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Los Alamos Neutron Science Center Contributions to the Development of Future Nuclear Power Reactors

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Abstract – *The Los Alamos Neutron Science Center (LANSCE) is a large spallation neutron complex centered around an 800 MeV high-current proton accelerator. Existing facilities include a highly-moderated neutron facility (Lujan Center) where neutrons between thermal and keV energies are produced, and the Weapons Neutron Research Center (WNR), where a bare spallation target produces neutrons between 0.1 and several hundred MeV.*

The LANSCE facility offers a unique capability to provide high precision nuclear data over a large energy region, including that for fast reactor systems. In an ongoing experimental program the fission and capture cross sections are being measured for a number of minor actinides relevant for Generation-IV reactors and transmutation technology.

Fission experiments makes use of both the highly moderated spallation neutron spectrum at the Lujan Center, and the unmoderated high energy spectrum at WNR. By combining measurements at these two facilities the differential fission cross section is measured relative to the $^{235}U(n,f)$ standard from sub-thermal energies up to about 200 MeV. An elaborate data acquisition system is designed to deal with all the different types of background present when spanning 10 energy decades. The first isotope to be measured was ^{237}Np, and the results were used to improve the current ENDF/B-VII evaluation. Partial results have also been obtained for ^{240}Pu and ^{242}Pu, and the final results are expected shortly.

Capture cross sections are measured at LANSCE using the Detector for Advanced Neutron Capture Experiments (DANCE). This unique instrument is highly efficient in detecting radiative capture events, and can thus handle radioactive samples of half-lives as low as 100 years. A number of capture cross sections important to fast reaction applications have been measured with DANCE. The first measurement was on $^{237}Np(n,\gamma)$, and the results have been submitted for publication. Other capture measurements in progress include ^{240}Pu and ^{242}Pu.

The United States recently announced the Global Nuclear Energy Partnership (GNEP), with the goal of closing the commercial nuclear fuel cycle while minimizing proliferation risk. GNEP achieves these goals using fast-spectrum nuclear reactors powered by new transmutation fuels that contain significant quantities of minor actinides. The proposed Materials Test Station (MTS) will provide the GNEP with a cost-effective means of obtaining domestic fast-spectrum irradiations of advanced transmutation fuel forms and structural materials, which is an important step in the fuels qualification process. The MTS will be located at the LANSCE, and will be driven by a 1.08-MW proton beam. The peak neutron flux in the irradiation region is $1.67x10^{15}$ n/cm^2/s, and the energy spectrum is similar to that of a fast reactor, with the addition of a high-energy tail. The facility is expected to operate at least 4,400 hours per year. Fuel burnup rates will exceed 4% per year, and the radiation damage rate in iron will be 18 dpa (displacements per atom) per year. The construction cost is estimated to be \$73M (including 25% contingency), with annual operating costs in the range of \$6M to \$10M. Appropriately funded, the MTS could begin operation in 2010.

I. Introduction

The Los Alamos Neutron Science Center (LANSCE, then known as LAMPF – Los Alamos Meson Physics Facility) completed its construction the early 1970s, providing its first beam in 1972. Its goal was to provide unique opportunities in fundamental research of medium-energy nuclear physics, by utilizing its 800 MeV proton beam to produce secondary particles such as pions. Considering the need to provide an intense source of neutrons, the accelerator was designed and constructed to be capable of accelerating both positive and negative hydrogen ions and of delivering those beams to multiple experimental areas simultaneously.

The current LANSCE layout is shown in Fig. 1. It was designated as a National User Facility in 2001, and its research program[1] has expanded substantially. In the 2003–2004

