliant with all medical systems which are used and it's a very effective method. And it's very easy to use.

C: J. Nickles: Thank you, this is a very useful tip.

Jeanne Link, Seattle: ^{3}He RFQ and Unique Targetry Problems

I am part of a collaboration with Jerry Bida from BRF. We are trying to develop targetry for an ^{3}He RFQ. I didn't expect to present anything here because we haven't tested our target yet, and we have presented some of this target information before. So I didn't bring any numbers on some of the issues that have already come up in this session. Jerry asked me to present because he just wanted people to know that there are some targets out there that are extending the limits of what we think is possible and that there are some big issues in these machines. What we are trying to do is accelerating $^{3}He^{2+}$ to 10.5 MeV using a linear accelerator, and it's an RFQ made up of four RFQ's in line. An RFQ has a 360 Hz pulse cycle with 2.5 % duty cycle. What that means is that we have 10-20 mA electrical, so 5-6 mA particle peak current, we have a 300 µA average current, which means we have 1,500 watts through the whole target, which isn't too bad, except that we have 50,000 watts peak current. So this makes it a challenging target.

The other issue is, ^{3}He has a much higher dE/dx than the protons or deuterons, so in fact we are working right now with a 4 to 6 micron Havar window that drops the energy 1 - 1.5 MeV, so we start with 10.5 MeV and we go down to 9.5 or 9 MeV. These windows, when they are not grid supported, can only have a Δp of 1 - 2 bar, this makes some of the issues on cooling much more difficult. We've opted to try and go towards more conventional targetry at the beginning. What we've done is we've spread the area out to 25 cm^2 , and the 25 cm^2 gives us about 2,000 watts per cm^2 which what I had heard was absolutely impossible till today when Karl said it. But we have three target types, we look at water targets, putting ^{3}He on ^{16}O, it' a circulating target, it runs at 3.7 or about 4 liters per minute. It's about 50 mL recirculating. That one we've actually put over 1 kW/cm^2 on the windows and it actually held that up. We've had full current density on the old machine on those windows, but not at the full current, in a smaller area.

We are also going to have a solid target or several solid targets, we are trying water backing and a gas cooling on the window. This is a challenge because you don't want to attenuate much of the helium in the gas. And we have a gas target that can be flow-trough or recirculating, but the issues becomes cooling in the gas target.

We are also looking at more experimental targets. Particularly, we're thinking about cryogenic windowless, we're not going that way to begin with, and we're lookin at grids. Jerry Bida has put a lot of work into gridded windows.

What we've done with this machine is, when we have the 10.5 MeV beam, we call it the high energy beam transport, we've designed it to be flexible. So there's a variable magnet and solenoids at the back where we can change the diameter of the beam to expand it or focus it from slightly over 1 cm^2 to a rectangular 25 cm^2 , which is 2 x 12.5 cm^2 . We've looked at the beam, we've a good beam on target now, but we are not up to 360 Hz yet. With this machine, we have only gone up to 120, and stable beam is still at 60 Hz. The beam we would like would be rectangular. It would be Gaussian in this dimension, and fairly flat across this dimension. We've looked at the beam, right now we have to do a little more tuning when the habit comes in, then it actually looks a little more like that. But that's our first pass on it. We are going to try and test our targets at a power density of 2,000 to 50,000 W/cm^2 . I am sure we will never get up to the 50,000 for more than about a pulse. The issues of this target haven't been answered, I can't tell you exact numbers right now. We are doing cooling trying to get into turbulent flow, and we are working with jetting the windows and the gases onto the front foils to try and get this turbulent flow. We have to have fairly good velocities, linear velocities to move it through.

The real interesting aspects of this are the challenges in the engineering of the target, but also what is going to happen if we put this much current in pulses on the target, what the radiochemistry is, the hot atom chemistry. We are going to see.

Q: T. Ruth: Can you tell us what the dedicated reactions are ?

