Parasitic Isotope Production with Cyclotron Beam Generated Neutrons

LANL Isotope Program
Chemistry Division

UNCLASSIFIED
### TABLE 3.2. POPULAR RADIONUCLIDES VERSUS THE PROTON ENERGIES REQUIRED FOR THEIR PRODUCTION

<table>
<thead>
<tr>
<th>Proton energy (MeV)</th>
<th>Radionuclides easily produced</th>
</tr>
</thead>
<tbody>
<tr>
<td>0–10</td>
<td>F-18, O-15</td>
</tr>
<tr>
<td>11–16</td>
<td>C-11, F-18, N-13, O-15, Na-22, V-48</td>
</tr>
</tbody>
</table>

**Intermediate energies ~60 MeV and higher**

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Operating Intermediate Energy Facilities Worldwide

- LANL, USA – 100 MeV, 250 μA
- BNL, USA – 200 MeV, 100 μA
- INR, Russia – 160 MeV, 120 μA

- iThemba, South Africa – 66 MeV, 250 μA
- PSI, Switzerland – 72 MeV, 100 μA
- TRIUMF, Canada – 500, 70 MeV, 100 μA
- ARRONAX, France – 70 MeV, 2X 375 μA

From a production volume perspective, the IPF at LANL is presently the leading high power facility.
## Operating Intermediate Energy Facilities Worldwide

<table>
<thead>
<tr>
<th>Facility</th>
<th>Beam Energy (MeV)</th>
<th>Operating Beam Current (μA)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ARRONAX, France</td>
<td>70</td>
<td>2X 375</td>
</tr>
<tr>
<td>LANL, USA</td>
<td>100</td>
<td>250</td>
</tr>
<tr>
<td>iThemba, South Africa</td>
<td>66</td>
<td>250</td>
</tr>
<tr>
<td>INR, Russia</td>
<td>160</td>
<td>120</td>
</tr>
<tr>
<td>BNL, USA</td>
<td>200</td>
<td>100</td>
</tr>
<tr>
<td>TRIUMF, Canada</td>
<td>500, 70</td>
<td>100</td>
</tr>
<tr>
<td>PSI, Switzerland</td>
<td>72</td>
<td>100</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>New Facility</th>
<th>Beam Energy (MeV)</th>
<th>Beam Current (μA)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Deajeon, Korea</td>
<td>100</td>
<td>600</td>
</tr>
</tbody>
</table>
### The LANL experience – commercial isotopes


#### $^{82}\text{Sr}$ and $^{68}\text{Ge}$ produced on a large scale

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life</th>
<th>Main Use</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{82}\text{Sr}$</td>
<td>25.5 d</td>
<td>Parent of $^{82}\text{Rb}$ used in cardiac perfusion studies with PET</td>
</tr>
<tr>
<td>$^{68}\text{Ge}$</td>
<td>270 d</td>
<td>Positron emitter used in calibration sources for every PET scanner in clinical use</td>
</tr>
<tr>
<td>$^{88}\text{Zr}$</td>
<td>83.4 d</td>
<td>Parent of $^{88}\text{Y}$ used as a tracer surrogate for $^{90}\text{Y}$ in oncological bio-distribution studies</td>
</tr>
<tr>
<td>$^{22}\text{Na}$</td>
<td>2.6 a</td>
<td>Test objects for PET studies, tracer for natural Na</td>
</tr>
<tr>
<td>$^{32}\text{Si}$</td>
<td>153 a</td>
<td>Tracer for environmental transport studies</td>
</tr>
<tr>
<td>$^{73}\text{As}$</td>
<td>80.3 d</td>
<td>Tracer for toxicology studies</td>
</tr>
<tr>
<td>$^{109}\text{Cd}$</td>
<td>462.6 d</td>
<td>Source for X-ray fluorescence</td>
</tr>
<tr>
<td>$^{67}\text{Cu}$</td>
<td>2.6 d</td>
<td>Treatment of non-Hodgkin's Lymphoma</td>
</tr>
<tr>
<td>$^{225}\text{Ac}$</td>
<td>10 d</td>
<td>Alpha emitter used in cancer therapy clinical trials</td>
</tr>
<tr>
<td>$^{186}\text{Re}$</td>
<td>90.6 h</td>
<td>Bone pain palliation, cancer therapy</td>
</tr>
</tbody>
</table>

Other facilities has similar experience
Overview – Isotope Production Facility (IPF) at LANSCE

Proton Injectors

201.25 MHz Drift Tube Linac

H+

750 keV

H-

100 MeV Transition Region

805 MHz Side Coupled Cavity Linac

H+ SCCL is 90% of accelerator length

H-

800 MeV

Proton Storage Ring

Line X

Line D

Line A

Area A

Weapons Neutron Research Facility

Manuel Lujan, Jr. Neutron Scattering Center
Overview - Large Scale Production

- Production targets are routinely irradiated with 250 μA of 100 MeV protons
- Pulsed beam has a ring-shaped profile
- Three targets in a stack with cooling channels in between
- Production occurs simultaneously in 3 production energy windows

