 Activation of Air and Concrete in Medical Isotope Production Cyclotron Facilities

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Outline

• Motivation
• Methods
• Air activation
• Concrete activation
• Conclusions
Motivation

- Neutrons produced in cyclotron target via \((p,Xn)\)
  - can activate materials in vault
  - air activation can pose a radiological hazard to workers
  - concrete activation can become a disposal problem
- Why the renewed CNSC interest?
  - cyclotrons have become much more powerful
  - old days ~ 60 µA and 2 hour runs for F-18 FDG production
  - now could have 500 µA and 6 hour runs for Tc-99m production
  - 25 times more activation!
Monte Carlo simulations

- MCNP5 (1% statistics)
- proton beams 18 & 24 MeV
- simplified vault geometry
- F-18 and Tc-99m neutron source spectra – assumed isotropic
- explore sensitivity of results to
  - changes in vault design
  - source energy
  - source location
  - polyethylene (with/without boron) shielding around targets
Methods (2/2)

Collaboration with UOIT co-op students

– Rob Shackelton (air activation)
– Devon Carr (concrete activation)
– Audrie Ismail (concrete activation)
Air activation

- **Nitrogen 78% of air**
  - N-14(n,p)C-14 (78% of air)
  - C-14: 5,730 year half-life, soft $\beta$ – little radiological consequence

- **Oxygen 21% of air**
  - O-16(n,p)N-16 (21% of air, 10 MeV threshold)
  - N-16: 7s half life, 6 MeV $\gamma$ – little radiological consequence

- **Argon ~ 1% of air**
  - Ar-40(n,$\gamma$)Ar-41 (0.93% of air)
  - Ar-41: 1.8h half life, hard $\beta$, 1.3 MeV $\gamma$

**Dominant hazard is Ar-41 activity**
Geometry and Materials

Go from complex to extremely simple
Reveals essential features and save computing time
Neutron Source Spectra (1/2)

- Neutron point source emulates target during irradiation (isotropic)
- F-18 thick-target spectrum from Mendez et. al. [1]
  \[ N(E) = 0.27 E^{0.45} e^{-\frac{E}{2.7}} \]
  - approximated by Maxwell fission spectrum
  - 150 logarithmically spaced energy bins
- Tc-99m spectrum from nested neutron spectrometer data [2]
  - Histogram representation
- All spectra automatically normalized by MCNP
Neutron Source Spectra (2/2)

- Maxwell fission
- F18 Spectrum
- Tc99 Spectrum (24 MeV protons)

Probability vs. Energy (MeV)
Neutron Source Placement

- 3m x 5m x 5m vault
- Eight source positions
- Starting at origin, moving diagonally into corner
- Considering statistical error, effect of source location is negligible

Effect of Source Placement on Ar-41 Production

- Percent difference from mean Ar-41 production

Distance from Origin (m)
Linearity result of large neutron mean free path in air ~62 m

Deviation from linearity for irregular shapes and very small vaults

Ar-40 captures vs. cube bunker edge length

Ar-40 Capture density vs. Cube edge length (m)
Cyclotron and Source Energy

• Iron cyclotron added to centre of the room
  – Slight decrease in production
  – Cyclotron density has no effect

• Source energy tests: (3m x 6m x 12m vault with cyclotron)

<table>
<thead>
<tr>
<th>Neutron energy</th>
<th>Ar-41 production/incident neutron/cm³</th>
</tr>
</thead>
<tbody>
<tr>
<td>F-18 spectrum</td>
<td>3.0 E-5</td>
</tr>
<tr>
<td>Tc-99 spectrum</td>
<td>3.6 E-5</td>
</tr>
<tr>
<td>Isotropic 1 keV</td>
<td>5.2 E-5</td>
</tr>
<tr>
<td>Isotropic 0.025 eV</td>
<td>12.0 E-5</td>
</tr>
</tbody>
</table>
Target Clamshell

- Polyethylene target shielding
  - 5, 10, 50cm thickness
  - No boron

- Critical thickness
  - Thin shield thermalizes neutrons
  - >10cm necessary to capture

- $\gamma$ shielding requirements
Partition Walls

- Wall placement below resulted in 15% decrease in Ar-41 production

- Objects in vault reduce air activation
- Justifies simplified geometry
Ar-41 Activity

• Results are in captures/neutron - How many neutrons per μA of beam?
  – IAEA TRS-468 gives saturation activity of F-18 at different beam energies; at saturation
• Neutron production rate = F-18 decays/s (Bq)
  – extrapolated to 24 MeV → 1.6 \times 10^{10} \text{n/s/μA}
• For Mo-99 (NNS @19 MeV) → 3.2 \times 10^{10} \text{n/s/μA}

• At saturation
• F-18 at 150 μA - (3 x 5 x 5 m vault) ∼2 mCi
• Tc-99 at 750 μA – (3 x 6 x 12 m vault) ∼ 12 mCi
## Dosimetry – Dose rates

All dose rates in $\mu$Sv/h

<table>
<thead>
<tr>
<th></th>
<th>F-18 @ 150 $\mu$A</th>
<th>Tc-99 @ 750 $\mu$A</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>3m x 5m x 5m</strong></td>
<td>5.1</td>
<td>17.3</td>
</tr>
<tr>
<td><strong>3m x 6m x 12m</strong></td>
<td>0.1</td>
<td>0.3</td>
</tr>
<tr>
<td><strong>5.1</strong></td>
<td>17.3</td>
<td></td>
</tr>
</tbody>
</table>

