INL/CON-08-13955
PREPRINT

Performance of a New Composite Single-Crystal Filtered Thermal Neutron Beam for Neutron Capture Therapy Research at the University of Missouri

13th International Congress on Neutron Capture Therapy

John Brockman
David W. Nigg
M. Frederick Hawthorne
Charles McKibben

November 2008

Performance of a New Composite Single-Crystal Filtered Thermal Neutron Beam for Neutron Capture Therapy Research at the University of Missouri

John Brockman[a], David W. Nigg[b], M. Frederick Hawthorne[a], Charles McKibben[a]

[a] *University of Missouri, Columbia, MO USA*
[b] *Idaho National Laboratory, Idaho Falls ID USA*

Abstract

Parameter studies, design calculations and initial neutronic performance measurements have been completed for a new thermal neutron beamline to be used for neutron capture therapy cell and small-animal radiobiology studies at the University of Missouri Research Reactor. The beamline features the use of single-crystal silicon and bismuth sections for neutron filtering and for reduction of incident gamma radiation. The calculated and measured thermal neutron flux produced at the irradiation location is on the order of 9.5×10^8 neutrons/cm^2-s, with a measured cadmium ratio (Au foils) of 106, indicating a well-thermalized neutron spectrum with sufficient thermal neutron flux for a variety of small animal BNCT studies. The calculated combined epithermal and fast-neutron kerma of the beam is approximately 1.0×10^{-11} cGy-cm^2, and the calculated incident gamma kerma is approximately 4.0×10^{-11} cGy-cm^2.

Keywords: BNCT, Neutron Source, Thermal, Activation

1. Introduction

Modern (post-1994) BNCT clinical trials conducted worldwide differ from earlier trials largely because of the availability of significantly improved, near-optimal, neutron sources and implementation of much more accurate computational and experimental dosimetry, including the required analytical chemistry. In contrast, there have been essentially no improvements in the boron targeting agents approved for human BNCT applications in the last 30 years. The three available approved compounds, while offering some attractive features, are still not optimal for the treatment of tumors of interest (Hawthorne and Lee, 2003).

The next significant advances in clinical BNCT efficacy are thus almost certain to be based on better alternatives to the currently-used boron targeting agents. In fact, it has been demonstrated that improved BNCT agents are essential to take full advantage of recent improvements in neutron source technology (Wheeler et al., 1999). Furthermore, it will be useful to study applications to a broader spectrum of tumor types, even with current agents (e.g. Kankaanranta et al., 2007; Dagrosa et al., 2007). And it is notable that several highly-promising new boron agents that may offer improved biochemical properties *and* that are potentially capable of treating a wider variety of tumor types are available (Hawthorne and Lee, 2003) but for various reasons they have not been systematically evaluated in small- and large-animal models to a sufficient degree to permit human trials, and some have not been evaluated at all.

In this context, the University of Missouri (MU), the Idaho National Laboratory (INL), the National Atomic Energy Commission of Argentina (CNEA), and the University of Missouri Research Reactor (MURR) are collaborating under the leadership of the MU International Institute for Nano and Molecular Medicine in a new initiative to further the development of improved BNCT agents and treatment protocols for a broader array of tumor types (Hawthorne et al., this meeting). A key first step of this effort has been the design and construction of a new thermal neutron beam irradiation facility for cell and small-animal radiobiological research at the MURR. In this paper we present the beamline design with the results of pertinent neutronic design calculations as well as initial neutronic performance measurements.

