1.2 Target Bodies

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Only rarely target material can be used directly as a target. Important is the design and fabrication of devices for holding and/or moving targets. Target support, target holder, target body and target box are used to describe these devices. During conversations and in literature, one is interchanging target and target holder frequently. We suggest to use the correct word in the right place. An example is the announcement of this morning session on "targets" and their design.

From what are target bodies made?
The materials used will have great influence on corrosion, activation, contaminants, cooling inserts, plating, etc. The choice of material depends on the following main factors as shown in the next table.

- nuclear reaction
- state of aggregation of target material
- enriched target material
- beam parameters
- work up of product
- workshop of the cyclotron facilities
- costs and ease of handling of the materials used

The design of cyclotron target bodies is primarily determined by the target material to be irradiated. Next to the choice of the nuclear reaction is the state of aggregation of main importance. There are big differences between solid, liquid and gas target bodies. Even more intriguing is the target body construction for the use of very expensive enriched isotopes, for instance ^{124}Xe. It is clear that the chemical/physical form of the radioactive product plays a major role as well, for example, the formation of ^{81}Rb from the target gas ^{82}Kr. The gas target holder has to be washed to get the solid Rb out. Of the beam parameters, the beam current has a high impact on cooling of the target body.

There are no standard target holder equipment because they are mostly manufactured in the workshop of the cyclotron facility. If we take for example the production of ^{81}Rb from ^{82}Kr by the (p,2n) or (d,3n) nuclear reaction, we see that this straight forward production method leads to as many different target bodies as institutions of production. This is demonstrated in the following table:

1.	Stainless steel	+ Al foil (conical shape)	Argonne NL
2.	Ni tube	+ Havar foil	Medi Physics
3.	Ni holder	+ Cu foil	BNL
4.	Stainless Steel liner (rotating)		Heidelberg
5.	Stainless steel	+ Ti foil	Gent
6.	Stainless steel	+ Al foil	Milano
7.	Al electrostatic	+ Al foil	Milano
8.	Cu cryo target		Milano
9.	Stainless steel	+ steel foil (conical)	Turku
10.	Al tube–anodized		Hammersmith
11.	Brass–Ni plated	+ Ta foil	Eindhoven

Although this list is not complete, it gives an impression of the variety of systems, and this is just for one product. The materials used for the target holder may differ totally, although the same nuclear reaction is used. Nevertheless, for a particular production method the target holders have some features in common as dictated by the nuclide production requirements. In the ease of research, target holder requirements are less strict as far as cooling and activation are concerned. For routine productions remote handling and control is very important and gives extra points of concern in the construction of the bodies. With this idea in mind, let us look into some aspects of the target bodies as shown below:

- corrosion
- activation
- contaminants
- fabrication
- cooling
- inserts
- plating

At first glance one may be surprised by the fact that so few details about these aspects of target bodies are in the literature. Review articles in this field are scarce. Some arguments can be found for the lack of extensive literature:

- economic reasons
- research reasons
- no need for optimisation
- problems may be institution dependent

Economic reasons may prevent publication of details from production facilities. In research institutions there may be no need to go into detail when the production of the desired nuclide is high enough. Problems may be institution dependent so its not much of use to others. Apart from that most people solve their problems on their own and publish only the final results if anything at all. Especially the long term behavior under irradiation conditions is not very popular in the literature.

As far as we know there are no reviews published on any of the subjects of this session, nor a combination of them. Useful information can be obtained from the book of J. Clark and P. Buckingham and from the section of F. Helus in the CRC issue on radionuclide production; although these were not meant to go in detail into the subjects of interest.

Corrosion can be divided into a few stages:

- corrosion during irradiation
- corrosion under work up conditions
- passive corrosion

The passive corrosion is of minor importance compared to the other two.

Favourable conditions for corrosion are:

- heat
- radicals and peroxides
- aqueous solutions

What are the main causes of corrosion?