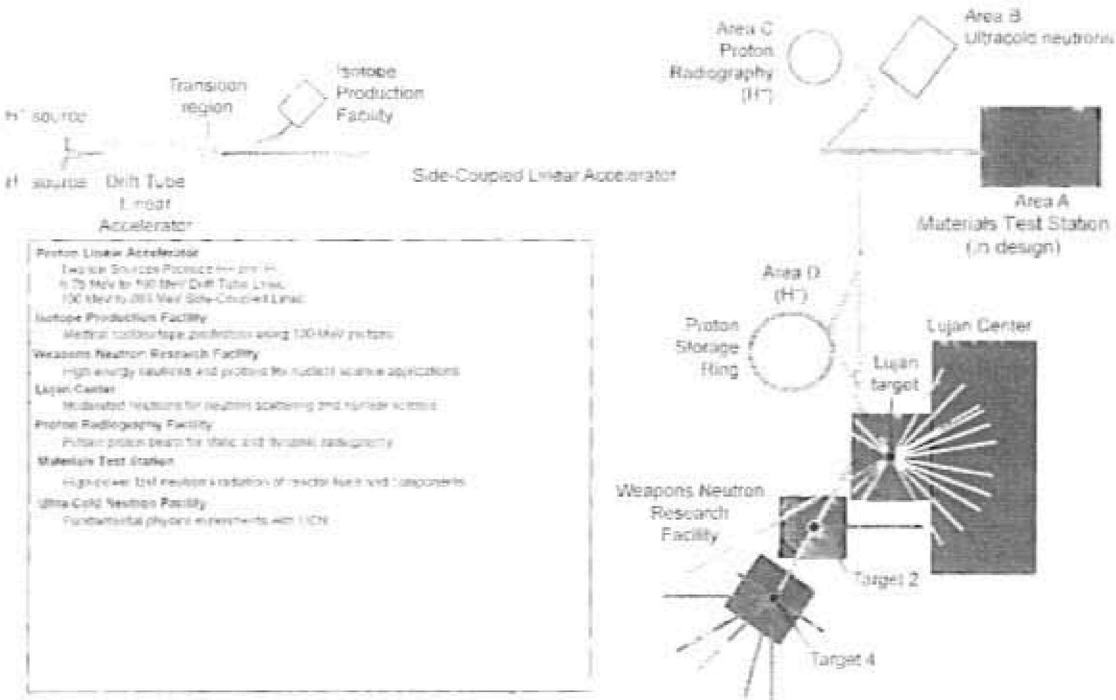

Fig. 1. Layout of the LANSCE user facility

operating period, there were over 1100 user visits during which over 350 experiments were conducted during 200 days of user facility operation. The core capability of the facility is an 800-MeV linear accelerator system that accelerates both negative hydrogen ions and protons. At present the accelerator supplies up to 135 μA of H- ions with pulsed beam timing patterns suitable for a wide variety of experimental programs at three user facility areas, and up to 250 μA of proton beam (H+) at 100 MeV for isotope production.

II. The current LANSCE facility

There are three major experimental areas: the Manuel Lujan Jr. Neutron Scattering Center (Lujan Center), the Weapons Neutron Research (WNR) Facility and the Proton Radiography Facility (pRad); these make up the user facility. In addition, there is an area for 100 MeV isotope production and an area for Ultra Cold Neutron production for fundamental physics.

The Lujan Center uses spallation neutrons produced by the 800-MeV H- beam, through moderators, to produce cold, thermal and epithermal neutrons in the meV to keV range, primarily for neutron scattering studies and nuclear science research. The H⁻ beam is injected into the Proton Storage Ring (PSR) that compresses the 750 ms-long pulses from the linear accelerator, to 125 ns wide pulses (FWHM) at a rate of 20 Hz. The proton beam, at a current of up to 135 μA, then strikes a light-water cooled split-tungsten target with a flux-trap and backscattering moderator system to give a high peak neutron flux. The neutrons are collimated to form beams for up to seventeen flight paths.

Research at the Lujan Center is supported by instrumentation on 12 flight paths that allows neutron scattering measurements for materials science and engineering, polymer science, chemistry, earth science and geology, structural biology, and condensed matter physics. In addition, there are three flight paths instrumented for neutron nuclear science and transmission spectroscopy.

The WNR Facility is primarily used for basic and applied nuclear science and for testing of semiconductor devices in neutron beams for industry. For most experiments, WNR uses up to 5 μA of H⁻ beam that is chopped and bunched before acceleration to give a pulse-to-pulse separation of typically 1.8 μs (the pulse separation can be adjusted based on the requirements of experiments). The pulse width is approximately 125 ps (FWHM). Neutron energy is determined at using flight paths between 10 and 90 meters long, using time-of-flight measurements. These pulses are selected out of the individual micropulses that form the 750-μs "macro-pulse".

WNR has two experimental areas that use the proton beam, "Target 2" and "Target 4".