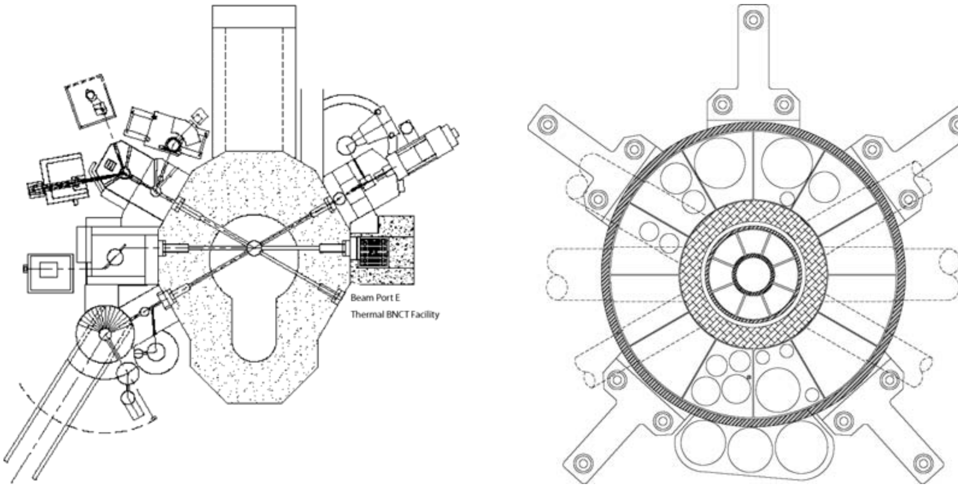

Figure 1. Top view of MURR reactor, shielding, and beamlines (L), and core detail (R) showing fuel annulus, beryllium reflector, and graphite reflector. Beamline E extends from the core horizontally to the right in this view.

2. Facility Description

The MURR reactor (Figure 1) features a compact light-water cooled and moderated fully-enriched annular core composed of eight plate-fuel elements. The maximum licensed power level is 10 MW. The outer radius of the core is approximately 14.9 cm, with an active height of 60.96 cm. The core is surrounded by a beryllium reflector, followed by a graphite reflector. Details of the new beamline design are shown in Figure 2. It will be located in an existing 15.24 cm (6") diameter MURR beam tube, referred to as Beamline E, which extends from the outer surface of the beryllium reflector, through the graphite reflector, and out through the biological shield wall as shown in the figures. Key features of the new beamline include the use of a single-crystal silicon neutron filtering section followed by a single-crystal bismuth section in a manner similar to that reported by Kim et al. (2007), but without cryogenic cooling of the crystals. The irradiation location is just downstream of the bismuth filter section, at a distance of approximately 3.95 meters from the central axis of the reactor. A shielding enclosure surrounds the exit port of the beamline as shown. A hydraulic lift inside this shield enclosure enables the remote placement of samples or animals being irradiated.

The single-crystal silicon section in the beamline provides the bulk of the spectral filtering, while the bismuth section provides some final neutron filtering along with its key function of reducing the incident gamma component in the beam. When the beam is not in use, the bismuth filter section rotates out of the beamline and is replaced by a Pb, steel, boral and polyethylene laminated shutter.

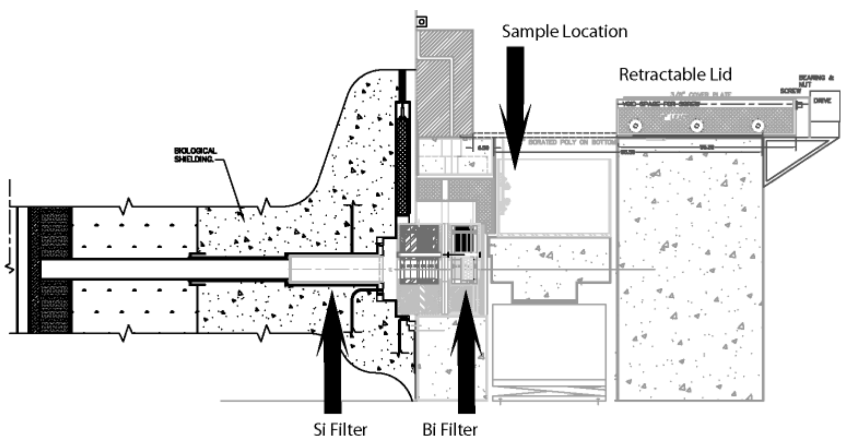

Figure 2. MURR thermal beamline design detail, shown in a closed configuration to allow access to samples.