- oxygen in starting materials (ozone)
- aggressive behaviour of target materials
- sweeping gases as F_2 in Ne
- washing of the target body
- peroxide formation in water (solution)

Oxygen in gaseous starting materials leads to ozone formation, which has strong oxidizing characterisitics. An example of aggresive target material is iodine or iodine compounds.

It is clear that water can cause corrosion. Although very reactive nuclear products do not cause corrosion on a large scale, the problems for the work up caused by reaction with the target body may be enormous. ^{18}F as F_2 or as HF is a good example. Another is $^{15}O_2$ on Cu. How to prevent corrosion? Choose the best materials!

Activation of the target bodies

This phenomenon depends on:

- material used
- accelerated particle
- energy of the particles
- beam current

For routine large scale production runs, activation of the target body may become a big problem, especially when something goes wrong during the irradiation and one has to repair as soon as possible to keep up with the delivery schedules.

Al seems to be an ideal metal for target holders for reasons well known to you. It is easy to work with and the activation is low because only short lived Si–nuclides are produced. Moreover, the material is cheap and the cooling is relatively easy.

Al:
- cheap
- easy to handle
- low activation
- good heat conductivity

Al(p,xn)..Si $t_{1/2} < 5$ sec

Cu (adsorbing oxygen) (brass = Cu + Zn) is easily activated:

Cu(p,xn)^{63}Zn and ^{65}Zn (243 d)
Cu(α,2n)^{67}Ga or Cu(^{3}He,n)^{67}Ga
Zn(p,xn)^{66}Ga and ^{67}Ga (78.3 h)

Ni: (monel = 67 % Ni and 30% Cu + Fe and Mn)
^{64}Ni(p,n)^{64}Cu (13 h)

Fe (stainless steel):

^{56}Fe(p,xn)^{56}Co (77 d) and ^{57}Co (270 d)
etc.

Contaminations

The final product can be contaminated by other radioactive nuclei or by chemical contamination.
In general the produced radioactivity in the target body is not easily transferred to the desired product and thus will not lead to major nuclide impurities.
Chemical contamination can be expected when an Al target body has to be washed with a basic or acidic aqueous solution. The target body may slowly dissolve and one ends up with an unpleasant chemical impurity in the final product.

Cooling

In general the cooling of foils is more complicated than cooling of target bodies.

1. Internal irradiations
   For internal irradiations the cooling of the target holder is a major problem due to high beam currents and no possibilities for beam spreading.
   For instance, over 4 kW is dissipated in 0.3 cm^2 when 22 MeV protons and 200 μA are used (thick target).
   Internal targets have a limited application due to technologically complex construction which leads to high costs.

2. External target bodies are heated to a lesser extent by the defocussed beam so cooling is in most cases not a big problem.
Especially gas target holders have a greater surface and are easily accessible for cooling.
For solid target bodies the thermal stability and heat conductivity of the materials used are important.

Fabrication

The problems encountered during fabrication of the target bodies depend strongly on the materials chosen.

Discussion

This session was started by J.Clark with a short introduction guided by a hand-out. The aim was to go straight into the problems without a concise overview. He introduced the topic by mentioning a target body or system in each category as seen in the hand-out.

T. Ruth opened the discussion by raising the following question: Who has experience with the static F_2-Neon target using different target body and foil materials? What are the best materials and what is the recovery under the possible conditions? According to J. Clark and P.Horlock, the Inconel foils are no good for this system. They observed a green deposit on the foil and a lower recovery of the $^{18}F_2$.

D. Schlyer used a solid Ni-target body and a Ni-foil at BNL. The target body is water cooled and the Ni-body is removed and polished at certain intervals.

R. Wagner uses a chemically Ni-plated passivated Cu-Be target body and a Havar foil for more than 2-1/2 years now. A minor decrease in performance has been observed. The back plate is water cooled. During irradiation the body contains 60 μmoles of F_2.

T. Ruth asked what the experience with target washings is. The EDTA washing helps to keep the ^{18}F recovery on an acceptable level. There seems to be an improvement after EDTA washing but there is a lack of basic knowledge. Just replacing the old foil seems to improve the recovery in some instance as well. The foil may even be the main cause of the yield drop.