- Assumes saturation production
- Skin dose – upper limit – assumes no clothing
1. Objects in vault reduces air activation
   • Justifies simplified geometry
2. Air activation is not a problem for F-18
   • Results are for saturation of Ar-41 (half-life 1.8 h)
   • Runs are ~ 3 hours and typical F-18 runs are shorter
3. For Tc-99 may be a problem
   • Runs are ~ 6 hours and beam current ~ 3 times higher
4. Ventilation reduces problem dramatically
   • Less time to build up Ar-41 in vault and less exposure time – 1 hour air exchange time reduces dose by a factor of ~10
Concrete Activation

This produces radioactive waste, affecting decommissioning costs

1. How deep does it go?
2. What do polyethylene layers (with/without boron) do?
3. Is it on all inner vault surfaces? Or is it localized?
Decommissioning a cyclotron [6]

20-year-old 17 MeV Scanditronix cyclotron (~40 μA)

- 40 tons of low-level radioactive waste including the concrete vault wall
- Activities with $\tau_{1/2} > 1$ year

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Measured activity (Bq/g)</th>
<th>UCL (Bq/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Co-60</td>
<td>0.068</td>
<td>0.1</td>
</tr>
<tr>
<td>Cs-134</td>
<td>0.005</td>
<td>0.1</td>
</tr>
<tr>
<td>Eu-152</td>
<td>0.083</td>
<td>0.1</td>
</tr>
<tr>
<td>Eu-154</td>
<td>0.010</td>
<td>0.1</td>
</tr>
<tr>
<td>Mn-54 *</td>
<td>0.016</td>
<td>0.1</td>
</tr>
<tr>
<td>Total</td>
<td>0.18</td>
<td>0.1</td>
</tr>
</tbody>
</table>

UCL: unconditional clearance level at which material can be thrown out as non-radioactive,

* Made by fast neutrons via (n,p) reaction rather than (n,γ)
Literature Review (2/2)

Reactor study [7]

Ordinary concrete sample in TRIGA reactor in Slovenia

30 minute exposure @ neutron flux of $6.8 \times 10^{12}$ n/s/cm$^2$

Principal activities found Eu-152 and Co-60 at 6 Bq/g

Conclusion – concrete activation could be a problem with the new cyclotrons
Absorption depth

Neutron capture density in concrete (F-18 production)

- No Polyethylene
- 10 cm thick Polyethylene
- 10 cm thick Borated Polyethylene

Concrete depth (cm)
## Polyethylene around target

<table>
<thead>
<tr>
<th>Poly layer thickness (cm)</th>
<th>Percentage of neutrons captured in poly layer</th>
<th>Percentage of neutrons captured in borated poly layer</th>
</tr>
</thead>
<tbody>
<tr>
<td>5</td>
<td>17.1</td>
<td>32.0</td>
</tr>
<tr>
<td>10</td>
<td>52.0</td>
<td>65.8</td>
</tr>
<tr>
<td>15</td>
<td>72.5</td>
<td>82.9</td>
</tr>
<tr>
<td>20</td>
<td>82.6</td>
<td>91.1</td>
</tr>
</tbody>
</table>

- Results show how deep a sacrificial layer should be (if used)
- Regular poly is almost as good as borated poly
- Either option much cheaper than sacrificial layers in the vault
- And can be put in after vault construction
Spatial Distribution in Vault

We now know activation goes down to depth ~20cm
But this was averaged over whole inner vault surface
Is it on all inner vault surfaces? Or is it localized?

Investigated
1. Tc-99 vs F-18
2. Moving the source position inside the vault
3. Lateral distribution of neutron capture density within 1 side of the wall
1. Neutron Escape Percentage for Regular Poly Around Target - Tc-99 versus F-18 source

F-18 neutrons escape the polyethylene layer more easily than Tc-99's higher energy (more penetration)
2. Moving the Source Position on Y-Axis With 20-cm Thick Polyethylene Layer
2. Relative Capture Density of the Left Wall With Respect to Tc-99 Source Position

\[ y = 3.4e^{-0.002x} \]
\[ R^2 = 0.99 \]
3. Lateral Distribution of Neutron Capture Density in Near Wall

- Inverse Square Law
- 50cm away WO poly
- 50cm away WITH poly
- middle WO poly

At 50-cm distance radius of activation ~ 150 cm (10%)
1) Compare cyclotron neutron flux with TRIGA reactor flux
   • After 1 year at full operation → 0.35 Bq/g of Eu-152 and 0.33 Bq/g for Co-60 *(measurable)*
   • After 25 years operation → 7 Bq/g for Eu-152 and 3 Bq/g of Co-60

2) 100 x regulatory limit *for disposal as non-radioactive waste*

3) If no steps are taken, it will impact decommissioning cost and possibly financial guarantee
Conclusions

• **Air activation** – Not a big deal and easily controlled through
  – ventilation
  – restricted access for a few hours (normal to allow cyclotron to cool off)
  – detection of Ar-41 by area monitor in vault

• **Concrete activation**
  – could be a challenge for decommissioning
  – localized to concrete near target – including floor

  **For both problems, suggest borated poly around target**

• **Experiment** – activate sample of vault concrete & analyze by $\gamma$ spectroscopy
• Reactor neutron spectrum not quite the same as cyclotron neutron spectrum
• Your concrete may have different impurities
Questions?


Initial Results

- 3m x 5m x 5m empty vault with point source