3. Computational Methods and Models

Independent deterministic and stochastic models of the coupled reactor core and beamline were developed using the DORT (Rhoades et al., 1993) two-dimensional radiation transport code, with a highly forward-biased angular quadrature set consisting of 315 angular directions, and the MCNP-5 Monte Carlo code (Breismeister, 2000), respectively. The BUGLE-80 47-neutron, 20-gamma group cross section library (Roussin, 1980) was employed for the DORT computations, in keeping with previous practice at the INL for analysis of a number of other NCT neutron facilities worldwide. Modeling of the MURR with the two-dimensional cylindrical geometry option in DORT required a vertically-oriented model for the core, coupled at the outer boundary of the bismuth reflector to a separate, horizontal, model for the beamline. ENDF/B Version 6.8 cross section libraries were used with MCNP, except for two specialized cross section sets for the single-crystal bismuth and silicon filters in the MCNP calculations that were provided to MU and INL for this study by the Korean Atomic Energy Research Institute (Lee, 2007). These cross section sets were prepared (Kim et al., 2007) according to models described by Freund (1983). In the case of the DORT model, the thermal (Groups 46 and 47) scattering cross sections for amorphous bismuth and silicon in the BUGLE-80 library were adjusted to account for the single-crystal form of these materials using modified thermal cross sections computed by MCNP with the single-crystal libraries noted above. The adjusted cross sections were obtained by flux-volume weighting the MCNP reaction rates and fluxes in the thermal neutron energy range in the filter regions and then dividing the former by the latter to obtain effective 2-group (0 – 0.1 eV and 0.1 to 0.414 eV) thermal cross sections that reflect the reduction in thermal neutron scattering when the material is in single-crystal form. The capture cross sections were unchanged.

A number of parameter studies were conducted with DORT and, independently, with MCNP, varying the thicknesses of the silicon and bismuth filter sections to find an optimum that maximized the thermal neutron flux while maintaining the fast-neutron and gamma components of the beam within acceptable ranges. Both the DORT and the MCNP beamline optimization computations led to the conclusion that the silicon filtering section should be 50-55 cm in thickness along the beamline, while the bismuth section should be 8-10 cm in thickness.

Neutron spectra at the irradiation location, computed using the DORT discrete-ordinates model, are shown in Figure 3 for the unfiltered beamline, for the beamline with 50 cm of silicon only, and for the fully-filtered Si(50cm)/Bi(8cm) configuration. The spectral shapes computed by MCNP were consistent with the DORT results shown, within the MCNP statistical uncertainties. Modification of the spectrum to reduce the above-thermal component relative to the thermal component from one case to the next is apparent in the spectrum plots. The total calculated thermal neutron flux (0 – 0.414 eV) delivered to the irradiation location by the fully-filtered beam (Si/Bi) with the reactor at 10 MW was approximately 9.6×10^8 neutrons/cm^2-s with an estimated uncertainty of approximately 10%. The DORT calculations yielded a combined epithermal and fast-neutron kerma for the beam of approximately 1.2×10^{-11} cGy-cm^2, and an incident gamma kerma of approximately 4.0×10^{-11} cGy-cm^2.

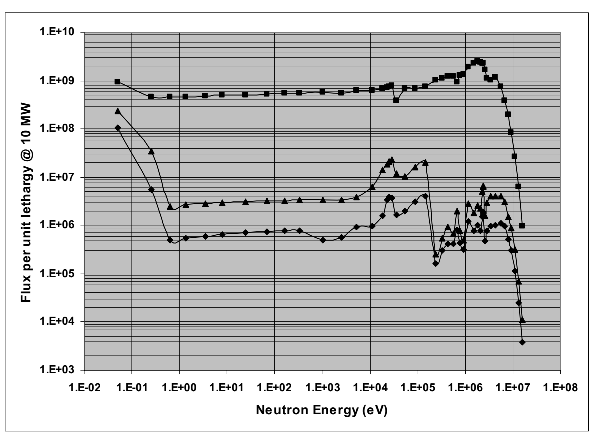

Figure 3. Computed unfiltered (■), silicon-filtered (▲), and Si+Bi filtered (♦) neutron spectra at the irradiation location for the baseline MURR thermal beamline design**.** Computations were performed with the DORT discrete-ordinates code with adjusted single-crystal silicon and bismuth cross sections.