A: The reactions are C-12 ... everybody knows this is helium, to C-11, so there's an 'n'.... no, alpha, I'm sorry. I told you I can't add standing up.... . So this is a low specific activity problem that we have been working on and a chemistry to try and increase the specific activity, but using the nucleogenic C-11 and getting it out of the matrix of the C-12. Another low specific activity reaction is again the 'alpha out' to make O-15, that one were not the only issues whether we could make high specific activity for making CO, because low specific activity oxygen and water aren't an issue. This is at this point a solid target, possibly thin foils, the O-16 is a water target. It's a water target because the O-16 reaction is also used to make F-18 (writing, counting particles)...so it's an 'n'. We do so many reactions, I can't remember them all. So this is one of our best yield reactions, this is when we actually had current on target, not full target, and we had good recovery of the fluoride. The issue is bringing the heat transfer down enough to bring the volume down.

The other reactions that we looking at are irradiating B-10 to make C-11. That's a solid target, probably boron oxide because boron is hard to work with. We haven't decided. That should be high specific activity. N-14 to give O-15, that will be a gas target and that will give us high specific activity. Another one we are investigating but the regulatory issues and the neutron production, because this is a neutron producing reaction, are Be-9.... should give us C-11. And that has a fairly good cross section, but it has got other problems. Neutron shielding on this machine is supposed to be real low, but at 10.5 MeV it's still a significant problem.

Q: Jeanne, can you tell us what the power requirements on the Linac are? It's a lot of power in the beam, what do you have to put in?

A: I can't, I could tell you tomorrow, I'm sure I have got them in the computer. I don't know, Ken, do you know what they are? 400 kW, ok, it takes a lot of power.

Q: And the dimensions?

A: The dimensions of the machine? Again, I would know this exactly. Each tank is about six feet, no, four feet, there are four tanks. The alpha magnets are about four feet long, it actually isn't the original configuration. What we have right now, the ^{3}He RFQ was originally designed to be a triple machine with three RFQs in close coupled RF and a He^{2+} source where the ion source would start here, and there would be three RFQ tanks accelerating up to 8 MeV. What happened was we couldn't get a high current He^{2+} source, so the next evolution of the design of the RFQ went to a low energy beam section which had the ion source here, beam goes this way, one RFQ and then what they call the medium energy beam transport system, which involved taking He^{+}, stripping it with a gas stripper, foil stripper isn't possible because of the high dE/dx again, and then bunching it into the next two RFQ's. The problem was, there were several problems, the ion source wasn't working, there were problems with the RF systems and the third problem was, they weren't getting bunching, they weren't getting the beam into the second RFQ. And then some of the reason for making the RFQ changed and we suggested to make it a larger energy. So we've gone now where the first front end of the machine is the same. There is a single RFQ tank that goes to 1 MeV, at which point the low energy system is changed and two alpha magnets were actually in there, about five quadropoles to take the beam, to strip the beam and refocus it and bunch it into the RFQ's. And now there are three RFQ's at this end, taking it to 10.5 MeV and the beam goes like this. It's actually been interesting here because the alpha magnets for taking ^{3}He and doing this kind of movement of the ^{3}He beam has never been done before, and they actually found out some stuff about space charge neutralisation and getting this to work.

So this is what the machine looks now. The actual size is about 20 feet by 6-8 feet wide. It's a very big machine, but the size of the machine isn't the RFQ. This machine has changed a lot, it was initially envisioned as a very small table top, one button push button machine. We really thought it was going to be, it's not. This machine has been rebuilt by Fermi Lab and they deserve a lot of credit for the work they put into it, but the machine we have now is a medium energy physics machine that is built like a brick house. The thing that they build now is the RFQ I've described, but they have put so many diagnostics in it, that this machine right now goes up about four feet above the RFQ filled with 19 inch rack diagnostics just to understand the mechanics of this beam and what's going on. There are RF towers, the RF system at this point was bought on salvage, so they wouldn't redo or develop new RF because of the budget. And the RF towers, there is a single RF tower for each of them, just the regular Linac RF station one of the ... about 20 years ago.

Q: Jeanne, where is the stripper in that diagram?

A: The stripper? I'm not sure, in the middle of this. It's in here, the stripper is in this part. I think it's right at the beginning. So the stripper is here and then there's these magnets to take it and I guess it's a way of keeping it somewhat isochronous and punch it back in, that's why they went with this design. You have to talk to the physicists, I don't fully understand the 270 degree bending design. But it was a way of keeping space down and yet getting the beam over a long distance back into these RFQs. It's quite a physics machine.

