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Spallation Production of Neutron Deficient Radioisotopes in North America

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SUMMARY. The United States Department of Energy produces a number of neutron deficient radioisotopes by high energy proton induced spallation reactions in accelerators at Los Alamos National Laboratory in New Mexico and Brookhaven National Laboratory in New York. Research isotopes are also recovered from targets irradiated at TRIUMF in British Columbia, Canada. The radioisotopes recovered are distributed for use in nuclear medicine, environmental research, physics research, and industry worldwide. In addition to the main product line of Sr-82 from either Mo or Rb targets, Cu-67 from ZnO targets, and Ge-68 from RbBr targets, these irradiation facilities also produce some unique isotopes in quantities not available from any other source such as Al-26, Mg-28, Si-32, Ti-44, Fe-52, Gd-148, and Hg-194. We will describe the accelerator irradiation facilities at the Los Alamos and Brookhaven National Laboratories. The high level radiochemical processing facilities at Los Alamos and brief chemical processes will be described.

1. INTRODUCTION

The United States Department of Energy produces a number of neutron deficient radioisotopes by high energy proton induced spallation reactions in accelerators at Los Alamos National Laboratory in New Mexico and Brookhaven National Laboratory in New York. Spallation reactions generally result from interaction of >100MeV particles with a nucleus.

This paper will describe the accelerator irradiation facilities at the Los Alamos and Brookhaven National Laboratories. The high level radiochemical processing facilities and brief chemical processes will be discussed.

International cooperation contributed to the year round supply of ^{67}Cu with target irradiation performed at all three facilities.

A number of rare and exotic radioisotopes not available anywhere else in the world are produced and distributed to researchers around the world. These radioisotopes range from ^{10}Be and ^{26}Al to ^{194}Hg and most are recovered from targets many mass numbers above.

2. THE ACCELERATORS

The US DOE operates two medium energy particle accelerators that have radioisotope production missions. One is located in New Mexico at the Los Alamos National Laboratory and the other is located in New York at the Brookhaven National Laboratory. Research isotope targets are also irradiated at TRIUMF in Vancouver, British Columbia and processed at one of the National Laboratories.

Isotope production is supplemental to the primary physics research at all three facilities and isotope production capabilities at each of the facilities are different.

Los Alamos National Laboratory[1] uses the LANSCE (Los Alamos Neutron Science Center) Facility. This accelerator generates 800 MeV H^+ protons at nearly 1 mA of current. This accelerator is nearly 1 km long and the isotope production facility is located in front of the beam stop. A total of nine target stations permit irradiation of nine targets at once with the beam traversing through each target in succession. A specially designed target holder can hold three experimental targets, although normally there

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is only one target per station. Greater than 90% of the initial 1 mA of 800 MeV protons impinge on the first target. Most of the current is deposited in these targets before the beam exits into the beam stop.

Brookhaven National Laboratory[2] utilizes the BLIP (Brookhaven Linac Isotope Producer) Facility. A maximum of 145 μA of 200 MeV H^- protons are stripped from the end of the linac injector for the Alternating Gradient Synchrotron research facility. Available energy ranges from 66-200 MeV in 21 MeV increments. There are seven target holders, each capable of holding three disc shaped targets. The initial 200 MeV energy protons can be used for isotope production by spallation reactions only in the first or second targets. The other targets are used to produce radioisotopes by high energy (p,xn) type reactions.

TRIUMF (Tri Universities Meson Facility) located at the University of British Columbia has a 500 MeV cyclotron. 150 μA of H^- are available for spallation isotope production on beamline 1A. Only one target at a time may be irradiated in this facility. This facility is also used by UBC and MDS Nordion for isotope production by spallation reactions.

Zinc oxide targets for production of 2.58 day ^{67}Cu have been irradiated at TRIUMF, shipped to Los Alamos National Laboratory for chemical processing and shipped around the world for medical research.