Target 2 is a low-background area with seven flight paths that can be configured to support a wide array of experimental instrumentation. A Lead Slowing Down Spectrometer[2] is currently utilized in fission cross-section measurements of actinide samples as small as 10 ng. At the WNR "Target 4", the beam strikes a tungsten target producing neutrons with useful energies from about 100 keV to above 600 MeV. Those neutrons are collimated to form beams for six flight paths viewing the neutron source at angles to the left (L) or right (R) relative to the incident beam direction of 15,30,60and 90 degrees. Research at the WNR is supported by a suite of flight-path instrumentation that allows measurements providing nuclear data for radiochemistry and astrophysics, cross-sections for neutron-induced reactions on actinide isotopes, fission cross-sections, energy production, fragment mass and neutron energy distributions, properties of nuclei in excited states ("isomers"), few-body interactions such as n–D scattering, reaction studies for theory and modeling, and electronics component and system performance in radiation environments.

The Proton Radiography Facility (pRad) provides a unique capability for study of dynamic processes using the proton beam and a magnetic lens imaging system. Because protons interact through both the strong nuclear force and the electromagnetic force, transmission measurements allow simultaneous imaging and determination of material properties with very high time resolution[3] . For pRad, 50 ns-wide H^- beam pulses with approximately 10^9 particles are spaced in time at intervals pre-determined by experiment requirements. The beam is then timed to strike test objects during the dynamic event. Transmitted and scattered protons are imaged by a magnetic lens system and recorded by cameras. This technique gives multi-frame radiographs that spatially resolve features with better than 1-mm accuracy using samples of up to 60 gm/cm^2 thick.

Over the past 20 years, LANSCE has supported the Los Alamos Radioisotope Program by irradiating target materials, originally at 800 MeV and now at 100 MeV. The Isotope Production Facility (IPF) project was completed in the fall of 2003 and is now fully operational. The facility has a new beam line and the equipment required to deliver 100 MeV proton beam from the existing LANSCE accelerator to a new target station designed specifically for the production of radioisotopes. The IPF was designed to operate without influencing scheduled beam delivery of proton beam to other experimental areas at LANSCE. In the IPF, production targets are segmented into different production regions and materials to allow optimized production of various radioisotopes. The new facility thereby produces a wide range of radioisotopes for medical diagnosis, treatment, and scientific research.

III. Plans for sustaining the user facility

The LANSCE accelerator provided the first 800 MeV proton beam in June of 1972. Since that time, its mission has evolved, but improvements to the physical infrastructure have not kept pace with evolving requirements for operation at an acceptable level of reliability. Over the past several years, work has been done to define the elements of a project necessary to sustain reliable operation for the next several decades. Specifically, a LANSCE Refurbishment (LANSCE-R) Project is planned that will improve facility operations. The goals are to replace or refurbish systems that (1) have an impact of 15% or greater on reliability, (2) to increase operational efficiency by eliminating sources of single-point failure that have long estimated times to repair, and (3) replace equipment that is beyond its predicted end-of-life and difficult to maintain.

Achieving this requires replacing radio-frequency equipment to achieve high reliability and providing adequate spare components; replacing hardware and software in the accelerator controls, data acquisition, and timing systems that have become virtually non-maintainable; refurbishing and replacing vacuum, cooling, and magnet power supplies for the accelerator and beam-transfer lines; and refurbishing and improving the beam-diagnostics systems. Along with replacement of injector components we plan to increase the H⁻ beam intensity by a factor of two.

LANSCE-R will cost roughly $200M and occur over a period of seven years, beginning in 2008. During this time LANSCE facility will continue user operations by installing equipment during scheduled outages. When completed, the investment at LANSCE will have been preserved and enhanced for the next decade and beyond.

IV. Fission Cross-Section Measurements

The fission cross section measurements at LANSCE are carried out at two different experimental locations in order to span 10 decades of incident neutron energy. By combining measurements at the Lujan Center and the WNR facility it is possible to obtain fission cross sections from 0.01 eV to 200 MeV, with an overlap in the 100-200 keV region.

The fission events are detected using gas-filled parallel-plate ionization chambers (PPIC's). These chambers are routinely used as flux monitors at different flight paths of WNR, and are described in detail in Ref[4]. The PPIC's are reliable, easy to operate, resistant to radiation damage and highly efficient for fission detection. Another important feature is the ability to discriminate between α-particles (from natural decay and (n,α)-reactions) and fission fragments. A high level of separation between the two particle types can be achieved by optimizing the plate spacing and gas pressure in the chamber.