C: K. Krohn: The stripper is just a Nissan fuel injector. This is actually one of the successes of the machine.

A: Yes. But a Mercedes Benz one doesn't work quite as well. It's true, they actually tested all that.

C: M. Welch: But can you say that in Germany?

A: You're right, I'm sorry. So, I apologize for my diagrams, as I said I didn't think since we haven't tested full beam. They are conditioning it now, we are up to 8 mA average current

extracted out of the ion source coming through. They can get about that much through. The extraction is not as high as they'd like, but the big problem right now is conditioning the RF. The RF still has to come up and they are not up to the rep' rate they need or the duty cycle they need.

Q: J.-L. Morelle: Is the motivation in this project for other applications and for PET?

A: Initially no. I think we are looking at other applications now. Now that we understand what this machine was like...

C: The need for the original application is going away.

Q: J.L. Morelle: Coming back to targetry, I have one question. What was the temperature increase during a single pulse?

A: You see, that is why I wish I had my numbers. It depends on the cooling. I have whole tables, there was quite a bit of heat calculation work done. And the temperature increase in a single pulse, depending on how much cooling goes from completely melting the Havar window above 750, or where we're running, it's down less than a hundred degrees.

Q: J.L. Morelle: That means that was in that time you really have a vibration that you can hear in your targets ?

A: We don't. We haven't seen that so far. You'd think you would and one of my biggest worries was the pulsing of the beam, but we have not seen that. We have not put in acoustical, we haven't put a microphone on it either. That's one of the things we don't understand is the flexing of what will happen with this pulse beam until we try it.

Andy Roberts, Madison: High Current Experience from Hammersmith

The MC40 at Hammersmith (here it's just the usual layout thing) has large body targets. They have at least a 2 cm window, generally pretty big things. In some cases that is desirable. But there's certainly a lot of reasons why you want to get away from that, and get to smaller targets, either for reasons of specific activity or you want to use enriched isotope, or you want to run at high pressure, high pressure water targets. The way they irradiate their targets, they have a spinner in the beam to give you a lower power density on the actual target. So we turned the spinner off and collimated it down to 9 mm and did some targetry development there. And what we found was, you can get very hot beams out of this positive ion machine, we are going through a 9.5 mm aperture with more than 95 % transmission, so it's a small beam and the foils don't like that. I sent a whole bunch of Havar all the way up here, so you have to find a way of dealing with those hot beam spots. The most convenient thing – since we have energy to burn – is to use an aluminum plate window of 0.7 mm thickness. It worked very well. We did some rough calculation, it should melt at 50 µA of 19 MeV. At that thickness you are burning about 3.5 MeV, so that's a couple of hundred watts into the window. It runs routinely at about 400 psi and shoot 40 to 45 µA on it. After I left they actually blew a window, there was a kind of mistake with the current, they guessed it was at about 70 µA.

Discussion:

Q: K. Erdman: When you had a total aluminum target and the Havar window in front, what does that do to this problem of getting weird stuff and not getting carbon out and so on ?

A: Nothing actually. For the F-18, it works like gangbusters, there is no problem with that at all.

Q: You haven't tried the CO_2.

A: I actually did a CO_2 run using the F-18 target after it had been used for the fluoride and everything else. And that worked really well. Once.

Q: J. Nickles: But you mentioned, that didn't go backwards?

A: No, you can't go backwards that way, but that's a separate issue. I was just working under the assumption that irradiated fluorine would clean out your target better than just about anything. And it did. There was very little cold carbon that came out of it.

Dave Schlyer: Thermal Modeling

I would like to give a short overview on the theoretical models. Some of the problems you'll hear about are the same ones that Jeanne talked about, and we have some of the similar. We are looking at the target design and doing window construction and heat transfer, some theoretical calculations. As I mentioned, we just got the accelerator running just before I left last week. So we don't have any experimental results yet. But this is the problem we both have to deal with, well, if you have a 20 μA cyclotron beam, you cannot see it, it's in the dust down here below, whereas here we are talking about a 10 mA pulse that comes through, but with some time in between the pulses. So what we are trying to do is build targets that will stand that kind of a pulse.

