Final Technical/Scientific Report
DE-SC0017215

Sponsoring Office: Office of Science, Isotope Program
Recipient: Dr. Jonathan Engle

Title: Final Technical Report on Nuclear Data for Spallation Neutron Radioisotope Production

Consortium:
iThemba National Laboratory, South Africa
Berkeley National Laboratory, California
Los Alamos National Laboratory, New Mexico

Disclaimer: The content of this report is appropriate for public release.

Abstract:

Over 50 million nuclear medicine procedures are performed annually, leading to a multi-billion dollar market for radioisotope production. The demand for new medical and research isotopes is growing, and radioisotope supply is insufficient. Most radioisotope production today utilizes charged particle or low-energy neutron irradiation of a target. Isotope production with tens to hundred MeV incident energies is a relatively unexplored option. There is a tremendous opportunity associated with a growing number of suitable domestic and international facilities buttressed by hundred million dollar global investments (e.g., the Los Alamos and Brookhaven Isotope Production Facilities, the European Spallation Source in Lund, and the Korean Multi-purpose Accelerator Complex in Gyeongbuk). In part due to a lack of supporting nuclear data that would make modeling radioisotope yields and purities possible, these facilities do not utilize their high-energy neutron fluxes for isotope production. This project attempted to measure neutron reaction excitation functions relevant to the large-scale production of critical radioisotopes, enabling development of cost-efficient isotope production methods, contributing to the improvement of theoretical models, and enhancing the value of national isotope production facilities. Reactions which form ^{67}Cu, ^{32}Si, and alpha-emitting isotopes like ^{225}Ac were chosen for their consistent prioritization by expert panels, representation of diverse reaction mechanisms, and relative lack of supporting nuclear data. Accurate measurement of these data is presently made only using quasi-monoenergetic neutron beams, which are produced by bombarding thin lithium targets with protons at only a few laboratories in the world. Ultimately, the projects efforts were incompletely accomplished due to a combination of multi-year technical issues at the only laboratory in the world which can make the proposed measurements, iThemba Labs in South Africa, and the onset of a global pandemic, which precluded the significant effort and time required to develop an independent measurement capability in the United States.

Project Summary

Significance and Impact

Most radioisotope production today utilizes charged particle or low-energy neutron irradiation of a target. Isotope production using neutrons with 10^$^{1-2}$ MeV incident energies is a relatively unexplored pathway. There is a tremendous opportunity associated with a growing number of suitable domestic and international facilities buttressed by hundred million-dollar global investments (e.g., the Los Alamos and Brookhaven Isotope Production Facilities, the European Spallation Source in Lund, and the Korean Multi-purpose Accelerator Complex in Gyeongbuk). In part due to a lack of supporting nuclear data that would make modeling radioisotope yields and purities possible, these facilities do not utilize their high-energy neutron fluxes for isotope production. This work was to measure neutron reaction excitation functions relevant to the large-scale production of critical radioisotopes, enabling development of cost-efficient isotope production methods, contributing to the improvement of theoretical models, and enhancing the value of national isotope production facilities. The radionuclide production market for nuclear medicine applications is projected to exceed 20 billion dollars by 2025. The demand for new medical and research isotopes continues to grow, and the Nuclear Science Advisory Committee (NSAC) has identified dozens of radioisotopes whose supply is insufficient. Reactions to form such radionuclides, e.g., ^{67}Cu, ^{32}Si, and alpha-emitting isotopes like ^{225}Ac, were chosen for their consistent prioritization by NSAC panels, representation of diverse reaction mechanisms, fit to project personnel expertise, and relative lack of supporting nuclear data. Accurate measurement of these data is presently made using quasi-monoenergetic neutron beams, which are produced by bombarding thin lithium targets with protons at only a few laboratories in the world. These laboratories' experimental focus has not yet been brought to bear on the potential for fast neutron-induced radioisotope production and is anticipated to be helpful to disciplines using fast neutron nuclear data for material sciences, dosimetry, and nuclear model creation. The project's engagement of graduate students in processes of radiochemical separations chemistry, nuclear data measurement, accelerator physics and Monte Carlo simulation techniques fills numerous explicitly identified voids in several national policy documents spanning decades, particularly in the areas of nuclear medicine, nuclear physics and radiochemical expertise. Students supported to engage this work had unique opportunities to engage research questions with real-world applications in the context of their dissertation research and through collaborative engagement with some of the most famous international facilities in the world, all the while maintaining a hands-on connection to investigations they were compelled to drive experimentally and conceptually from within an institution internationally reputed for the quality of its mentoring.

Original Hypotheses/Objectives

The project's original objectives were to:

1. Establish collaborative experimental arrangements and procedures with facilities that can produce quasi-monoenergetic neutron (QMN) fluxes in the range of 10-200 MeV.
2. Prioritizing radioisotopes emphasized by 2009 and 2014 NSAC meetings, measure energy-dependent formation excitation functions between 10 and 200 MeV relevant to

production of these isotopes at DOE isotope production facilities (IPF, BLIP). Measurements will be accomplished using the activation method.
3. Validate experimental and analytical methods employed by energetically overlapping measurement of suite of established excitation functions using different facilities, and use collected data as feedback for the improvement of theoretical nuclear codes (e.g., MCNP).
4. Modify libraries of existing spectral deconvolution codes (e.g., SAND-II) with measured data to incorporate neutron-induced reactions with tens of MeV threshold energies, and
5. Characterize neutron flux-energy distributions at relevant radionuclide production facilities using foil activation experiments.
6. Validate collected nuclear data with radioisotope yields measured from reference “white” spectra activation experiments.

