PARASITIC ISOTOPE PRODUCTION WITH CYCLOTRON BEAM GENERATED NEUTRONS*

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Abstract

Several LINAC and cyclotron facilities worldwide generate high intensity beams with primary beam energies in the range 66 MeV to 200 MeV for isotope production purposes. Many of these beams are almost fully subscribed due to the high demand for isotopes produced via proton induced reactions, leaving little beam time available for production of smaller quantities of research isotopes. Modelling and preliminary experimental measurement of the high power proton beam interaction with targets at the Isotope Production Facility (IPF) at Los Alamos show a high potential for parasitic small scale production of isotopes utilizing the secondary neutron flux generated around the target. Such secondary neutron flux can also be exploited by cyclotron facilities, especially emerging modern commercial 70 MeV machines with total beam currents approaching 1 mA and more.

INTRODUCTION

Popular radioisotopes whose production requires a proton beam with primary energy of 30 MeV or higher include ^{124}I, ^{123}I, ^{67}Ga, ^{111}In, ^{11}C, ^{18}F, ^{13}N, ^{13}O, ^{82}Sr, ^{68}Ge, ^{22}Na and ^{48}V [1]. Some of these isotopes are exclusively made in large quantities at facilities with energy capability beyond 60 MeV. Limited beam time for small-scale production of other research isotopes is therefore a known problem at most operating intermediate energy isotope production facilities such as those listed in Table 1. Usually the production of one or two commercial isotopes dominates operations, consuming most of the beam time.

Table 1: Operating Intermediate Energy Isotope Production Facilities

Facility	Country	Beam Energy and Current	Type
IPF, LANL	USA	100 MeV, 250 μA	LINAC
BLIP, BNL	USA	200 MeV, 100 μA	LINAC
INR	Russia	160 MeV, 120 μA	LINAC
iThemba	South Africa	66 MeV, 250 μA	Cyclotron
PSI	Switzerland	72 MeV, 100 μA	Cyclotron
TRIUMF	Canada	500, 70 MeV, 100 μA	Cyclotron
ARRONAX	France	70 MeV, 2x375 μA	Cyclotron

THE LANL EXPERIENCE

Production operations also dominate irradiations at the 100 MeV Isotope Production Facility of the Los Alamos National Laboratory (LANL), which is based on a linear accelerator. Presently, the production of the commercial isotopes ^{82}Sr and ^{68}Ge represents more than 95% of the production volume. Tightly scheduled routine production runs proceed at a beam current of 230 μA, employing a standardized production target stack consisting of two RbCl targets and one gallium target [2]. Recent decline in beam availability resulted in the concurrent decline of small-scale production of research isotopes to almost non-existent levels. This prompted the consideration of alternative parasitic production approaches.

Secondary Neutrons at IPF

The IPF's high proton beam current produces a secondary neutron flux with a scale that is typically beyond the reach of low energy cyclotrons (<30 MeV) and energetically distinct from reactor neutron fluxes. This potential for research in novel methods of isotope production and materials science is as yet unexploited. Current irradiations utilizing a standard production target stack result in a secondary neutron flux of ~10^{12} n s^{-1} cm^{-2}, approaching the scale of medium flux research reactors at the location directly behind the target stack (see Fig. 1).

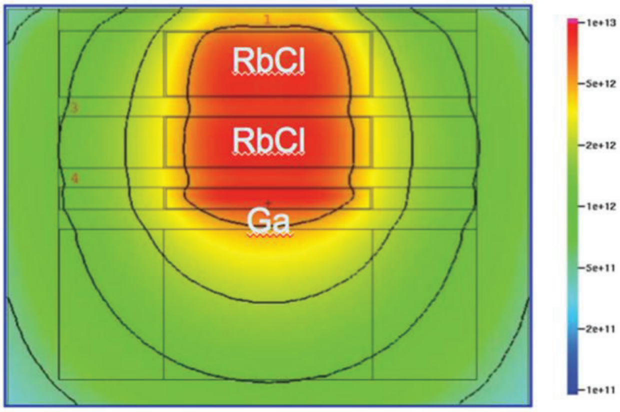

Figure 1: Neutron flux distribution (n s^{-1} cm^{-2}) around the IPF target stack as predicted by MCNPX [3]. The proton beam direction is downward.