The cross sections are measured relative to ^{235}U, which is a standard at 0.0253 eV and from 150 keV to 200 MeV[5]. The time-of-flight method (TOF) is used to determine the incident neutron energy. Each channel on the PPIC's has an identical and nearly independent data acquisition system in order to avoid dead-time correlation issues. Each data acquisition system is triggered by a signal from the PPIC channel above a set threshold. The raw PPIC signals are driven to the data acquisition system (DAQ) by fast current-sensitive preamplifiers mounted on the chamber. A beam pick-off for the accelerator provides the start signal for the TOF measurement. The relative arrival time of the PPIC pulse is recorded and used to determine the neutron time-of-flight. The PPIC pulse height is also recorded for each event and used to discriminate between fission fragments and other charged particles.

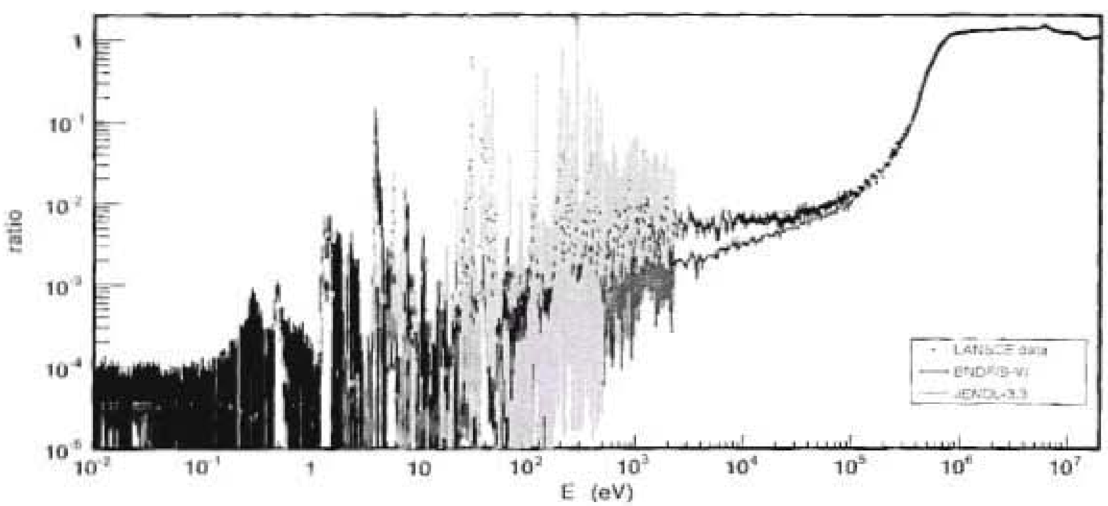

Fig. 2. The measured fission cross section of ^{237}Np from compared to the ENDF/B-VI and JENDL-3.3 evaluations.

Since the measurements cover a large energy region there are many types of corrections to consider in the analysis. There has been much effort in reducing the systematic uncertainties introduced by the different corrections in order to approach a 1% level of uncertainty. Improvements in the data acquisition system (DAQ) and analysis procedure have been made to achieve this level of precision. One such improvement was a modification to the DAQ in order to measure a type of background called wrap-around events. These events are caused by slow neutrons from one proton pulse overlapping with fast neutrons from a later pulse. By using an innovative