G. Meyer noted deposits on the passivated stainless-steel foils and concluded that foil corrosion resulted in recovery problems which gave a significantly lower yield.

M. Guillaume found no difference between a Ni and a stainless-steel 316 target body as long as he did not open it to air and passivated the body. He used 0.15 % F_2 in Ne and found a recovery of 90 % of the $^{18}F-F_2$ of the theoretical value as calculated from the BNL and Nozaki figures. A recovery of 60-70 % would be more likely and participants were disputing the 90 % recovery. M. Guillaume also tested a golden target body. This resulted in no recovery at all. He did not give more details on this subject.

According to T. Ruth, the passivating of the target bodies is not as difficult as one was led to believe by the earlier papers in this field (T. Ruth will add some additional information). Somebody suggested to collect the white or grey powder from the target body and analyse it.

R.J. Nickles mentioned an electrostatic target body to shrink the surface in contact with the $^{18}F_2$. One would expect less surface interaction. Beam on Neon glow, voltage 1500 volts and a current in the system of some mA.

O. Solin tested an electrostatic target body as well and used in his experiments a beam current of ca. 1 μA. During the discussion no figures on yields and recovery were given.

B. Wieland mentioned the effect of beam strike on product composition and recovery. He noticed that the recovery was better when beam strike was avoided. Also better results were obtained as far as the radiochemical purity of the $^{18}F-F_2$ is concerned. The percentages of CF_4 and NF_3 tend to rise when the beam hits the wall. The N and C come from air. B. Wieland mentioned a 4 cm^3 Ni target body with $^{18}O_2$ as target at 20 atm. He did not give experimental details.

G. Bida had some experience with such a target body and obtained 25 % recovery. He did not prove the identity of the obtained product. The activity behaves chemically as $^{18}F-F_2$ but could be $^{18}F-OF_2$ for instance. He did not mention the addition of a carrier amount of F_2 to the used $^{18}O_2$.

R.J. Nickles did not have success with such a target body. At least in some of the experiments he used a mixture of $^{18}O_2$, Ne and 0.1 % of carrier F_2. Further (technical) details were not given.

Then O. Solin went back to the problem of determining the exact yield and recovery. In some experiments he recovered from the Ni target body $^{20}Ne(d,\alpha)^{18}F$ 90 % of the carrier F_2 while only 70 % of the ^{18}F was obtained.

P. Malmborg mentioned that most people use the I_2-titration method and that in such cases only the oxidizing potential of the total contents of the target body is measured.

Then, B. Wieland started the discussion on the $^{14}N(p,\alpha)^{11}C$ production and the behavior of the used target bodies over longer periods of time. Does the production of $^{11}CO_2$ go down after a more or less considerable period of time?

G. Meyer noticed only a few percent drop in yield after some time when using an Al-target body. The following statement was put forward: When pure nitrogen gas (99.9999 %) is used the oxygen in the system is burned out. Not everybody agreed on this. People who have no problems may have an oxygen leak.

C. Crouzel had no problems with recovery of $^{11}CO_2$ in static 30 minutes irradiation runs. He compared the halogenation of methane by chlorine, which leads under radical favouring conditions to tetracarbonchloride (90 % in 1 min) with the ^{11}C-production from ^{14}N by irradiation of a mixture of 95 % pure (99.9999 %) N_2 gas and 5 % H_2. One would expect $^{11}CH_4$ but you get only $^{11}CO_2$ instead (How about ^{11}CO?). There seems to be still enough oxygen in the system to get $^{11}CO_2$ (Remark from the editor: The results of irradiation of N_2/H_2-mixtures can be found anywhere in literature) Oxygen from Al-oxide covered Al-target body influences a high specific activity of the produced ^{11}C. With a new system or a system with a new foil you need many runs to get your good specific activity again.

