Workshop on the Management of Non-Nuclear Radioactive Waste

Managing Hospital and Medical Accelerator Waste

Dr. Carlo Bergamaschi
Radiation Protection Expert

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What is a cyclotron?
A cyclotron is a particles accelerator. It is an electrically powered machine which produces a beam of charged particles that can be used for medical, industrial and research processes. As the name suggests, a cyclotron accelerates charged particles in a spiral path, which allows for a much longer acceleration path than a straight line accelerator.

How does a cyclotron work?
A cyclotron body consists of electrodes, called 'dees' because of their shape, in a vacuum chamber. This vacuum chamber is flat and sits in a narrow gap between poles of a large magnet which creates a perpendicular magnetic field. A stream of charged particles is fed into the centre of the chamber and a high frequency alternating voltage is applied across the electrodes. This voltage alternately attracts and repels the charged particles causing them to accelerate.

The magnetic field moves the particles in a circular path and, as they gain more energy from the accelerating voltage, they spiral outwards until they reach the outer edge of the chamber.

Modern cyclotrons accelerate negative ions created in a plasma. When these negative ions reach the outer edge of the chamber the excess electrons are stripped off the ions forming positive particles such as a proton or deuteron, which can then be extracted from the cyclotron as a beam. The size of the vacuum chamber determines the length of the spiral path and hence the amount of energy attained by the particle.
Medical cyclotrons

Medical cyclotrons produce proton beams which are used to manufacture radioisotopes used in medical diagnosis. Radioisotopes produced in a cyclotron decay by either positron emission or electron capture. Positron emission tomography (PET) and single photon emission computed tomography (SPECT), which utilizes the gamma rays associated with electron capture, are two imaging techniques that rely on cyclotron-produced radioisotopes.
Example of not self shielded cyclotron

**Siemens Eclipse HP**

- 11 MeV protons
- 60 μA (x2) max. current
- MAX. POWER: 660 W (x2)

**IBA Cyclone® 18/9 -HC**

- 18 MeV protons
- 9 MeV deuterons
- 150 μA max. current
- MAX. POWER: 2700 W
Not self shielded cyclotron: typical installation
Example of self shielded cyclotron

GE PETtrace Self-Shielded

GE PETtrace

- 16 MeV protons
- 8 MeV deuterons (option)
- 60 µA max current

MAX. POWER: 960 W

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Self shielded cyclotron: typical installation

Self-shielded (~30 cm walls)

- Shielding (neutrons)
- NCRP 144
- Reinforced floor
- Beam direction
- Negative pressure
- Conduits – Power and utilities
- Trenches – Product delivery
- Corner Pit – Decay in storage
- Power supply room
- Support room
- Control room – Vault door
- Work bench – targets service
- Product distribution valves
The dark side of the cyclotron

Open Cyclotron
All parts requiring maintenance can be isolated from the cyclotron vacuum
Target chamber

Target chamber is made up of silver. The front flange supports the beam into the target chamber for irradiation.

Two thin metal foil windows (Havar foils) are used to separate the target chamber from the cyclotron vacuum. Havar foil forms the entrance of proton beam into a highly pressurized $^{18}\text{O}$ enriched water target used for the production of $^{18}\text{F}$. 
Example of synthesis circuit of $^{18}$F

Nuclear Reaction:

$^{18}$O (p,n)$^{18}$F

$\tau = 109.8 \text{ min}$

Target Holder Material
- Titanium
- Silver
- Niobium
- Tantalum

He gas (500 psi)

Water Cooling

H$_2^{18}$O (> 95%)

$\iff$ recover

Synthesis Unit

$^{18}$F Reaction Vessel

Precursor

$^{18}$F trap

Purification

Final Product

Batch

Yields
Target: 5-10 Ci
Trapping: > 95%
Labeling: 30-60%
Purification: ~ 95%

Specific Activity: NCA
Commercial Distribution: YES!
Cyclotron waste

CONSUMABLES

Foils
Various fluids
Radiochemical cartridge

MAINTENANCE MATERIALS

Target body, copper grid, etc.
Various metal parts activated
Air strainer

DECOMMISSIONING
2 ITECH Coaxial Detector
- 1 N-Type Crystal
- 1 P-Type Crystal
- INTERWINNER Software
- WINNERTRACK for MC

2 ORTEC Coaxial Detector
- 1 PROFILE –S P-Type Crystal
- 1 DX 100 PORTABLE P –Type Crystal
- GAMMAVISION Software

- Energy and Efficiency Calibration
  - Eckert & Ziegler Multi-gamma check source
  - 46.5 – 1836 keV; 60 – 1836 keV: different geometries
- Lead shielded enclosure
  - Low sample counts
  - Important to minimize background
  - 48 hr background spectrum before run
The chamber containing the target material is isolated with two metal sheets in Havar. 