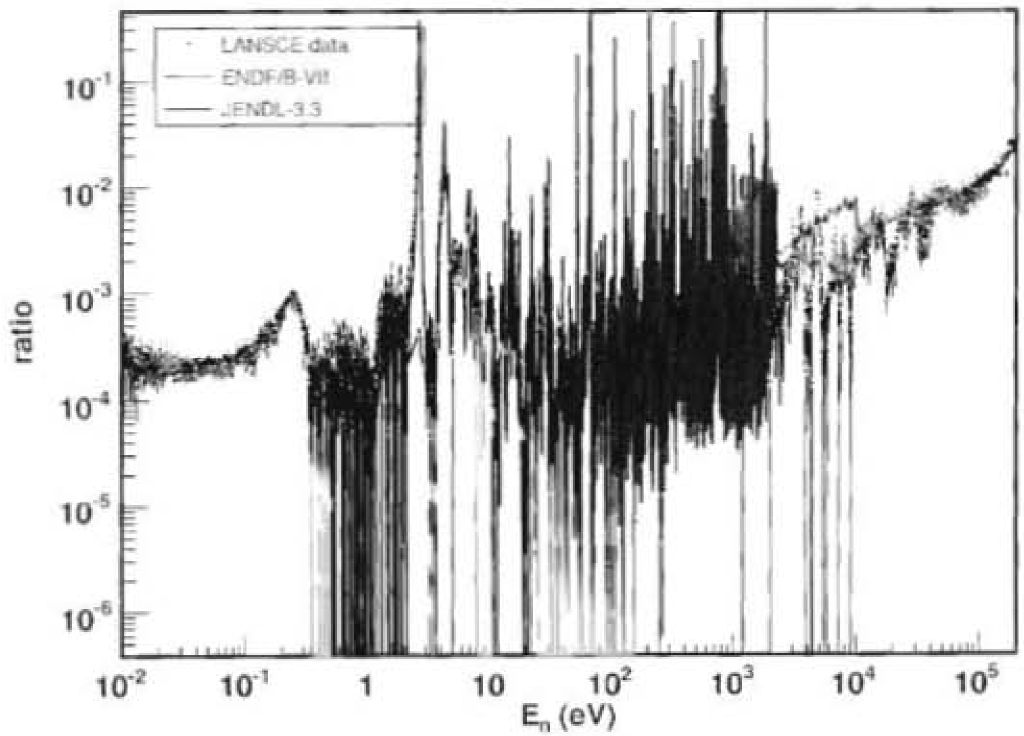

Fig. 3. The measured cross section ^{242}Pu as a ratio to ^{235}U. Please not that the data needs to be corrected for fission events from the 0.01% contamination level of ^{241}Pu. The evaluations have been modified to include the contamination.

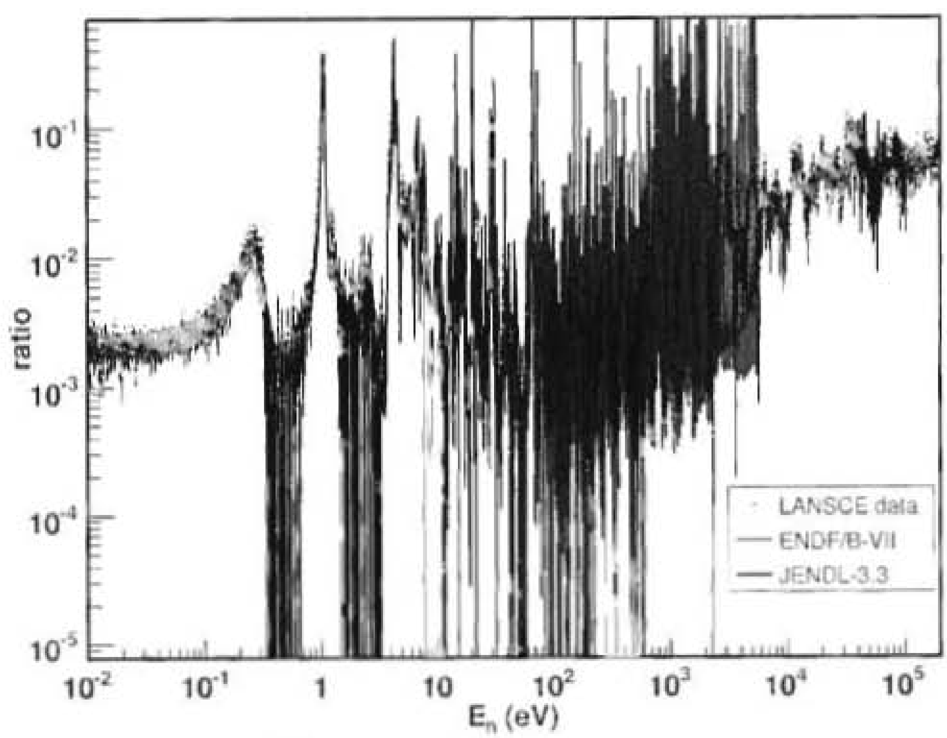

Fig. 4. The measured cross section ^{240}Pu as a ratio to ^{235}U. Please not that the data needs to be corrected for fission events from the 0.01% contamination level of ^{241}Pu. The evaluations have been modified to include the contamination.

modification to the DAQ it is possible to accurately measure and fit this background. shown in Fig. 2, together with the ENDF/B-VI[6] and JENDL-3.3[7] evaluations. The measurement clearly favors the JENDL-3.3 evaluation in the unresolved resonance region. However, above fission threshold the data agrees better with the ENDF/B-VI evaluation. The measurement data have been submitted to the ENDF/B-VII project, and a new evaluation which is in better agreement with the measurement will appear in the official release.