C. Crouzel noticed some problems with the chemistry afterwards when the system was in use for a very long time.

P. Malmborg once had an old crummy Al–target body for the $^{11}CO_2$ production which never failed. He decided to build a neat system with stainless steel lines, etc. and stumbled into recovery problems. The extraction of the $^{11}CO_2$ from the target body dropped significantly after some time. He decided then to add 0.1 % O_2 to the pure N_2 and since then has had no problems.

S.–J. Heselius had the same problems with their fancy target body made of Al and he was successful with the same treatment.

J.R. Dahl suggested the use of glass liners and referred to session 1.4 of this workshop. He also had a substantial drop in the recovery of $^{11}CO_2$ from his target body. He checked the activity in the target body and found a half–life of 20 minutes, so it had to be ^{11}C. Its hanging in the target body and nobody seems to know in what form. Sticking to the walls did not have a high probability according to the audience.

A. Paans uses a target body of Al, 0.5 mm Al foil and stainless steel liners. Pure nitrogen (99.9999 %) under 12 atm. is used for the production of $^{11}CO_2$ with a specific activity of approximately 1 Ci/μmole. There are no recovery problems over the years. The high pressure makes an air–oxygen leak unlikely.

D. Schlyer suggested that in some cases the beam hits the wall of the target body resulting in a destruction of the rather inert Al–oxide layer. If this happens a very reactive surface is available for the ^{11}C produced during bombardment of the N_2 gas. People should look carefully into the beam parameters of their system.

J. Clark wondered if people find that $^{18}F^-$ does not behave like $^{19}F^-$ chemically when the ^{18}F is produced in a water target. The 2–FDG synthesis is screwed up very easily by Al, Cu or other metal ions.

R. Weinreich found that in his system, the frequently used titanium target body did result in ^{18}F that did not behave properly in the subsequent chemistry. An explanation could be that the thick Ti–foil (fixed energy of 72 MeV protons) leads to Ti–F complexes which may be too stable to allow sufficient free $^{18}F^-$ for the chemistry. He switched to a solid silver target body with a 0.7 mm Ag foil. AgF is a known fluorinating agent. In doing so, a very good chemical behaviour of the ^{18}F was observed. J. Clark noticed the oxidation of the titanium back–plate of the target body resulting in a white spot. Somebody stated that Ag catalyses peroxide decomposition while Ti does not! The peroxides may be formed during the irradiation of $H_2{}^{18}O$.

J.R. Dahl uses a water target with a Ni–plated brass target body for the ^{13}N–production. The brass is covered by four 25 μm layers of Ni and in a four year period there were no failures of the system. It seems that a very thin layer of Cu has to be brought onto the brass before the chemical Ni–plating is started (hypophosphate reduction). He noted a cloudy product under certain conditions, for instance, a long stay (over night) of the H_2O in the target body. He expects the cloudy material to be Ni–oxide. He concludes that Ni is not the optimal material for water targets. Then, a discussion on dry–wet target bodies started.

F. Helus used glass liners for many years and is content with his system.

I. Trevena uses an Al–target body for the p,2n reaction on ^{124}Xe. They need to dry the target body after washing, otherwise they get serious retention of the ^{123}I in the next run. He considers the target walls as an ion–exchange column. If you treat the walls as such, the product comes off quickly.

J. Clark complained about the drop in the yield of Rb when an Al pressure vessel was used as target body for the Kr–gas. The Karlsruhe group uses a chromium plated stainless steel target body for the Rb production. In their situation it makes no difference.

Drying and not drying between runs gave the same results.

O. Solin uses a stainless steel 316 target body with a tantalum back plate (low neutron production?)

D. Schlyer used in Riad an Al target which he dried by pumping only and the recovery of Rb was adequate.

M. Guillaume mentioned a stainless steel target body for the Kr gas. He did not dry between the runs, on the contrary, he keeps the target walls wet and obtains up to 95% of Rb from this system.

