#### **Isotope Production at High Energy**

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### **Facility Description**

- The Brookhaven Linac Isotope Producer (BLIP) was the world's first facility to seriously exploit the isotope production capabilities of a high energy proton accelerator.
  - The use of higher energy particles allows the use of relatively thick targets, where the large number of target nuclei can compensate for the generally smaller nuclear reaction cross sections compared to low energy reactions.
  - The BLIP, built in 1972, utilizes the excess beam capacity of the 200 MeV proton Linac that injects into larger synchrotrons (Booster, AGS, RHIC) at BNL. A 30m long transport line delivers the protons to a shielded target area for radioisotope production. The facility has been upgraded twice, in 1986 and 1996.
  - The target area consists of an underground 2.44m diameter tank containing sand shielding, a water filled 40cm diameter shaft, 9.2m high, and a 20cm diameter inspection shaft.
  - The target assembly is immersed at the bottom of the shaft and cooling water is forced individually past the faces of the target disks.



#### Figure 1. BNL Accelerator Complex (AGS=Alternating Gradient Synchrotron, RHIC=Relativistic Heavy Ion Collider)





### **Brookhaven LINAC Isotope Producer (BLIP)**

Figure 2. The LINAC supplies protons and polarized protons to the Booster for nuclear physics. Excess pulses (~85%) are diverted to BLIP. Energy is incrementally variable from 66 to 202 MeV and can be changed pulse by pulse.

Figure 3. The BLIP beam line directs protons up to 115µA intensity to targets; generally parasitic operation with nuclear physics programs.







#### Figure 4. LINAC BEAM LINES







Figure 5.

### **BLIP Operating Parameters**

- Energy 66, 93, 118,139,161, 81, or 202 MeV variable pulse by pulse
- Pulse current 38mA, pulse rate 6.67 Hz, pulse width 420µs
- Maximum integrated current 115µA, typical integrated current (parasitic mode) -85µA
- Beam profile Gaussian, FWHM horizontal 2.2cm, vertical 1.5cm
- Power 7.6MW instantaneous, 22 kW integrated
- Polarized protons pulse current 250µA, pulse rate 2 per 3.8s, pulse width ~400 µs; pulse to pulse switching with high intensity protons
- Target capacity up to 8 simultaneously, dependent on beam energy & target thickness
- Schedule Due to high operating costs BLIP generally runs in a parasitic mode, sharing pulses (~85% share), and costs (~30% share) with the driver nuclear physics programs at RHIC. The schedule and duration of Linac operation is largely determined by the plans and funding of the nuclear physics experiments, not isotope production needs. Recent increases in isotope sales have allowed extended runs of 26 weeks per year independent of RHIC.
  - Production coordination with other sites in Los Alamos, S. Africa, Russia has helped year round availability of longer lived, high value isotopes, such as Sr-82 and Ge-68.



### **Typical BLIP beam intensity profile**

normal Gaussian tune





# **Scientific Approach**

- Nuclear physics issues
  - Investigate proper nuclear target by determination of nuclear excitation functions for desired isotope and probable impurities by literature search, calculation or direct measurement. Irradiations for physics needs, particularly radiation damage studies, are also often performed (for FermiLab, KEK, and soon FRIB)
- Target design
  - Pick material that has appropriate physical and chemical properties (density, isotopic abundance, solubility, purity, availability, low corrosivity), good mechanical and thermal properties (high melting and boiling points, high thermal conductivity, high stability, low vapor pressure)
- Radiochemistry development
  - Design chemical separation scheme to yield desired isotope in high chemical and radiochemical purity. Ion exchange chromatography, solvent extraction and distillation techniques used
  - Develop assay methods for radiopurity by gamma ray spectroscopy and/or beta counting by liquid scintillation
  - Develop assay methods for chemical purity by ICP-OES, ICP-MS &HPLC
  - Develop labeling methods in template systems, for example by linking to monoclonal antibodies, as a final test of product usefulness
- Package and ship
  - Assorted sizes of DOT approved containers are used for radioisotope transport both domestically and internationally



# **Target Types**

- Target thermal properties are the most critical issue due to the high average and pulsed power
  - Target thermal and mechanical behavior in beam are modeled with finite element software
  - Pure metal disks are favored when possible, but dissolution for processing can be slow
  - Pressed salt pellets often used if elemental form is not practical
- Since all the targets are immersed in radioactive cooling water, most BLIP targets are encapsulated in metal cladding in order to obtain high chemical and radiopurity
  - The cans are typically fabricated as rings (2.75"OD) with thin electron beam welded windows(0.012")
  - Cladding materials are usually stainless steel, but inconel is sometimes used for enhanced corrosion resistance
  - Liquid Ga used to produce Ge-68 is encapsulated in niobium