THE REACTIONS

Spallation reactions generally result from interaction of >100 MeV particles, either neutrons or charged particles, with a nucleus[3,4]. Reaction products generally reach a maximum yield at 10-20 mass numbers below the target mass, drop off rapidly and rise again in the low mass nuclei. For instance, a common product in all of the targets is ^7Be, produced as a spallation fragment. The choice of target therefore has a large effect on the desired product's yield and radiopurity. Spallation products are generally neutron deficient and the spallation fragments are neutron rich. This permits the production of such exotic radioisotopes as long-lived neutron rich ^{10}Be with very high specific activity which may be recovered after the co-produced ^7Be decays away. Typical radioisotopes and yields from a molybdenum metal target for spallation production of ^{82}Sr from LANL molybdenum targets are listed in Table I as an example of spallation yields. Only one example of each of the elements detected is listed.

TABLE I
Typical Production in Mo Targets*

^{95m}Tc	300-500 mCi
^{91m}Nb	2000-3000 mCi
^{88}Zr	5000-6000 mCi
^{88}Y	5000-8000 mCi
^{82}Sr	8000-12000 mCi
^{83}Rb	5000-7000 mCi
^{75}Se	2500-3500 mCi
^{73}As**	800-1000 mCi
^{68}Ge	200-400 mCi
^{65}Zn	300-500 mCi
^{58}Co**	150-250 mCi
^{59}Fe**	10-20 mCi
^{54}Mn**	10-20 mCi
^{51}Cr**	20-40 mCi
^{48}V**	5-10 mCi
^{22}Na**	5-10 mCi
^7Be	300-500 mCi

* 30 days after End of Bombardment 45 day irradiation.
** Identified/assayed - not recovered for distribution

THE TARGETS

All three facilities use disc shaped targets that the proton beam traverses. Large heat deposition from the proton beam must be considered in the target choice and design to prevent leakage or vaporization while in beam. Desired products and recovery chemistry also dictate the choice of target material. The products will be concentrated at 10-20 mass numbers below the mass of the target as demonstrated by the choice of molybdenum with an average mass of 96 to produce ^{82}Sr.

5. THE FACILITIES

The main radioisotope chemical processing facilities at Los Alamos National Laboratory

consist of twelve inter-connected hot cells plus a universal dispensary cell. All targets and waste enter and exit the facility from this dispensary cell. A large electric train running under the cells permits easy transfer of materials between cells. An additional facility with sixteen hot cells in two separate banks of eight is also available for chemical processing if needed.

Additional facilities in the main radiochemistry laboratories consist of six chemical fume hoods that are used for recovery of small quantities of radioisotopes and for chemistry procedure development. Analysis of stable metal elements content is possible on all radioactive products using either computer controlled DCP-AES or ICP-AES instruments located within the facility.

Radiochemical analysis for gamma emitting radioisotopes produced at Los Alamos is performed with three high purity germanium detector systems connected to a MicroVAX computer system.

These detector systems are located throughout the chemical processing facility and are calibrated for activity levels ranging from nanocuries to curies. Analysis for low energy x-ray, beta and alpha emitting radioisotopes is done using capabilities of the Radiochemistry Counting Room. This facility has nearly seventy different state of the art computer controlled detector systems available for use.

Recent major upgrades at the Brookhaven National Laboratory[2] include the addition of two new hot cells for a total of seven available for radiochemical processing. An upgraded ventilation system and liquid waste handling disposal system were also installed to bring BNL into compliance with current environmental regulations.

As with the sister laboratory at Los Alamos, Brookhaven National Laboratory also has available a number of radiochemistry hoods. A recently installed ICP-AES permits stable metal element analysis of isotope products.

Radiochemical analysis for gamma emitting radioisotopes produced at Brookhaven National Laboratory is performed with high purity germanium detector systems. These detector systems are located within the chemical processing facility.

Both BNL and LANL chemistry facilities are approved by the U.S. Food and Drug Administration for production of Bulk Pharmaceutical Components.

6. THE CHEMISTRY

Spallation reactions are not selective and elements ranging from the next element above the target down to hydrogen are produced in varying quantities. Chemical separation techniques have been developed to recover the radioisotopes of interest in both high radiochemical purity and yield. These chemical processes have also been required to reduce or eliminate the generation of mixed waste.

The only isotope common to all three facilities is ^{67}Cu produced in natural abundance ZnO targets. The actual production reaction mechanism is complex with other high energy reactions such as (p,2p) and (p,α) on ^{67}Zn and ^{70}Zn contributing significant quantities of product. This radioisotope has been chemically processed only at LANL and BNL. Each facility uses a different method of chemical recovery of the ^{67}Cu.