On request of J. Clark, B. Wieland gave some information on electroformed sleeves of nickel. This technique uses Al mandrels which are electroplated with 125 um Ni layer. Then the Al is dissolved leaving a conical or cylindrical shaped sleeve depending on the starting form. These sleeves are guaranteed up to 140 kpsi (9.7 x 10^3 atm). B. Wieland can add the address of the company in New Jersey and has details on the prices. (Servometer Corp., 501 Little Falls Rd., Cedar Grove, NJ 07009 (201)-785-4630)

J. Clark: Ni welding is not a big problem but with Al it is much more complicated. One has to be very careful and it is expensive and don't forget to specify the leak testing.

B. Wieland mentioned a leak tester up to 10^{-6} cc/sec for 500-600 US dollars. It is also very useful for leak testing on lines and water traces in the cyclotron.

J. Nickles advocated the use of a torch for Al brazing. He obtained very good results and the price is low. At this point we run out of time and the discussion was stopped.

From the foregoing it is clear that about many things (too) little is known. In a discussion afterwards, three main subjects for further evaluation were chosen:

- Optimal materials for ^{18}F water target bodies and effects on chemical behavior of the produced $^{18}F^-$.
- Dry-wet targets; procedures and materials
- $^{11}CO_2$ target body performances. Basic understanding of $^{11}CO_2$ recovery.

Special Examples of Target Bodies

$^{11}CO_2$ Gas Target:

Target Gas	–	N_2 – corrosion: no problem
Material	–	Aluminum – commonly used
	–	Surface pretreatment not well documented
Contaminants	–	easily operated no carrier added
Cooling	–	usually used

$^{15}O_2$ Gas Target:

Target Gas	–	$N_2 + O_2$ – corrosion by NO_x not insignificant
Material	–	Aluminum – commonly used Hard anodising may help control corrosion
Contaminants	–	cannot be operated no carrier added due to wall losses as $^{15}NO_x$
Cooling	–	usually used

$^{18}F_2$ Gas Target:

Target Gas	–	Ne/F_2 – corrosion not a significant problem if only dilute mixtures used, e.g. 0.2 % F_2
Material	–	Nickel – machined from solid or welded
Activation	–	Significant problem, use of disposable liner may be advantageous
Contaminants	–	Can only be operated with carrier F_2 Radiochemical losses significant problem
Cooling	–	Optional, all metal targets are often run hot (approx. 250 ^{o}C) using beam power

^{43}K, ^{81}Rb, ^{123}Cs Gas Targets:

Target	–	Argon, Krypton, Xenon operated in wet/dry modes– corrosion potential
Material	–	Variety of materials tried No clearly defined preference
Activation	–	Walls hit by beam due to multiple scattering especially at high Z. Conflict with desire for small inventory of enriched gases
Contaminants	–	As targets operated in wet/dry modes corrosion and radiolytic interferences possible

^{18}F Liquid Target:

Target	–	H_2O
Material	–	Variety of metals tried, e.g. Ti, Ni, SS, Au, Ag and electroplated variants are some good and some bad!
Activation	–	Problem with above metals but scope for reduction by using entry and exit window configuration
Contaminants	–	Trace dissolved metal impurities and possibly radiolytic products interfere with subsequent chemistry
Cooling	–	Essential to avoid boiling

^{13}N Liquid Target:

Target	–	H_2O to produce $^{13}NO_3^-$ and $^{13}NH_3$ – corrosion not a problem
Material	–	Aluminum
Activation	–	Long term build up not a problem
Contaminants	–	Can be operated no carrier added
Cooling	–	Essential to avoid boiling

^{123}I – H_2O/NaI solution:

Target	–	Solution of NaI in H_2O – corrosion anticipated
Material	–	Stainless steel – grade unknown
Activation	–	Significant as with all stainless steel systems
Contaminants	–	Building up of corrosion debris in liquid

^{123}I Organic Liquid:

Target	–	CH_2I_2/I_2 solution
Material	–	Ti corrosion not a problem
Activation	–	Significant
Contaminants	–	Mainly due to radiolysis of target material to e.g. ethylene

^{123}I – NaI melt:

Target	–	Molten NaI heated by beam and oven
Material	–	Stainless steel
Activation	–	Significant
Contaminants	–	No apparent problem – target material sublimation blocks pipes