Parasitic Production Potential at LANL

Parasitic production target material inserted at the rear of the target stack may advantageously utilize this neutron flux. As the energetics of the IPF neutron field are also predicted to differ significantly from typical reactor

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energy spectra, novel opportunities for isotope production become available.

The (p,xn) reactions induced by the incoming proton beam result in emission of neutrons with a spectrum of energies which extends from a thermal minimum up to a maximum near the energy of the incoming proton. According to Monte Carlo predictions, approximately half of the secondary neutron flux has an energy in excess of 1 MeV. This portion of the flux is therefore generally capable of inducing threshold reactions such as (n,xn), (n,p), (n,pxn), (n,α), and (n,αxn), which are relevant to production of small quantities of no-carrier-added (NCA) isotopes. Present candidates targeted for parasitic production at LANL include isotopes such as ^{36}Cl, ^{47}Sc, ^{63}Ni, ^{64}Cu, ^{67}Cu, ^{85}Kr, ^{89}Zr, ^{224}Ra, ^{225}Ac, ^{229}Th, ^{231}Pa and ^{237}Np.

SECONDARY NEUTRONS FROM CYCLOTRONS

In 2006 there were more than 200 radioisotope-producing cyclotrons worldwide [4]. Of these about 10% were capable of producing proton beams with energies beyond 40 MeV, and, depending on available beam intensity, these machines could potentially take advantage of parasitic production using secondary neutrons. The existing operating isotope production machines with energies >66 MeV (see Table 1) are obvious candidates while facilities based upon the emerging commercial high current 70 MeV cyclotrons are particularly interesting in this regard. These machines are capable of delivering dual beams at a maximum beam current of 375 μA each.

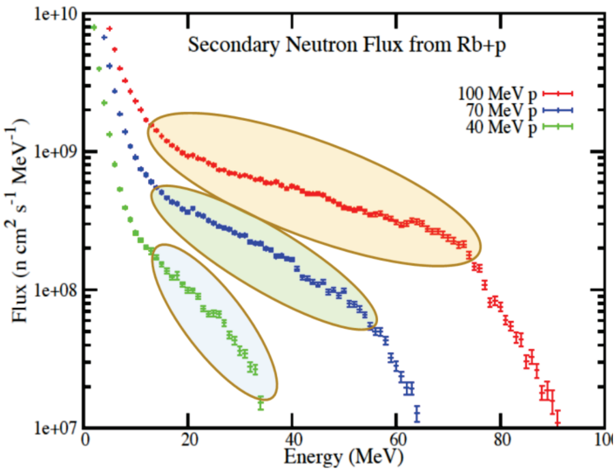

Figure 2: Comparative differential neutron flux distribution (n s^{-1} cm^{-2} MeV^{-1}) predictions for different 250 μA proton beams. The shaded areas indicate the most productive regions for threshold reactions. Secondary neutrons originating from a 40 MeV incident proton beam were modelled to impinge upon a target of liquid ^{nat}Ga, since the production of ^{82}Sr from bombardment of ^{85}Rb is not possible in this energy range.

Figure 2 compares predicted secondary neutron flux distributions for cyclotron generated proton beams with different energies. In order to provide a sense of the relative potential for parasitic production, neutron spectrum ranges with the greatest potential utility are indicated in the shaded areas.

Figure 3 depicts representative excitation functions for the neutron induced reactions that we have found to be relevant to parasitic radioisotope production using the IPF secondary neutron flux. The wide distribution of energies in a secondary neutron flux entails advantageous use of differential decay rates, where possible, to achieve desired radioisotopic purities.