812 J/pulse

0.625 ms Beam on
32.675 ms Beam off
13,333 μA (1.3 MW)

Power density (kW/cm²)
Distance (cm)

IPF beam profile (ideal FWHM=1.27 cm)
One Target Stack Tuned for $^{82}\text{Sr}$ and $^{68}\text{Ge}$

Represents majority target configuration for IPF Challenge to allocate beam time to research and new isotopes
Lots of Secondary Neutrons

- For the $^{82}$Sr/$^{68}$Ge production target stack
- Model calculations predict a secondary neutron flux of $\sim 10^{12}$ n/s/cm² around the targets

MCNPX prediction

“nuisance” neutrons cause unwanted activation and unwanted production of impurities

Comparable with that of a medium-flux research reactor
Parasitic Production of NCA Isotopes Utilizing Secondary Neutrons

- Presents potential for useful production of No-Carrier-Added isotopes via (n,p), (n,2n) and (n,α) threshold reactions
- Candidate isotopes include $^{36}$Cl, $^{63}$Ni, $^{64}$Cu, $^{67}$Cu, $^{85}$Kr, $^{89}$Zr, $^{212}$Pb, $^{225}$Ac, $^{229}$Th, $^{231}$Pa, $^{237}$Np
- Inspection of reaction thresholds on target nuclei over a wide mass range shows that thresholds generally vary in the range 0-10 MeV

<table>
<thead>
<tr>
<th>Target Nucleus</th>
<th>(n,p)</th>
<th>(n,2n)</th>
<th>(n,α)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{16}$O</td>
<td>10.2</td>
<td>16.6</td>
<td>2.3</td>
</tr>
<tr>
<td>$^{36}$Cl</td>
<td>0</td>
<td>8.8</td>
<td>0</td>
</tr>
<tr>
<td>$^{70}$Zn</td>
<td>5.8</td>
<td>9.3</td>
<td>0.2</td>
</tr>
<tr>
<td>$^{148}$Nd</td>
<td>4.1</td>
<td>7.4</td>
<td>0</td>
</tr>
<tr>
<td>$^{226}$Ra</td>
<td>2.9</td>
<td>6.4</td>
<td>0</td>
</tr>
</tbody>
</table>
Parasitic Production of NCA Isotopes Utilizing Secondary Neutrons

- Approximately 50% of the neutrons have energies >1 MeV (25% > 5 MeV, 15% > 10 MeV)
- This high energy component is very suitable for inducing (n,p), (n,2n) and (n,α) type reactions
- MCNPX model calculations and experimental verification measurements suggests small scale production is feasible
- Work is championed by Post Doctoral Fellow, Jonathan Engle

Intermediate energy cyclotrons can take advantage
Commercial 70 MeV cyclotrons for large-scale production

ARRONAX machine, France

<table>
<thead>
<tr>
<th>Beam</th>
<th>Particle</th>
<th>Energy (MeV)</th>
<th>Current (μAe)</th>
<th>Dual beam</th>
</tr>
</thead>
<tbody>
<tr>
<td>Proton</td>
<td>H-</td>
<td>30-70</td>
<td>&lt;375</td>
<td>Yes</td>
</tr>
<tr>
<td></td>
<td>HH+</td>
<td>17.5</td>
<td>&lt;50</td>
<td>No</td>
</tr>
<tr>
<td>Deuteron</td>
<td>D-</td>
<td>15-35</td>
<td>&lt;50</td>
<td>Yes</td>
</tr>
<tr>
<td>Alpha</td>
<td>He++</td>
<td>68</td>
<td>&lt;70</td>
<td>No</td>
</tr>
</tbody>
</table>
Secondary neutron flux comparison

Secondary Neutron Flux from Rb+p

Energy (MeV) vs Flux (n cm² s⁻¹ MeV⁻¹)

- 100 MeV p
- 70 MeV p
- 40 MeV p
Example of small-scale production of NCA $^{225}\text{Ac}$

Production approaching mCi scale is possible at 100 MeV, 250 μA

- Sufficient to support pre-clinical research efforts
- 70 MeV and 40 MeV potential is lower but
- High current of emerging commercial machines can still take advantage
Advantages are compelling

- Production is parasitic with no impact on proton beam schedule
- No proton beam heating of targets
  - Allows a wide range of target material
  - Containment of target material is greatly simplified
- Recycling of expensive highly enriched target material is much simpler
  - Target mass can range from $mg$ to $g$ scale and beyond
  - No sophisticated hot cell based target fabrication capability required
**Summary**

- Several isotope production facilities are operating at intermediate energies, including high power cyclotrons.
- Often primary beam time is consumed for production of one or two isotopes (Sr-82, Ge-68 at LANL).
- Challenge to allocate beam time for small quantities of research isotopes.
- At LANL parasitic utilization of secondary neutrons shows promise for research scale production via threshold reactions such as (n,p), (n,2n) and (n,α).
- This approach can benefit other facilities, especially high power cyclotron facilities such as iThemba, ARRONAX and TRIUMF.
Acknowledgements

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