Medical Isotope Production With The Accelerator Production of Tritium (APT) Facility

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1.0 INTRODUCTION

In order to meet US tritium needs to maintain the nuclear weapons deterrent, the Department of Energy (DOE) is pursuing a dual track program to provide a new tritium source. A record of decision is planned for late in 1998 to select either the Accelerator Production of Tritium (APT) or the Commercial Light Water Reactor (CLWR) as the technology for new tritium production in the next century. To support this decision, an APT Project was undertaken to develop an accelerator design capable of producing 3 kg of tritium per year by 2007 (START I requirements). The Los Alamos National Laboratory (LANL) was selected to lead this effort with Burns and Roe Enterprises, Inc. (BREI) / General Atomics (GA) as the prime contractor for design, construction, and commissioning of the facility. If chosen in the downselect, the facility will be built at the Savannah River Site (SRS) and operated by the SRS Maintainance and Operations (M&O) contractor, the Westinghouse Savannah River Company (WSRC), with long-term technology support from LANL. These three organizations (LANL, BREI/GA, and WSRC) are working together under the direction of the APT National Project Office which reports directly to the DOE Office of Accelerator Production which has program authority and responsibility for the APT Project.

2.0 DESCRIPTION OF APT

The APT, which is shown schematically in Figure 1, is made up of four major subsystems which are described briefly below[1].

2.1 Accelerator System

The Accelerator System consists of the following subsystems:
- Proton injector to develop and maintain a continuous 100-mA current.
- Radio Frequency Quadrupole (RFQ) to focus and accelerate the proton beam to 7 MeV (kinetic energy)
- Coupled-cavity Linac (CCL) to accelerate the proton beam to 211 MeV.
- Superconducting Linac (SCL) to accelerate the proton beam to its final energy of 1700 MeV. The design is modular so that the portion of the accelerator up to 1030 MeV can be completed to produce 1.5 kg/yr of tritium and then a decision can be made as late as 2002 to add the last section if the START I production level is still required.

Fig. 1 APT Plant Layout
2.2 Target/Blanket (T/B) System

The Accelerator System provides a proton beam that is expanded and directed to a T/B assembly shown in Figure 2. The T/B assembly consists of a tungsten clad target in which neutrons are produced by spallation surrounded by a blanket in which additional neutrons are produced in lead. The neutrons are thermalized by collisions in the lead and in light-water and are subsequently captured in He-3 to produce tritium.

2.3 Tritium Separation Facility (TSF)

The TSF operates by extracting tritium from a tritium, hydrogen, and He-3 mixture returned from the T/B System in a recirculating He-3 loop. The He-3, hydrogen and tritium mixture also contains impurities such as water, methane, ammonia and small quantities of radioactive materials. Hydrogen isotopes are separated from the He-3 and sent to an Isotope Separation System where the tritium is separated from hydrogen. The He-3 is purified and recycled to the T/B assembly.

2.4 Balance of Plant (BOP) Systems

The BOP Systems support the integrated operation of the accelerator, T/B, and TSF, and provide the facility buildings that house them. The BOP System designs are driven by the required electric power input, the generated waste heat to be removed throughout each facility and the need to handle radioactive materials remotely.

3.0 MEDICAL ISOTOPE PRODUCTION WITH APT

The APT T/B will produce a high energy, high flux proton and neutron irradiation environment that is unique in the field of isotope production[2]. In addition to converting He-3 to tritium, it can be used to create isotopes for medical applications. Although the primary mission of the APT is to create tritium for the nuclear weapons deterrent, an ancillary isotope production mission is possible without significantly impacting tritium production. Because of the unique irradiation environment, and the large volume of space available, the APT has the potential to provide a significant source of research, diagnostic, and therapeutic isotopes to the medical community.

The capability for the irradiation of medical isotope targets is feasible within the scope of the T/B material surveillance program. The centerpiece of this program is a "rabbit" system that allows the insertion and removal of material surveillance coupons directly into several (up to seven) locations within the T/B assembly. Double wall tubes with a continuous flow of water provide a cooled environment for small (approximately 1.4 cm diameter by
6.3 cm long) target capsules. These capsules are moved in and out of the irradiation positions using hydraulic pressure. The design of the tungsten neutron source in the APT T/B allows for a large volume of potential irradiation positions directly in the high proton and neutron flux regions.

A small hot cell located nearby in an adjoining room is used to remove the capsules from the “rabbit” tubes and load them into shipping casks for transfer to a processing facility. Location of a private processing facility at the site boundary is a possibility. As an option to shipping casks, it is also possible to use pneumatic transfer tubes from the hot cell to the processing facility to speed the transfer.