The preliminary results obtained for ^{242}Pu and ^{240}Pu are shown in Figs. 3 and 4, respectively. The data still needs to be corrected for a 0.01% contamination of ^{241}Pu in the samples. The ^{241}Pu(n,f) cross section was recently measured and the result will be used for the corrections. The correction will strongly affect the result in thermal region, but will only change the result in the unresolved resonance region with a few percent. The ENDF evaluation of the ^{242}Pu(n,f) cross section is a typical example of the problem observed for many non-fissile minor actinides. The evaluation exhibits an un-physical "step" in the evaluated fission cross section at 10 keV, probably caused by inconsistencies between measurements performed in the sub-threshold and above-threshold regions. Since the experimental data set reported here goes right through the 10 keV region it clearly resolves this inconsistency.

V. Materials Test Station

In January 2006, the US Department of Energy announced plans for its Global Nuclear Energy Partnership, which seeks to reduce the risks of nuclear proliferation while simultaneously expanding the use of nuclear power worldwide. Central to achieving these goals is the development of a closed nuclear fuel cycle where spent fuel from nuclear reactors is recycled, resulting in more effective utilization of the world's limited uranium resources and a significant reduction in the volume of high-level radioactive waste.

Recycling spent fuel involves transmuting minor actinides (Np, Am and Cm) in fast-spectrum nuclear reactors. While such reactors have existed for over half a century, they have not traditionally been designed to transmute minor actinides (MA's). This function requires the development of new fuel forms containing significant quantities of minor actinides that can survive high fast-spectrum neutron fluence ($>10^{23}$ n/cm^2). Qualification of new fuel forms will require testing in prototypic spectra. Unfortunately, only a few fast-spectrum irradiation facilities exist around the world. There are no fast reactors currently operating in the USA, and the earliest a new reactor could reasonably be expected to begin operation would be towards the end of the next decade.

Recognizing the need to establish a domestic fast-spectrum irradiation capability, in 2005 the US Congress appropriated funds to design the Materials Test Station (MTS) at the Los Alamos Neutron Science Center (LANSCE) within Los Alamos National Laboratory (LANL). The primary purpose of this facility will be to irradiate candidate MA-bearing fuels and cladding being developed under the Global Nuclear Energy Partnership. The MTS will differ from all other fast-spectrum irradiation facilities in that it is not a nuclear reactor. Rather, the primary source of neutrons is spallation reactions induced by directing 800-MeV protons onto a tungsten target.

For nearly three decades, the LANSCE accelerator reliably delivered 1 mA of 800-MeV protons to pion production targets in support fundamental nuclear physics experiments. While the LANSCE accelerator has continued to deliver 800-MeV H$^-$ beam to other targets, the 1-mA proton beam has not been utilized since 2000. The DOE is planning to refurbish the LANSCE accelerator so that it can continue to reliably deliver beam for another two decades or more. This refurbishment consists primarily of replacing antiquated power supplies that deliver radiofrequency power to the accelerator structure. Once refurbished, the LANSCE accelerator will be capable of delivering 1.35 mA of 800-MeV protons, or 1.08 MW of proton beam, to the MTS.

The basic configuration of the MTS target system, depicted in Figure 5, consists of two spallation target regions separated by a fuel irradiation region. The spallation targets are tall, narrow blocks of tungsten cooled by liquid lead-bismuth eutectic (PbBi). The fuel irradiation region is 2 cm wide and can hold up to 40 fuel pins, each with an active fuel pellet stack height of 12 cm. The pulsed nature of the proton beam allows alternate beam pulses to be directed onto first one, then the other, spallation target in a straightforward and direct manner.