Approaches
Collaborative experimental plans and PAC proposals with investigators at iThemba National laboratory were established by 2017. These PAC proposals were necessary to secure the devotion of significant resources at the iThemba quasi-monoenergetic neutron (QMN) user facility, the only one of its kind in the world to have developed validated neutron flux energy distributions suitable to make energy differential measurements of the nuclear formation excitation functions targeted by this proposal. iThemba PAC proposals defended remotely in 2017 and 2018 were awarded experimental beam time in the 2018 and 2019 run cycles, respectively, accomplishing the first objective above.

The methods for so-called “activation-style” quantification of nuclear formation excitation functions are well established by 60 years of experimental nuclear data science, and the facility at iThemba labs is designed to accommodate these measurements. Following PACS beam time awards, preliminary experiments were planned as proposed and materials with natural isotopic distributions procured. Additional isotopically enriched materials (sulfur-36) were also purchased anticipating follow-on full-scale experiments where simulations predicted radiochemical separations and high purity starting materials would be necessary to achieve quantification of residuals production with satisfactory uncertainty.

Unforeseeably, staff at iThemba identified and communicated problems related to operation of the QMN facility beginning in early 2018. Early problems were technical in nature, with facility staff believing they needed additional beam and primary lithium target upgrades as a matter of operational course, and the larger collaboration of interested international researchers (which included users from multiple European and South African laboratories) agreed to defer experiments to the 2019 run cycle. Plans were developed to share beam time amongst multiple target assemblies (possible because of the low stopping power for neutrons for most of the materials being irradiated) to maximize the user efficiency of the 2019 cycle. In 2019, the iThemba facility informed its global user base that a neutron dosimetry problem in the secondary target fault made operation under established conditions unsafe, and planned mitigation and remediation efforts that would entail disassembly of the neutron collimating shield leading into the secondary target position. These efforts were expected to require the majority of the calendar year and caused an additional one year delay of all objectives in the planned experiments for this project.

To hedge against the possibility that experiment at iThemba would be unsuccessful, the project team also undertook an unproposed attempt to establish an independent excitation function measurement capability in the U.S.. This effort benefited from collaboration with Lawrence Berkeley National Laboratory (LBNL) and the University of California – Berkeley (Bernstein group). A set of lithium targets of varying thickness was designed and fabricated with Inconel-welded encapsulation for the primary beam energies of the 88” cyclotron facility. Funding requests were made to additional DOE mechanisms to use these targets to characterize the excitation functions of several important reactions that would have been useful to develop the facility in Cave 5 at LBNL and at the Crocker Nuclear Laboratory’s analogous 88” machine and train experimentalists at this location (e.g., LAB17-1763), but were unfortunately unsuccessful.

The unfunded collaborative relationships with the University of California and Lawrence Berkeley National Laboratory were nevertheless successful in irradiation experiments using the thin Inconel-encapsulated lithium targets at the 88" cyclotron at incident energies of 15, 22 and 40 MeV. These experiments were focused on characterization of the emitted QMN flux and employed a mixture of neutron targets giving access to well-described reactions in the hopes of using the activation method to unfold the differential neutron energy distribution (Al, Co, In, Ni, Zn, and Zr). Solid state Si detectors and a ToF system were also tested for the first time during these experiments, with data taken at 0° and 20° to explore subtraction of the effects of the anticipated low energy tail of the neutron energy spectrum. Chris Kutyreff, a graduate students at the University of Wisconsin, traveled to Berkeley and spent 8 days working locally with LBNL staff (Lee Bernstein, Darren Bluel, Bethany Goldblum) and students (Andrew Voyles, Jonathan Morrell) to conduct the experiments and assay the irradiated materials. Analysis of results showed anticipated levels of activation reactions forming products via n,2n and n,3n channels, and provide a solid foundation for moving to higher energies which are more comparable to the energies accessible at iThemba, LANL and BNL. These measurements were presented at the International Nuclear Data meeting in Beijing in 2019.

Los Alamos National Laboratory was also engaged as a collaborator. White spectrum activation experiments were planned at the Los Alamos IPF in 2018 and 2019 but delayed by Los Alamos staff due to problems obtaining consistent beam parameters in beam development runs. A student from Wisconsin, Kaelyn Seeley, traveled to LANL when these experiments were anticipated, and was able to make and publish two other proton excitation function measurements when the neutron slot was unavailable. Preliminary analysis of these data were also presented as the International Nuclear Data meeting in Beijing in 2019.

Between 2020 and 2022, the global pandemic eliminated any hope of international travel for scientific purpose and stopped planning for awarded experimental beam time at iThemba. In the hopes that white-spectrum activation measurements of high-value targets could continue past the project’s endpoint, isotopically enriched starting materials transfer agreements between the UW and LANL for sulfur-36, are ongoing.

Conclusion

The potential of high energy secondary neutrons from spallation sources remains an important avenue for research investigation of isotope production possibility, and for characterization of the products of high-energy charged particle productions that may be influenced by a secondary neutron flux. This project was delayed over four years by unforeseeable and experimentally catastrophic extenuating circumstance that will require revisioning of original ambitions and, ideally, funded development of domestic QMN facilities capabilities in the post-pandemic environment.