Radioisotopes production in the APT target/blanket was analyzed to determine production rates and radiopurity of several isotopes that are of interest to the medical community. These include Cu-67, Ge-68, Sr-82, In-111, Re-186, Sm-153, Pd-103, P-32, Sc-47, and Ga-67. The isotopes were produced by nuclear spallation of natural (non-enriched) target materials by high-energy protons and neutrons. Significant production rates, high radiopurity and specific activity were achieved for most of the isotopes. In addition, the calculations showed that for 11 liters of target volume placed directly in the proton beam the decrease in tritium production was less than 2%.

4.0 THE POTENTIAL IMPACT OF APT ON MEDICAL ISOTOPE NEEDS

Leaders of the biomedical science community met recently in Dallas[3] to discuss the importance of developing an adequate supply of radionuclides to support clinical practice, research, education and training, and new treatments. The group acknowledged that the present supply of radionuclides is insufficient to meet current and projected future needs.

As an example, the recent development of specific delivery agents such as monoclonal antibodies that can target specific tumor cells and carry radiotherapeutic materials to those cells has led to a major advance in radiotherapy. Other healthcare areas with great potential include bone pain palliation, brachytherapy applications in inoperable tumors, nuclear cardiology and positron emission tomography (PET) procedures. For these to become available in routine practice there must be a supply of reasonably priced, high purity radionuclides.

The proposed APT facility has the potential to supply the much-needed radionuclides. Although the charter for APT does not include radionuclide production, the facility has the potential to make a significant contribution with little impact on tritium production. The biomedical community leaders learned of preliminary calculations of production yields, specific activities and radionuclidic purities for the nuclides listed above. This wide variety is possible from APT because, unlike reactor facilities, both energetic protons and neutrons are available in quantity.

Working as a team, the group developed the following statement in support of including radionuclide production in the APT charter:

“The APT facility will provide a unique resource for the production of substantial quantities of high specific activity radionuclides resulting in enormous scientific, research and education opportunities. These radionuclides will be especially useful in advancing healthcare for diagnosis and in the rapidly growing area of radionuclide therapy. We therefore support an expansion of the APT project charter to include designing into a biomedical radionuclide production capability. This initiative should not draw from existing radioactive materials research or production programs, as it is fundamental to the overall DOE mission, extending rather than replacing existing efforts, with downstream benefits which are vast for the nation.”

There was general agreement within the group that any new opportunity for radionuclide production and development gained by construction of the APT facility should be jointly pursued by the National Institutes of Health (NIH), DOE, and the Department of Defense (DOD), with NIH being the lead agency.

5.0 CONCLUSIONS

The APT will produce a high energy, high flux proton and neutron irradiation environment that is unique. Medical isotope production in APT has the potential to provide downstream benefits which have been judged by the biomedical community to be “vast for the nation”. Although the primary mission of the APT is tritium production, significant medical isotope production is possible with little impact on tritium production.

References

Medical Isotope Production with the Accelerator Production of Tritium (APT) Facility

Presented by M. R. Buckner (Westinghouse Savannah River Company)

Contributors: M. Cappiello and E. Pitcher (Los Alamos National Laboratory), H. O’Brien (O’Brien & Associates)

Linac 98
XIX International LINAC Conference Chicago, III.
August 23-28, 1998
DOE Dual Track Tritium Strategy

- Purchase Irradiation Services or Commercial Reactor
- Build Advanced Light Water Reactor (Small or Large)
- Build Modular High Temperature Gas-Cooled Reactor (MHTGR)
- Build Heavy Water Reactor (HWR)
- Build Proton Accelerator (APT) system

Commercial Reactor Option(s)
- Clarify institutional issues
- Resolve public policy issues

Backup Primary Path

DOE Decision FY 1998
- Resolve key technology elements
- Confirm / Improve cost estimate

Evaluation FY 1994 - FY 1995
Engineering Development FY 1996 - FY 2001
Commissioning Operation FY 1989
The APT Mission is to Provide a Tritium Production Facility for National Defense Using Accelerator-Based Technology

- Comprehensive, tested design that is cost-optimized and flexible, maximizing operability and safety, and minimizing environmental impacts.

- Annual production capacity sufficient to meet START-I requirements.