#### **Target array schematic**





# **Target Issues**

- Window bowing
  - Welding process can cause window deformation during fabrication
  - Two atmospheres of pressure from air and water depth can deform windows as can internal melting of target material
  - Window bowing can be inward or outward
  - This effect makes prediction of the energy loss of the protons propagating through many layers of water and multiple targets somewhat uncertain
- Failures can occur at peripheral welds mostly due to mechanical strain and metal fatigue or in the faces due to pitting (Ga targets have been most problematic)
- The Gaussian beam profile creates very high power density at its peak
  - Target heat distribution is non uniform, especially with salt targets having poor thermal conductivity (e.g., in SrCl<sub>2</sub> central temperature >874°C, surface temperature ~100°C)
  - Experiments to create a flatter beam profile with sets of octupole magnets are underway



### **Target Thermal Processes**

- Complex coupled problem
- Material properties are highly dependent on temperature
- Solid Target thermal path involves
  - Conduction
  - Heat transfer coefficient (water film)
- Molten Target
  - Buoyancy forces that drive convection
  - Heat transfer coefficient (inside)
  - Partially molten target (volume and shape of melt zone





#### Ga target thermal model (ANSYS) with convection



#### Ga target stress analysis from beam heating



### **Considerations in Gas Targets**



U.S. DEPARTMENT OF ENERGY

#### Hot cell sealable target:



- Enriched <sup>68</sup>Zn (to make <sup>67</sup>Cu) and <sup>86</sup>Sr (<sup>86</sup>Y) is quite expensive and economy requires recovery and reuse.
- Recovered target will be radioactive (eg. <sup>65</sup>Zn & <sup>85</sup>Sr) after first irradiation-hot cell target assembly is required
- Both halves are Al for better conductivity
- Head screws large enough for remote manipulator handling
- Silver coated stainless steel C-ring to provide adequate seal
- Target design was successfully irradiated without any leakage and can be opened and sealed in a hot cell.
- Modification to increase active area in beam may increase production levels but will require more enriched <sup>68</sup>Zn or <sup>86</sup>Sr to be used.



### **The Target Processing Laboratory**

- After irradiation all targets require chemical processing to separate the desired isotope from many others coproduced and from the bulk target material. To support this effort we have:
  - 9 hot cells
  - 8 radiochemistry development labs
  - Instrumentation lab for radionuclide and chemical assays
  - radwaste handling and storage facilities for both liquids and solids
  - Staff machine shop
  - For human use isotopes we maintain current good manufacturing practice (CGMP) registration with FDA



Figure 7. View of several processing hot cells



### **Radiochemical Processing Techniques**

#### Ion exchange chromatography

 Widely applicable, highly selective, reasonably rapid, adaptable to remote or automated operation

#### Solvent extraction

 Can be selective, rapid at small scale, adaptable to remote or automated operation but may involve toxic solvents

#### Distillation

Limited applicability and speed

#### Precipitation

Limited to low specific activity situations, outside hot cells



### Cu-67 process flow chart



## Transportation

- Radioisotopes are shipped every day
  - In the US FedEx does this routinely and reliably
  - Rigid, detailed Department of Transportation regulations apply (not DOE or NRC)
  - Approved containers vary widely in size and can be very expensive
  - International shipments are more difficult, and often go by passenger plane. The pilot has final say.
  - International Air Transport Association (IATA) rules, DOT and destination country rules all apply



#### **Additional Needed Support Facilities**

- Central fabrication facility with advanced capabilities
- Central hazardous and radwaste storage and disposal facility
- Trained isotope packaging & shipping group with assorted sizes of approved containers for radioisotope transport both domestically and internationally



### **Present BNL Isotope Menu**

<u>Isotope</u> Be-7	<u>~# shipped/yr</u> <1	<u>Half- life</u> 53.3d	Typical application γ source
Fe-52	2	8.3h	Nanoparticles
Zn-65	25	244d	Zn tracer
Cu-67	4	61.9h	Radioimmunotherapy
Ge-68/Ga-68	60	271d/68m	PET scanner calibration
As-73	1-2	80.3d	environmental tracer
Sr-82/Rb-82	70	25.5d/75s	Cardiac diagnosis
Y-86	3	14.7h	Cancer imaging
Y-88	2	106.6d	Y-90 tracer, γ source
Tc-95m	<1	61d	Tc tracer

Previously developed: Na-22, Mg-28, Sc-47, Co-55, Rb-81, Tc-96, Ru-97, Cd-109, Sn-117m, Xe-122, Xe-127, Pb-203



## Acknowledgements

#### This work was supported by the United States Department of Energy, Office of Nuclear Physics - Isotope Research and Production Program, under Contract # DE-AC02-98CH10886.