Los Alamos National Laboratory uses an electrochemical method of recovery of the ^{67}Cu from sulfate solutions after dissolution of the zinc target in dilute H_2SO_4. This is followed by simple anion and cation exchange for final purification.

Brookhaven National Laboratory has developed a recovery method based on use of Chelex 100. After target dissolution in HCl the material is evaporated to dryness and redissolved in buffer solution. The zinc target material is not sorbed on the Chelex and after washing the resin the ^{67}Cu and other radioisotopes are desorbed and purified using conventional anion and cation exchange methods.

The separated zinc solutions can be further

processed to recover ^{48}V, ^{49}V, ^{7}Be and isotopes of scandium. These solutions have recently been found to contain significant quantities of ^{44}Ti and investigations are underway to recover and distribute this rare isotope to the research community.

Large quantities of ^{68}Ge have been produced by spallation reactions on RbBr targets[5,6]. After dissolution in HCl the ^{68}Ge is recovered by distillation from 6 M HCl followed by extraction in CCL_4 to separate spallation produced selenium and arsenic radioisotopes. Very high specific activity ^{73}As has been produced as a byproduct of this ^{68}Ge production.

Microcurie quantities of ^{26}Al and ^{32}Si have been separated from KCl targets irradiated for 4-6 months[7]. The irradiated KCl target is dissolved in H_2O and filtered. The filtrate containing the ^{32}Si is acidified, converted to a heteromolybdate complex and sorbed on Sephadex. After washing away the KCl target material the pure ^{32}Si is stripped from the column with dilute base. The water insoluble fraction is dissolved in HCl and the ^{26}Al is recovered by ion exchange.

Brookhaven National Laboratory uses 192 MeV protons on KCL targets to produce ^{28}Mg by spallation. The targets is dissolved in H_2O and the Mg recovered by precipitation and solvent extraction.

Tantalum targets have provided large quantities
of ^{148}Gd in addition to microgram quantities of $^{178m2}Hf$ produced by spallation reactions. Dissolution of the tantalum in HNO_3-HF was followed by precipitation on CaF_2. Dissolution of the CaF_2 in HCl-HBO_3 was followed by ion exchange for purification.

In the past silicon targets have been used to produce ^{10}Be, and ^{194}Hg has been recovered from bismuth targets. The ^{10}Be was recovered after distillation of the silicon target material as the fluoride. No carrier added ^{194}Hg was recovered by extraction into ethyl acetate after the bismuth target was dissolved in dilute HNO_3.

Molybdenum targets irradiated for ^{82}Sr production are dissolved in H_2O_2 and the isotopes of Zr, Y, Sr, Rb, Zn, Fe, Mn, Cr, Na, and Be sorbed on a cation exchange resin[8,9]. These cations are stripped from the resin with HCl, evaporated and resorbed on a new cation exchange column. Washing the resin with dilute H_2SO_4 removes all of the isotopes present except ^{82}Sr and ^{88}Y. The ^{82}Sr is removed from the resin with dilute HCl and the ^{88}Y is not removed from the resin. The recovery of ^{88}Zr, ^{83}Rb, ^{65}Zn and ^{7}Be from the sulfate solution is accomplished by ion exchange.

The molybdate eluate from the first column is used to recover ^{68}Ge and ^{95m}Tc using Sephadex ion exchanger and Reillex HPQ resins.

192 MeV protons on ^{58}Ni targets have been used at Brookhaven National Laboratory to produce ^{52}Fe. Recovery and purification of the ^{52}Fe was accomplished by anion exchange.

CONCLUSIONS

The high currents available at the three accelerators combined with spallation reactions are an effective method to produce quantities of unique radioisotopes not possible at any other accelerator in the world. These radioisotopes have found use in nuclear medicine, environmental research, physics and industry worldwide.

The US DOE has successfully irradiated ZnO targets in three facilities throughout North America, chemically processed and distributed the recovered spallation produced ^{67}Cu to research facilities throughout the world. Irradiation at all three facilities was needed to continually produce ^{67}Cu year round in spite of the maintenance shutdown requirements of the various facilities.

Specific chemical techniques have been developed for the recovery of the spallation produced radioisotopes.

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