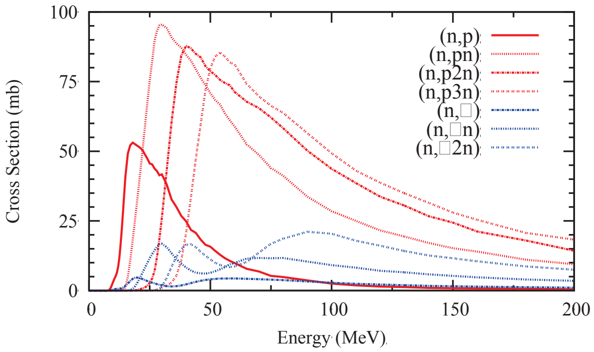

Figure 3: General excitation functions for neutron-induced transmutation reactions, with (n,pxn)-type reactions shown in red and (n,αxn)-type reactions shown in blue.

The relative production potential of the IPF neutron flux is further illustrated in Fig. 4, assuming a 250 μA beam and using the production of ^{225}Ac via ^{226}Ra(n,2n)^{225}Ra$\rightarrow$^{225}Ac as an example. This isotope is one of the priority therapy isotopes targeted for production at LANL. The figure shows that millicurie-scale parasitic production is possible with a 250 μA, 100 MeV beam. This is sufficient to support preclinical

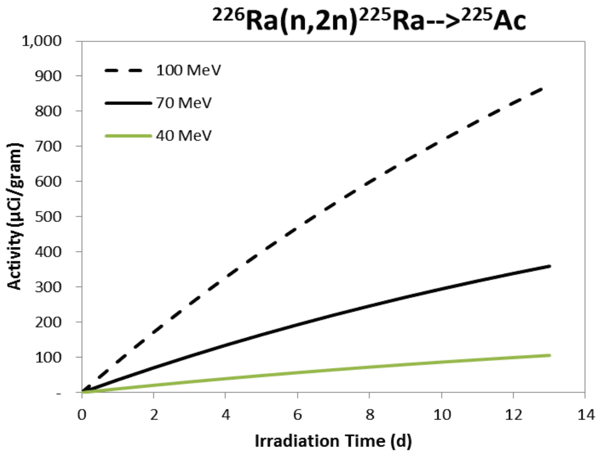

Figure 4: Parasitic secondary neutron production yields for the ^{225}Ra precursor of therapy isotope ^{225}Ac, using the ^{226}Ra(n,2n)^{225}Ra$\rightarrow$^{225}Ac production route as an example.

research efforts. The figure also shows that, as expected, machines delivering lower energy proton beams have lower parasitic production potential. However, some of the more modern machines have higher beam currents, which to some extent will compensate for the lower production rate. This is especially true for the emerging commercial 70 MeV cyclotrons designed for isotope production and for drivers in the generation of radioactive beams.

CONCLUSIONS

Several isotope production facilities are presently operating at intermediate energies. These include high power cyclotrons at national facilities. As is the case at LANL, primary beam time at these facilities is often dedicated to the large-scale production of one or two commercial isotopes, making it a challenge to allocate adequate beam time for production of small quantities of research isotopes and for R&D efforts. MCNP modelling predicts that the 100 MeV proton beam at IPF generates a secondary neutron flux of ~10^{12} n s^{-1} cm^{-2} with an MeV-scale energy distribution that favours transmutation reactions. Parasitic utilization of the secondary neutrons holds promise for research scale radioisotope production via threshold reactions such as (n,xn), (n,p), (n,pxn), (n,α), and (n,αxn) without disruption of important production irradiations. More importantly, this approach can also benefit high power cyclotron facilities, including iThemba, TRIUMF and PSI as well as newer isotope production facilities such as ARRONAX, which are based upon emerging high-intensity commercial 70 MeV cyclotrons.

ACKNOWLEDGMENTS

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