- Operational at Savannah River Site no later than 2007.
APT Project Management Team

DOE Program Manager, Staff Advisors
Single DOE Contract Officer (CO)
(Cost, Schedule, scope of contract with Prime Contractor
provides authority and direction to
Contract Officer Technical Representatives)

Project Director
(Project Direction, Principal COTR)
(Technical direction authority, task ordering, evaluation)
Staff Advisors, COTR Staff
Deputy Project Directors (LANL, BREI, WSRC)

Technology
(Conceptual Design, Engineering Development and Demonstration)
Los Alamos National Lab (Lead)
BREI, GA, WSRC
LLNL, BNL, SNL, TJNAF

Plant Project
(Plant Design, Construction, and Commissioning)
Burns & Roe Enterprises, Inc. (Lead)
GA, Other Laboratories
WSRC

Operations
(Production, Operations)
Westinghouse Savannah River Corporation (Lead)
LANL

- Small, dedicated staff
- Single Contracting Officer
- Best business practices
- Single point of authority
- Unified incentives and scorecard
Preferred APT Location Identified at SRS

Savannah River Site:
- 300 square miles
- Approximately 30 min. from Augusta airport
Top-Level APT Schedule - 3 kg/year Capacity CDR Plant

### Fiscal Year

- **Fiscal Year:** 96 97 98 99 00 01 02 03 04 05 06 07 08

#### Milestones:

- **CD-1 Approval of Mission Need**
- **Conceptual Design**
- **CD-2 Approve Cost and Schedule Baseline**
- **Preliminary and Final Plant Design**
- **Technology Downselect**
- **CD-3 Approve Construction Start**
- **Plant Construction**
- **Commissioning**
- **Production Certification**
- **CD-4 Plant Acceptance**
- **Plant Operation**
Target/Blanket:
- Neutron production from modular tungsten-lead neutron source
- tritium production from capture in $^3$He
- designed for 3 kg/year capacity

Tritium Separation Facility:
- Continuous Tritium separation
- Mature technology used at SRS and LANL for decades

Beam transport
RF power: Standard klystrons and power supplies
High-Energy Accelerator: Super-conducting linac with only two different structure designs
Low-Energy Accelerator: Normal-conducting
- Injector
- Radio Frequency Quadrupole
- Coupled cavity linac

Design capacity is 3 kg/year, with off-ramp to 1.5 kg/year operation
The Spallation Process

First Stage: The Intranuclear Cascade

- high-energy proton

Second Stage: Evaporation

Second Stage: Evaporation

What's Left: Spallation Product
Target/Blanket Major Design Features

Performance:
- Tritium Prod.: 11 gm/day*
- Tritium Inventory: 80 gm
- Beam Power: 170 MW
- Deposited Power: 127 MW

* Equivalent to 1000 MW-t Reactor

Vacuum Lines
Vessel Lid
Cavity Flood Pipe
Coolant and Gas Jumpers
Beam Diagnostics
Cavity Vessel
Beam Expansion Magnets
Beam Entrance Window
Target/Blanket Modules (17 Total)
Shielding
Target/Blanket Modular Arrangement

- Reflector
- Row 1
- Row 2
- Row 3
- Row 4
- Row 5
- Decoupler
- Peak Flux Irradiation Position
- Tungsten Neutron Source Ladder Structure
- Lead Blanket with 3He for Tritium Production
Proton Flux

Graph showing proton flux with axes labeled x (cm) and z (cm). The flux values are indicated on the right side of the graph.
Neutron Flux
Rabbit Tubes Provide Mechanism for Target Irradiations

- Rabbit Tube
- Front View of Ladder No. 2
- Stacked Rabbits Containing Material Surveillance Coupons or Isotope Production Targets
- Tungsten Neutron Source Ladder Structure
- Double Wall Rabbit Tube; Light Water Cooling
Rabbit Targets Removed/Packaged in Hot Cell

- Rabbit targets irradiated for desired time
- Moved to hot cell using hydraulic pressure
- Removed from piping and placed in transfer cask
- Optional: transferred to processing facility using pneumatic transfer line
How is APT Different from Other Isotope Producers?

- Reactors typically rely on low-energy neutron capture reactions that produce isotopes of the same element as the target isotope, e.g.,

\[ ^{102}\text{Pd} \ (n,\gamma) \ ^{103}\text{Pd} \]

whereas accelerators traditionally use proton-induced reactions, e.g.,

\[ ^{103}\text{Rh} \ (p,n) \ ^{103}\text{Pd} \]

- APT has both intense proton and intense neutron fluxes at its disposal for isotope production
  - reactors have only neutrons
  - accelerators traditionally rely solely on protons
How is APT Different from a Reactor?

