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Determination of spallation neutron flux through spectral adjustment techniques

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A B S T R A C T

The Los Alamos Isotope Production Facility (IPF) creates medical isotopes using a proton beam impinged on a target stack. Spallation neutrons are created in the interaction of the beam with target. The use of these spallation neutrons to produce additional radionuclides has been proposed. However, the energy distribution and magnitude of the flux is not well understood. A modified SAND-II spectral adjustment routine has been used with radioactivation foils to determine the differential neutron fluence for these spallation neutrons during a standard IPF production run.

1. Introduction

The Isotope Production Facility (IPF) produces isotopes both for medicine and research using a 100 MeV proton beam generated by the Los Alamos Neutron Science Center (LANSCE). The proton beam is impinged on a target stack whose multiple targets subtend different energy regions of the incident proton beam. In practice, the high demand for certain routinely produced isotopes results in oversubscription of the primary proton beam time.

The interaction of the proton beam with the target stack is known to also generate secondary spallation neutrons. Use of these spallation neutrons has been proposed to symbiotically generate nuclei of medical and scientific interest, especially those that cannot be produced by the IPF primary proton beam or in reactors where the neutron energy distribution is distinctly different and dominated by thermal neutrons. The secondary neutron flux created at IPF is of particular interest to create isotopes not feasible through other methods.

Proton bombardment is useful for creating neutron-deficient nuclei, generally positron emitters. Low-energy neutrons can be used for capture reactions, which produce nuclei one neutron richer than the target. Neutron-rich nuclei further from stability can only be produced through high energy neutron reactions, i.e. (n,p), (n,pn), (n,α), (n,2n), (n,3n), etc. Additionally, high energy neutrons induce nuclear transmutation, forming radionuclides absent added carrier mass. These small masses of radioactive product are nevertheless highly detectable thanks to their radioactive emissions, enabling study of biological processes without perturbing the physiological system or treatment of disease that is highly specific to receptor systems with very low in vivo density. Radionuclides of interest to produce using high energy neutrons include ^{67}Cu and ^{47}Sc, both 2 neutrons richer than stability and desired in quantity by the medical community.

For the purposes of yield and purity predictions of the residual nuclei created, the IPF secondary neutron flux has been modeled in MCNPX but has not been directly verified experimentally due to spatial challenges imposed by the production facility configuration. Direct measurement of the neutron flux via time-of-flight is not feasible as the target chamber for IPF is located 12 m below a hot cell in a compact, heavily shielded water-filled irradiation chamber.

The measurement of neutron fluxes in reactors, which often present similar spatial challenges, has been accomplished using radioactivation foils as an indirect probe. Spectral adjustment techniques are then used to adjust predictions of the flux using quantified radioactive residuals produced in these foils. A similar approach is presented here, using the radioactivation of selected foils to probe the secondary neutron flux produced at IPF. The results are compared with calculated activities from an MCNPX simulation, and the calculated neutron flux is adjusted using a modified SAND-II code in order to maximize agreement with the experimental measurements.

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2. Material and methods

This experiment and resulting activity measurements are described in detail in [1]. In summary, high-purity metal reaction foils were placed downstream of the traditional foil stack used in IPF production irradiations. A schematic of the standard target stack used in the production of ^{82}Sr and ^{68}Ge is shown in Fig. 1. This stack consists of two targets of Inconel-encapsulated rubidium chloride and a third of niobium-encapsulated liquid gallium metal, in which the proton beam is stopped. Seven foils, punched into discs with diameter 10.32 ± 0.05 cm to match calibrated geometries for later analysis, were used as activation foils: Al, Co, Y, Au, Bi, Ni, Zn. The activation foils were mounted directly downstream of the standard RbCl-RbCl-Ga IPF target stack, placing the foils in a neutron flux consistent with IPF production conditions. Radioactivation foil samples were enclosed in two single layers of 25 μm thick Kapton tape to facilitate trapping of gaseous, radioactive products and to isolate samples from one another during irradiation.

The irradiation foil stack was exposed to standard IPF production conditions. The target stack received a 200 μA proton current for four minutes to thermally condition upstream RbCl targets followed by a 230 μA proton current for one hour.

Following irradiation, the foils were removed and transported to the LANL Nuclear and Radiochemistry group counting room for γ-spectrometry using a high-purity germanium (HPGe) ORTEC GEM p-type aluminum-windowed detector. Details of the counting procedure and subsequent analysis have been reported previously in [2,3]. The photopeaks from the γ-spectra were analyzed using an in-house code, SPECANAL, using gamma energies and intensities from the National Nuclear Data Center's (NNDC's) database. The activity at end of irradiation was determined from an exponential fit to the measured decay curve.