Havar is an alloy consisting of:

<table>
<thead>
<tr>
<th>Percentage</th>
<th>Element</th>
</tr>
</thead>
<tbody>
<tr>
<td>42.5%</td>
<td>Co</td>
</tr>
<tr>
<td>20%</td>
<td>Cr</td>
</tr>
<tr>
<td>17.9%</td>
<td>Fe</td>
</tr>
<tr>
<td>13%</td>
<td>Ni</td>
</tr>
<tr>
<td>2.8%</td>
<td>W</td>
</tr>
<tr>
<td>2%</td>
<td>Mo</td>
</tr>
<tr>
<td>1.6%</td>
<td>Mn</td>
</tr>
<tr>
<td>0.2%</td>
<td>C</td>
</tr>
<tr>
<td>0.04%</td>
<td>Be</td>
</tr>
</tbody>
</table>
Foil
Foil

May 3rd, 2017
- Automated radiochemistry system
  - GE Heath Care Design
  - Converts externally produced 18F-fluoride into [18F]-Tracer

- FDG production via individual tracer cassettes

- We find a lot of this cartridge in collected waste
FDG Citrate Cassette

End Tubing
Trapping Cartridge
Eluent Vial
Left Hand 6 ml Syringe
Reaction Vessel
Reagent Vials
Hydrolysis Cartridge
Transfer Tubing
Purification Cartridge
Right Hand 6 ml Syringe
Alumina Cartridge
Radioactive waste

- Short-lived wastes
  - F18 and other PET isotopes
  - Decay-in-storage method (DIS)

- Long-lived wastes
  - Target components
  - **FDG Cassettes**
  - Need to characterize
    - Isotopes, activities
Isotope composition at irradiation

Co-56, 33.4%
Co-57, 0.9%
Mn-52, 7.7%
Mn-54, 0.5%
Ni-57, 5.3%
Re-181, 1.7%
Re-182, 0.4%
Re-184, 0.4%
Tc-95, 5.7%
Tc-95m, 1.8%
V-48, 0.1%
Y-88, 1.2%
Zn-65, 0.3%
Zr-89, 0.1%

May 3rd, 2017
Activity by counting sample

- Eluent Vial + 1 ml Syringe: 11.9%
- End Tubing: 3.6%
- Right Hand 6 ml Syringe: 0.5%
- Purification Cartridge: 0.9%
- Alumina Cartridge: 6.7%
- Transfer Tubing: 1.6%
- Hydrolysis Cartridge: 5.0%
- Reagent Vials Unit: 0.1%
- Reaction Vessel: 8.9%
- Trapping Cartridge: 47.6%

May 3rd, 2017
Summary of results

16 isotopes in 4 cassettes

- 0.8 – 312 day half-lives
- 122 – 1434 keV gammas
- 370 – 74k Bq activities
- More of these isotopes have half-lives longer than 75 days
- For the Italian law currently in force aren’t exempted
Activated and contaminated liquids

GAMMA SPECTROMETRY AND LSC ON ACTIVATED AND CONTAMINATED LIQUID
Summary of results
Activated and contaminated liquids

Vial 2014: H3: 1457 Bq/ml
Vial 2016: H3: 79557 Bq/ml
Activated materials

COPPER GRID END OF BEAM AT 2012

COPPER GRID END OF BEAM AT 2014
Activated materials

COPPER GRID END OF BEAM AT 2016
Activated materials

BODY TARGET F-18 END OF BEAM AT 2012

May 3rd, 2017
Management of radioactive waste gases from PET radiopharmaceutical synthesis can be integrated with a cyclotron safety system.
DECOMMISSIONING PROCEDURES FOR AN 11 MeV SELF-SHIELDED MEDICAL CYCLOTRON AFTER 16 YEARS OF WORKING TIME
In January 2004, it was decided to shut down the Cyclotron CTI RDS I 12 after more than 16 years of continuous operations in isotope production at the Nuclear Medicine Department of “Istituto di Ricovero e Cura a Carattere Scientifico (IRCCS) San Raffaele”.

The expected activation levels of various components, as reported in literature for high energy machines, are summarized in the following Table (European Commission 1999).
Before the beginning of the dismantling of the machine, in order to evaluate the risk for the staff involved, a Monte Carlo (MC) simulation code (MC-PX, MCNP4C2) was adopted to predict the activation of the cyclotron's components.

Table 2. Possible (p,n) reactions that generate radioactive elements with half-lives greater than 30 d in the extractor body and targets.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Reaction</th>
<th>Activated element</th>
<th>Half-life</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{65}\text{Cu}$</td>
<td>(p,n)</td>
<td>$^{65}\text{Zn}$</td>
<td>244.06 (d)</td>
</tr>
<tr>
<td>$^{109}\text{Ag}$</td>
<td>(p,n)</td>
<td>$^{109}\text{Cd}$</td>
<td>461.00 (d)</td>
</tr>
<tr>
<td>$^{181}\text{Ta}$</td>
<td>(p,n)</td>
<td>$^{181}\text{W}$</td>
<td>121.20 (d)</td>
</tr>
</tbody>
</table>
In general, a satisfactory agreement has been found between simulated and measured data.