This is just a general idea what we want to do with these, we want them to be reliable, simple and we want to be able to work on it without getting a incredible exposure.

This is some modeling of the window stress. This has been done before. These formula come from engineering diagrams. The purpose that we had in doing this is repeating some of these results to find out if you use a supporting grid, that is if you take a foil and a thick barrier on the back of it, drill holes in it, an then stretch it, will that formula accurately represent the stress on the foil, and will it break at the point predicted.

We carried out these experiments. Indeed, you do get predicted yield stresses here, and we do get exactly the foil failure at the point one would expect to get it within experimental error. There was a difference between foils, especially thin aluminum foils. You can see the thickness here is very small....trying to get aluminum foils that would have no crincles.

The next thing we wanted to look at was heat transfer in gaseous targets. This is based on some work that was carried out by Dick Lambrecht. The prediction of the temperature rise and the pressure rise in the target when you put beam on. We used that general equation that they developed and expanded that a little bit so that we can actually determine what the film coefficient for heat transfer out of a gaseous target is, using some experimentally determined parameters. You then can extrapolate beyond your measured conditions and predict what the temperature is and the conditions in the target will be way beyond where you can actually do the testing.

These are some target temperatures that we measured some time ago when Sven-Johan was at Brookhaven. These are temperatures for two different energies of beam, and we came to the conclusion that we had circular distribution.

The temperature rise inside the target goes up relatively slowly, over a few seconds time period, if you have a gas target. We were looking at this in particular with respect to gas targets with these pulses.

We were talking about Jeanne's prediction on the temperature rise in the foil during that single pulse, this is for protons at lower energy, but you get typical values if you calculate temperature rise in the foil. You get 48,000 degrees per second, but because of the 125 µsec pulse it is only a 6 degree temperature rise.

This is just a picture of the target we use based on these theoretical calculations and predictions. That's the one that is sitting on the end of an RFQ now, waiting for irradiation.

Discussion:

Q: T. Ruth: What kind of support is necessary for gridding the foil? Do they have to be in intimate contact?

A: Yes. What we use is the pressure inside the target to push the foil. We don't bond the foil to the grid. I know that other people are talking about that, actually bonding the foil to the grid.

Q: Does that add more thermal conduction ?

A: We are using Havar windows with ¼ mil or less, that's 5 micron Havar windows or less. At that, there's absolutely no way you can conduct the heat out of a small hole in the grid. You have to conduct it to a better conducting grid, aluminum or something, we are using aluminum. And the better you can make that contact, the better off you are getting the heat out of the foil. We are not putting any gas on the front of the foil, no cooling gas, we cannot afford the energy loss. We are strictly counting on conduction to remove the heat from the front foil. That's why we have to use a support grid that has the good thermal characteristics.

C: M. Welch: That window assembly would be very similar to the one we have used on the TCA for about four years. We have about 50 % transmission and to make oxygen we run 170 µA, 3.2 MeV.

A: I have confidence that it will work on this accelerator also.

R.J. Nickles: Special Targets at University of Wisconsin

We've got about a number of targets at Wisconsin, that we do special purpose work with. The first one I'll just briefly touch on, it's to obvious that there are many occasions where you need neutrons, fast neutrons, for fast neutron activation analysis. So we shoot together or put together a fairly carefully thought out beryllium, thick beryllium target, with the intention being to be able to get the secondary target that is going to intercept the fast neutrons that are basically going forward it, with 11 MeV protons onto thick beryllium, we are getting as close as possible to the proton beam strike on the beryllium. We managed to get it down to about two or three mm so as to allow us to get about 2 x 10^{11} fast neutrons per second per cm^2 going through about a basically 1 cm^2 slot. So it works for teaching, it works for fast neutron activation and for small samples.