2.1. Modifications of adjustment procedure

The SAND-II [4] code is a statistical adjustment procedure that iteratively modifies the neutron flux based on measured activation foil yields and calculated yields determined by using the input flux and known cross sections, where the input flux's characteristics are generally obtained from a simulation such as MCNP. The activity (a) in each activation foil is calculated by:

$$a(t) = \lambda_n m_o \int_0^{\infty} \sigma_{mn}(E) \int_0^{t} \phi(E, t')dt'dE \tag{1}$$

where λ_n is the decay constant for nucleus n, m_o is the initial number of target atoms, σ_{mn} is the cross section for the reaction producing the atom n and ϕ is the differential flux, which is a function of energy and time. The calculated activities are compared with the measured activities to obtain a weighting function. The resulting correction term is given by

$$C_j^{[k]} = \sum_{i=1}^{n} w_{i,j}^{[k]} lnR_i^{[k]} \Bigg/ \sum_{i=1}^{n} w_{i,j}^{[k]} \tag{2}$$

where $R_i^{[k]}$ is the ratio of measured to calculated activity and $w_{i,j}^{[k]}$ is the activity weighting function:

$$w_{i,j}^{[k]} = \frac{1}{2}\left(A_{i,j}^{[k]} + A_{i,j-1}^{[k]}\right) \Big/ A_i^{[k]}. \tag{3}$$

The flux distribution is then adjusted:

$$\phi_j^{[k+1]} = \phi_j^{[k]} \exp c_j^{[k]} \tag{4}$$

These equations are applied iteratively until the solution converges to a specified standard deviation between calculated and measured activities or a change of less than 0.1% between the standard deviations of run iterations is achieved.

Modification of the SAND-II code was required to adjust fluxes up to 100 MeV, as the code has defined energy bins that extend only to 20 MeV. Historically, this procedure has been applied to reactors and critical assemblies where the neutron energies are well within the 20 MeV maximum window allowed by the code. The energy binning was adjusted to accommodate these larger energies, now extending to 200 MeV to allow for future adjustments of faster spallation neutron fluxes. A new cross section library was also required as the energy bins of the cross section

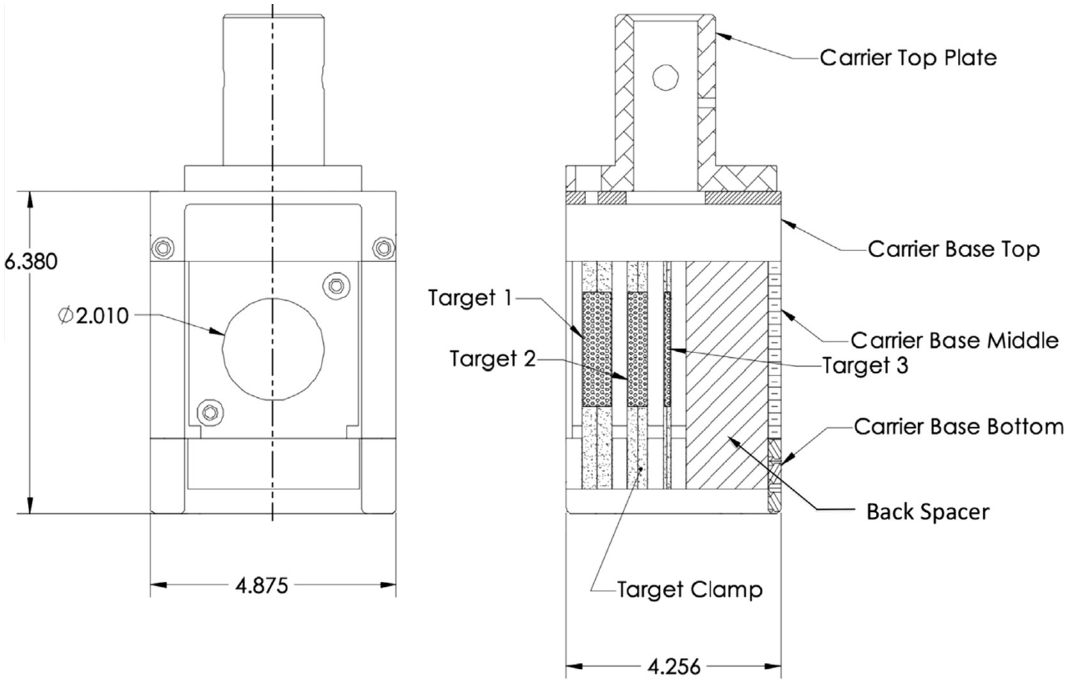

Fig. 1. Schematic of the IPF target stack from the front (left) and side (right). All dimensions are in inches.