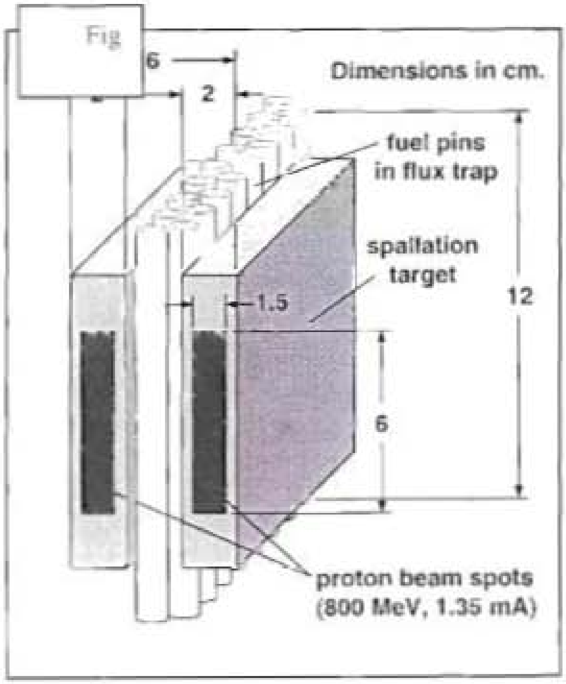

Figure 5. Basic configuration of the MTS target assembly.

The reason for using two spallation target sections and alternating the beam pulses between them is to achieve nearly uniform time-averaged neutron flux in the fuel irradiation region, which is located between the two spallation targets. ure 6 shows the spatial distribution of the neutron flux from a horizontal cut at target mid-plane. It shows the flux gradient across the 2-cm gap between spallation targets to be quite low. The peak flux occurs directly in the spallation targets where most of the neutrons are created. The peak flux in the irradiation region is 1.67×10^{15} $n.cm^{-2}.s^{-1}$.

The neutron spectrum at the peak flux position in the irradiation region is compared to the spectrum of a fast reactor in Figure 7. The fast reactor chosen for comparison is the proposed Advanced Burner Test Rector (ABTR), which is currently undergoing pre-conceptual design. The MTS spectrum is quite similar to that of a fast reactor, with the addition of a high-energy tail above 10 MeV that reaches up to the incident 800-MeV beam energy. The only significant impact of this high-energy tail is high helium production in steels. The peak flux position has about 60 times greater helium production rate in iron than that in a fast reactor at an equivalent flux level. For the ferritic/martensitic steels that are currently under consideration as fuel clad material for fast reactors, high concentrations (>700 appm) of helium can lead to embrittlement. The impact of higher-than-prototypic fast reactor helium production in the MTS must be given careful consideration when irradiations are performed.

The spallation targets consist of tall (18 cm), narrow (2.1 cm) tungsten plates stacked along the direction of proton beam propagation. The plates start out thin (4.4 mm) at the front end of the target, and grow progressively thicker toward the back. Separating the plates are 1-mm-thick cooling channels. The plate thicknesses are adjusted in this manner so that the heat flux at the cooling surface is limited to 600 W/cm^2, which does not challenge the thermal-hydraulic capability of the PbBi coolant.

A critical parameter for obtaining meaningful irradiation data is the temperature at which the irradiations are performed. In order to achieve conditions similar to that of a fast reactor, the fuel clad temperature must be controlled within a specified tolerance, and irradiation temperatures up to 550 °C must be attainable in the MTS. This requirement is very difficult to achieve using water as the fuel pin coolant. After considering a number of coolant options, PbBi has been selected as the coolant for the test fuel pins. This coolant does not react exothermically with water or air, has a high heat transfer coefficient, and is liquid over a large temperature range. It has the disadvantages of not being a liquid at room temperature (requiring trace heating on loop piping and components), requiring active oxygen control to reduce corrosion, and producing ^{210}Po as an activation product. Polonium-210 is an alpha emitter with a 138-d half-life, and limiting its release during off-normal events will require special attention to the design of safety systems.

The high heat transfer coefficient of PbBi means the film drop is relatively low (~30 °C) and predictable within 30%. Thus by measuring the inlet and outlet temperatures of the PbBi, the fuel clad temperature at any point along the fuel pin height should be known to within 10 °C. The fuel clad temperature can be controlled by adjusting the PbBi inlet temperature.