A suitable calculation program, such as the cited Monte Carlo codes MCNPX and MCNP4C2, may be of great assistance in predicting activation generated by the direct proton beam and secondary neutrons nuclear reaction with the accelerator and vault components.

The decommissioning of a self-shielded cyclotron or 11 MeV, after a 16-y working life, represents no risk for the staff involved in the decommissioning, after a quite long time from the shut-down.
Due to presence of residual long-lived activated elements in various parts of the accelerator and shields, it’s unable to release the components into agreement of the waste classification, according with regulations and national laws.

Limits and legal prescriptions currently in force in Italy require the customer to store activated parts indefinitely in authorized areas for radioactive waste disposal.

In any facility, this aspect should be considered prior to a decision regarding cyclotron installation, especially if there are no spaces and permits for storage.
On order of IRCCS San Raffaele Milan, in 2016 CAMPOVERDE started to analyze the parts currently stored at the institute, needed to verify whether 13 years after the shutdown the radioactivity levels still present allow release in according to the applicable clearance levels.
Sample containing iron chips produced by drilling

Radionuclide | Concentration (Bq/g) | Uncertainty (Bq/g) | MAR (Bq/g)
--- | --- | --- | ---
$^{60}$Co | 3.85E-02 | 1.98E-03 | 3.42E-04
Decommissioning

R. Calandrino and others
Health Physics Department IRCCS San Raffaele. Milano, Italy

SAMPLE CONTAINING CONCRETE PRODUCED BY DRILLING

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Concentration (Bq/g)</th>
<th>Uncertainty (Bq/g)</th>
<th>MAR (Bq/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{60}$Co</td>
<td>1.97E-02</td>
<td>1.00E-03</td>
<td>5.52E-04</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>7.03E-02</td>
<td>4.75E-03</td>
<td>3.55E-04</td>
</tr>
<tr>
<td>$^{154}$Eu</td>
<td>8.07E-03</td>
<td>6.30E-03</td>
<td>4.76E-04</td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>3.57E-04</td>
<td>1.60E-04</td>
<td>2.52E-04</td>
</tr>
</tbody>
</table>
In a 400 ml Becker the sample aliquots are attached with 10 ml of HNO₃ 14M, excepted aluminum for which HCL 12M was used.
Adding distilled water, all the samples are brought to a final volume of 100 ml.
Separating the iron on TRU resin (Eichrom), the nickel is not retained and remains in solution.
Iron is eluted from the resin with HNO₃ 3M and collected in a 20 ml vial.

Nickel is separated with resin NI Eichrom, using C₄H₈N₂O₂ as extraction on inert support. Nickel is eluted with HNO₃ 3M and collected in a 20 ml vial.
Decommissioning

R. Calandrino and others
Health Physics Department IRCCS San Raffaele, Milano, Italy

<table>
<thead>
<tr>
<th>Isotopo</th>
<th>ID Campione</th>
<th>Concentrazione [Bq/kg]</th>
<th>Isotopo</th>
<th>ID Campione</th>
<th>Concentrazione [Bq/kg]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{55}$Fe</td>
<td>C-1</td>
<td>&lt;MAR</td>
<td>$^{63}$Ni</td>
<td>C-1</td>
<td>&lt;MAR</td>
</tr>
<tr>
<td></td>
<td>C-4</td>
<td>&lt;MAR</td>
<td></td>
<td>C-4</td>
<td>&lt;MAR</td>
</tr>
<tr>
<td></td>
<td>C-5</td>
<td>&lt;MAR</td>
<td></td>
<td>C-5</td>
<td>59,7 ± 2,1</td>
</tr>
<tr>
<td></td>
<td>C-6</td>
<td>&lt;MAR</td>
<td></td>
<td>C-6</td>
<td>42,9 ± 1,4</td>
</tr>
<tr>
<td></td>
<td>C-8</td>
<td>&lt;MAR</td>
<td></td>
<td>C-8</td>
<td>22,5 ± 5,8</td>
</tr>
<tr>
<td></td>
<td>C-9</td>
<td>&lt;MAR</td>
<td></td>
<td>C-9</td>
<td>81,4 ± 1,6</td>
</tr>
<tr>
<td></td>
<td>C-10</td>
<td>&lt;MAR</td>
<td></td>
<td>C-10</td>
<td>57,0 ± 2,2</td>
</tr>
<tr>
<td></td>
<td>C-11</td>
<td>&lt;MAR</td>
<td></td>
<td>C-11</td>
<td>46,9 ± 1,3</td>
</tr>
</tbody>
</table>

QUENCHING CURVE

misure standard $^{55}$Fe
$a_0+a_1x+a_2x^2+a_3x^3+a_4x^4+a_5x^5$, R²=0.997

misure standard $^{63}$Ni
$a_0+a_1x+a_2x^2+a_3x^3+a_4x^4+a_5x^5$, R²=0.999
Thank you