The second thing, which is a B-10 target with the intent of making C-10 from $^{10}B(p,n)^{10}C$. And this is in a centered fused boiling mass of elemental boron and boron oxide. And so for

learning about the centering process we are mocking up the beam power with an oxygen-hydrogen torch and doing pyrometry on this boiling mass. A second whole class of weird stuff that we make requires the need of a vertical switching magnet. About half a dozen cyclotrons around the world have such a magnet and our intent with this was to be able to handle the group six elements in order to be able to make the halogens through (p,n). The group six elements are terrible as targets, they've got high vapor pressure at that temperature, they've got a low melting point, they've got an absolutely abysmal heat conductivity, they're toxic, they're corrosive. S, Se and Te in particular, these are the ones I am talking about. And so the major intent for this was to be able to irradiate Te-120, which I found a price at $40,000 for a target load would have allowed us to do this actually. I haven't been able to even find that source any more. So it's a double focussing magnet that brings the odious beam down to beam spot. We are irradiating a simple beaker of water, and in fact you can see it's about a mm across by about 2 or 3 mm in the other dimension.

What we are making right now are stents, and this is a stent, that little thing right there, it's a little tube, a slotted tube that is stainless steel 316. And it's used in PCTA balloon angioplasty. About half a million of these are done in the US alone, probably many times that around the world. It's overtaken the cardiac bypass and these stents.... OK, balloon angioplasty has got a dark side in a sense that about half of the people who have it, come back in six months with restenosis. The use of stents, this chainlink fence that is left behind in about half of these balloon angioplasty operations somewhat cuts this number down, but not very much. If you now however irradiate this stent, to leave a few µCi of activity on it, enough to give about 10 to 20 Gy over the next six weeks, that is remarkably effective just spectacularly effective in cutting down the proliferation that is behind restenosis. So what we are doing is both electroplating and directly activating by direct irradiation of the 316 stainless to make these things slightly radioactive. This is a new initiative that we are trying to do to broaden our base.

Part of the chart of the nuclides that we've been looking at over the past few years would be pushing the RDS to make different things, the weird stuff that I'm speaking of. And between Li-7 up to W we've had some fairly good yields in about 130 different reactions. And this between Ca and Zr show you the transition metals that are the most interesting to us as we are making stents and making most of our things. Where the color scale shows you that you are going from mCi up to Ci end of saturated bombardment on a pure material. What's clear, is that although we talk about these small cyclotrons as though they are PET machines, you can either choose to ignore the versatility that's there or you can make use of it in the midnight hours to broaden your base.

Q: M. Welch: How do you hold the stents in the beam?

A: I didn't show that, it's a rotating mandrel that's water cooled, with a water screw that brings the water up. It's virtually out in the room, so it's rotating at 1800 rpm and oscillating up and down and spinning around.

Q: J. Clark: Jerry, I'm sorry I can't resist. What is the scientific basis of these radioactive stents actually ablating a throma ?

A: The same major response, proliferation, think radiation. It's killing off some of these proliferating tissue cells that are either generating or sensing.

Clark: Is this your hypothesis or is that...

A: No, I don't know anything about this..

C: M. Welch: There's an awful lot of literature on this, and there are whole companies that have been formed to do this one way or the other.

A: Yes, very aggressive companies.

C: M. Welch: Filling balloons with matters, putting activity in the outer rim of the balloon, there are various approaches. I think the problem. There have been at least three symposiums in the States on this, on irradiating after angioplastic in the last two years. Having had angioplastic twice I sort of fall for that.

Q: J. Nickles: Have you tried irradiating your own stents?

A: M. Welch: I haven't. A thing on the side: The interesting thing is, the figure is 30 % of people who have angioplastic restenosis. But the thing that people don't know about this approach is that whether it stops that restenosis, or simply delays it. Because if you have a balloon angioplasty now they don't let you travel for six months in case of restenosis. And if it doesn't in six months, in 90 % it's certainly is not going to. Of the 30 % still going to restenose where is it going to be extendable to three to four years. That's sort of the interesting question.

C: J. Nickles: Actually the foremost group is the University of Heidelberg, the group of Hehrlein et al., who've been doing this for about five or six years now. We're in the hot bed of it right here.

C: K. Erdman: I just want to make one comment. In the vertical plane cyclotrons like our TR-19 one plane goes uphill and one beam extracted goes downhill. So this is an ideal place without using a bending magnet to use liquid targets. And in fact some of those are being developed at TRIUMF now for use.

Nickles: A pity, the PET trace wasn't built that way.

Q: How did you measure the neutron flux ?

A: By activation of sodium, aluminum and five different things. It was in the IJARI a couple of months ago.