- Neutrons in APT are much more energetic than those in a reactor
  - mean energy of neutrons in the neutron source of APT is 14 MeV
  - mean energy of neutrons in a fast reactor is ~300 keV

- This allows APT to utilize neutron-induced reactions with high threshold energies that are not readily accessible to reactors, e.g.,

\[ ^{32}\text{S} \,(n,p) \, ^{32}\text{P} \]

whose threshold energy is 0.96 MeV
Characteristics of APT-Produced Radioisotopes

- Can produce neutron-deficient isotopes not produced by reactors due to the proton flux (e.g., $^{68}$Ge, $^{76}$Br, $^{82}$Sr, $^{52}$Fe, $^{72}$Se)

- Can produce significant quantities of certain neutron-rich isotopes due to the high energy of the neutrons (e.g., $^{166}$Dy, $^{188}$Re, $^{105}$Rh, $^{143}$Pr)

- High production rates due to high neutron and proton fluxes

- Typically lower radiopurity than reactor-produced isotopes because spallation produces more than a single isotope of an element

- Typically higher specific activity than reactor-produced isotopes because the target element can be different from that of the medical isotope
Representative Radioisotope Production in APT

- 100-cc target volumes irradiated behind the second ladder in the tungsten neutron source
  - other positions may be considered upon further optimization, but this position provides peak production rates for the isotopes considered to date

- Targets irradiated for a time equal to the half-life of the radioisotope (i.e., half saturation)

- Assumed cycles per year based on 90% usage of APT's 75% availability
# Representative Isotope Production Rates

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Target</th>
<th>Irradiation Time (days/cycle) / (cycles/year)</th>
<th>Single-Cycle / Annual Production* (Ci / kCi)</th>
<th>Radiopurity (%)</th>
<th>Specific Activity (kCi/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu-67</td>
<td>Zn</td>
<td>2.58 / 96</td>
<td>140 / 13</td>
<td>52</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>(3 days cooling)</td>
<td></td>
</tr>
<tr>
<td>Ge-68</td>
<td>As</td>
<td>271 / 1</td>
<td>77 / 0.077</td>
<td>94</td>
<td>0.033</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>(100 days cooling)</td>
<td></td>
</tr>
<tr>
<td>Sr-82</td>
<td>Nb</td>
<td>25.4 / 10</td>
<td>190 / 1.8</td>
<td>43</td>
<td>2.6</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>(10 days cooling)</td>
<td></td>
</tr>
<tr>
<td>In-111</td>
<td>Sn</td>
<td>2.8 / 88</td>
<td>400 / 35</td>
<td>76</td>
<td>94</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>(1 day cooling)</td>
<td></td>
</tr>
<tr>
<td>Re-186</td>
<td>Os</td>
<td>3.78 / 65</td>
<td>780 / 50</td>
<td>11</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>(10 hours cooling)</td>
<td></td>
</tr>
<tr>
<td>Sm-153</td>
<td>Gd</td>
<td>1.93 / 128</td>
<td>31 / 4.0</td>
<td>91</td>
<td>7.0</td>
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<td></td>
<td></td>
<td>(2 days cooling)</td>
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<tr>
<td>Pd-103</td>
<td>Ag</td>
<td>17 / 14</td>
<td>830 / 12</td>
<td>91</td>
<td>28</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>(10 days cooling)</td>
<td></td>
</tr>
<tr>
<td>P-32</td>
<td>S</td>
<td>14.3 / 17</td>
<td>3100 / 53</td>
<td>98.6</td>
<td>108</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>(10 hours cooling)</td>
<td></td>
</tr>
<tr>
<td>Sc-47</td>
<td>Ti</td>
<td>3.35 / 74</td>
<td>1000 / 77</td>
<td>70</td>
<td>190</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>(1 day cooling)</td>
<td></td>
</tr>
<tr>
<td>Ga-67</td>
<td>Ge</td>
<td>3.26 / 76</td>
<td>110 / 8.1</td>
<td>86</td>
<td>24</td>
</tr>
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<td></td>
<td></td>
<td></td>
<td>(3.3 days cooling)</td>
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*Based on a 100-cc target volume.
APT Medical Isotope Chronology

7/97  A study, "Evaluation of Medical Radionuclide Production with the APT Facility", demonstrated the significant production capabilities of APT.

11/97  Regional Workshop in Augusta - strong interest from biomedical community but better quantification of potential yields needed.

5/98  Medical Isotope Workshop in Dallas - 24 biomedical leaders signed proclamation supporting incorporation of medical isotope production capabilities in APT.

6/98  Panel discussion on "Promising New Treatments in Nuclear Medicine" at National Press Club in Washington, D.C. - review of conclusions/recommendations from Dallas Workshop. Follow-up meeting with DOE officials to emphasize the interest of nuclear medical community.
The APT will produce a high energy, high flux proton and neutron irradiation environment that is unique. Medical isotope production in APT has the potential to provide “downstream benefits which are vast for the nation”.

Although the primary mission of the APT is tritium production, significant medical isotope production is possible with little impact on tritium production.