4. Preliminary Measurements and Results

Basic beam performance measurements for an initial configuration of the beamline with 50 cm of single-crystal silicon and 8 cm of single-crystal bismuth in place, but without the rotating shutter, were focused on quantifying the thermal neutron flux intensity and the approximate spectral quality. They were conducted using gold foils with and without cadmium covers as well as with flux wires composed of natural copper alloyed with 1.55% gold by weight. The gold foils were nominally 0.0254 mm (0.001") in thickness and 12.7 mm (0.5") in diameter, with masses of approximately 60 mg. The flux wires were 1 mm in diameter and approximately 10 mm in length, each with a mass of approximately 70 mg. Some scoping measurements to estimate the gamma dose rate at the irradiation location were also performed using Landauer TLD-100 dosimeters.

Initial measurements were conducted for the following beamline filter arrangements: 1) The open, unfiltered, beamline, 2) The bismuth filter and no silicon filter, 3) The silicon filter and no bismuth and, 4) The silicon filter followed by the bismuth filter. This allowed independent evaluation of the performance of each of the two filter components separately, and in combination. The activation foils and wires and the gamma dosimeters were placed in the center of the beamline at the approximate axial location of the irradiation position in the planned final configuration. The irradiation times varied from 5 minutes in the voided beam configuration to a maximum of 10 minutes in the silicon and bismuth filtered beams. After irradiation, the absolute activities of the foils and wires were measured using a high-purity germanium detector and converted to saturation activity per atom in the usual manner.

The results of the preliminary measurements are listed in Table 1. The "measured" thermal flux values given in Table 1 were obtained by dividing the difference between the bare and cadmium-covered foil activities by a computed (MCNP) effective average thermal (0-0.414 eV) cross section for the bare foils (91.3 b ± 3%).

Table 1. Preliminary performance results for the thermal-neutron BNCT facility at MURR

	Voided Beamline	8 cm Bi Crystal	50 cm Si Crystal	50 cm Si + 8 cm Bi
Saturation Activity, Bare Gold Foil (decays/atom-s)	1.31×10^{-12} (5%)	3.82×10^{-13} (5%)	2.38×10^{-13} (5%)	8.67×10^{-14} (5%)
Saturation Activity , Cd Covered Gold Foil (decays/atom-s)	4.11×10^{-13} (5%)	7.49×10^{-14} (5%)	3.64×10^{-15} (5%)	8.21×10^{-16} (5%)
Difference in Saturation Activity, Bare-Cd (decays/atom-s)	8.95×10^{-13} (8%)	3.07×10^{-13} (5%)	2.34×10^{-13} (5%)	8.59×10^{-14} (5%)
Measured Thermal Flux (n/cm^2-s)	9.80×10^{9} (11%)	3.36×10^{9} (8%)	2.56×10^{9} (8%)	9.41×10^{8} (8%)
Calculated Thermal Flux from DORT (n/cm^2-s)	9.38×10^{9} (10%)	3.81×10^{9} (10%)	2.22×10^{9} (10%)	9.62×10^{8} (10%)
Cadmium Ratio (Bare/Cd)	3.19 (7%)	5.10 (7%)	65.4 (7%)	105.6 (7%)
Wire saturation activity ratio (Au/Cu)	36.4	28.4	22.4	22.4

Note: Reactor power is 10 MW. Uncertainties (1σ) are shown in parentheses.

The silicon filter increased the cadmium ratio from 3.1 in the voided beamline case to 65.4 indicating that it is effectively removing epithermal and fast neutrons while transmitting thermal neutrons. The measured thermal flux with both the silicon and bismuth filters in place was 9.40E+8 n/cm^2·s with a Cd ratio of 105.6, indicating a well-thermalized beam with sufficient thermal neutron flux for a variety of small animal BNCT studies.

The measured cadmium ratios for the various configurations are very consistent with expectations from the DORT and MCNP computations and show the anticipated trend toward greater thermalization of the beam as filter components are added. The ratios of gold activity to copper activity induced in the flux wires for each configuration confirm the spectral trends shown by the foil data. This ratio approaches a theoretical minimum of approximately 22 (i.e. the corresponding thermal cross section ratio at room temperature) as the beam is thermalized by the various filter combinations. The preliminary gamma dose measurements using the Landauer TLD-100 dosimeters were suspect, due to potential neutron sensitivity of these particular dosimeters as well as issues related to prompt gamma emission from some of the temporary beamstop shielding materials that were, of necessity, present during the measurements.

5. Conclusions and Future Work

Parameter studies, design calculations and initial performance measurements have been completed for a new thermal neutron beamline for neutron capture therapy cell and small-animal radiobiology studies at the University of Missouri Research Reactor. Once the rotating beam shutter and the permanent beamstop shielding components have been installed, a much more comprehensive set of activation measurements will be conducted to characterize the neutronic performance of the final system. The incident gamma component of the MURR neutron source will also be measured using a set of FarWest™ paired ion chambers, in keeping with the recommendations of the International BNCT Dosimetry Exchange (Järvinen and Voorbrack, 2003).

6. References

Breismeister, J.F., 1999. "MCNP – A General Monte Carlo N-Particle Transport Code, Version 4C, LA-13709-M, Los Alamos National Laboratory, USA .

Dagrosa, M.A., et al., 2007. Optimization of Boron Neutron Capture Therapy for the Treatment of Undifferentiated Thyroid Cancer, International Journal of Radiation Oncology, Biology, and Physics, **69**:1059-1066.

Freund, A.K., 1983. Cross Sections of Materials used as Neutron Monochromators and Filters, Nuclear Instruments and Methods, **243:**495-501.

Hawthorne M.F. and Lee, M.W., 2003. A Critical Assessment of Boron Target Compounds for Boron Neutron Capture Therapy, Journal of Neuro Oncology **62**:33-45.

Järvinen, H., and Voorbraak, W.P., 2003. Recommendations for the Dosimetry of Boron Neutron Capture Therapy, Report 21425/03 55339/C, NRG Petten, The Netherlands.

Kankaanranta, L., et al.,2007. Boron Neutron Capture Therapy in the Treatment of Locally Reccurent Head and Neck Cancer, International Journal of Radiation Oncology, Biology, and Physics, **69**:475-482.

Kim, Myong-Seop et al., 2007. Development and characteristics of the HANARO neutron irradiation facility for applications in the boron neutron capture therapy field, Phys. Med. Biol. **52** 2553-2566

Lee, Byung-Chul, 2007. Korea Atomic Energy Research Institute, Private communication.

Rhoades, WA, et. al., 1993. TORT-DORT: Two and Three-Dimensional Discrete-Ordinates Transport, Radiation Shielding Information Center, Oak Ridge National Laboratory, CCC-543.

Roussin, R.W., 1980. BUGLE-80 Coupled 47-Neutron, 20 Gamma-Ray P3 Cross Section Library, DLC-75, Radiation Shielding Information Center, Oak Ridge National Laboratory, USA.

Wheeler, F.J, Nigg, D.W., et al., 1999. Boron Neutron Capture Therapy: Implications of Neutron Beam and Boron Compound Characteristics, Med. Phys. **26**:1237-1244.

This work was supported by the University of Missouri Institute for Nano and Molecular Medicine. Some in-kind material contributions were made by the US Department of Energy through the Idaho National Laboratory.