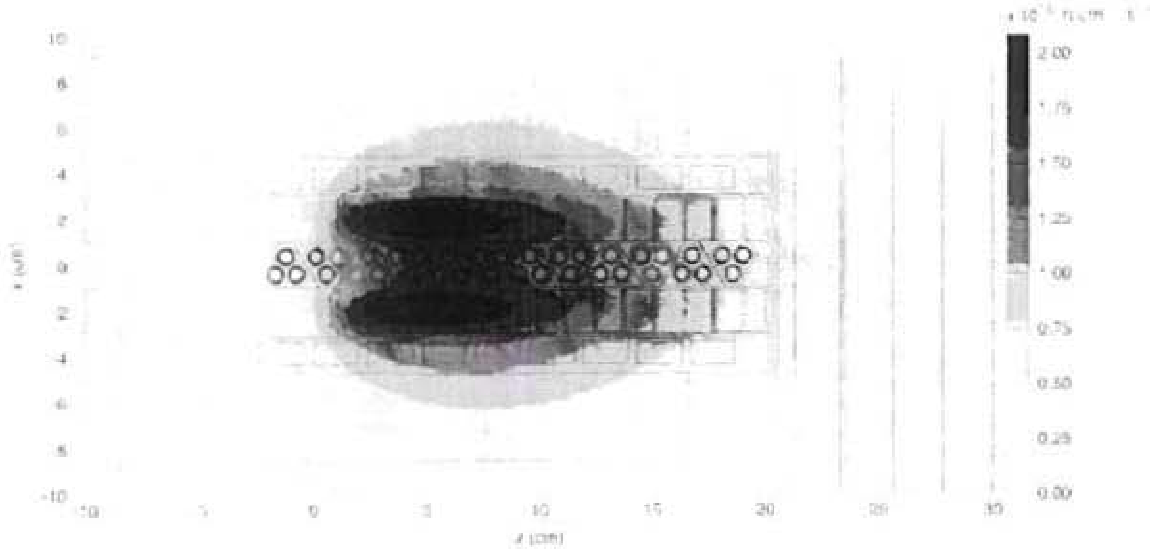

Figure 6. Spatial distribution of the neutron flux at target mid-plane.

Figure 7. Neutron energy spectrum at the peak flux position in the irradiation zone of the MTS compared to that of a fast reactor (ABTR).

The beam spot on each spallation target is 1.5 cm wide by 6 cm tall. A nearly uniform current density over the beam spot is achieved by rastering a small beamlet (nominally 3 mm wide by 6 mm high, FWHM) over the beam spot dimensions. This results in a peak current density on target of 75 μA/cm^2. Over a calendar year, the MTS is expected to operate 4400 hours at a beam current of 1.35 mA. The annual radiation damage in T91 alloy, a ferritic/martensitic steel that is the leading candidate material for the fuel pin housing and spallation target housing, at this current density is calculated to be 31 displacements per atom (dpa). For reference, a tungsten target at the ISIS facility at the Rutherford Appleton Laboratory in the United Kingdom was replaced at a dose of 13 dpa due to the failure of thermocouples used to monitor plate temperatures. At the time of replacement, the target was performing satisfactorily. Thus the anticipated minimum life of the target is expected to be 6 months, but the expectation is that it will not require more than annual replacement.

Located laterally outboard of the spallation targets are additional irradiation positions where materials samples may be placed. The neutron flux gradient in the lateral direction is rather high region, about 4% per mm, yet this is acceptable if the samples are sufficiently thin in one dimension (no more than 250 μm), as is typically the case with modern test specimens. The peak flux in this region is about 20% less than in the flux trap.

The highly activated target assembly will be serviced in a hot cell located downstream of the target operating position. The target assembly is being designed to be remotely serviceable. Spent activated components will be loaded into transfer casks and transported to appropriate facilities for disposal. Reasonably rapid retrieval of irradiated fuel pins is accomplished through the use of a gasketed cover plate on top of the fuel module. Once removed from the fuel module and loaded into transfer casks, the irradiated pins can be shipped to hot cells at LANL or other laboratories for post-irradiation examination.

Use of the existing LANSCE accelerator and the Area A experimental hall results in significant cost savings over a green field site for the MTS. A bottoms-up cost estimate completed within the last year yielded a total project cost for the MTS of $73M, including 25% contingency. This estimate includes the cost of preparing Area A for siting the MTS, which is approximately $13M. Other major cost components are the accelerator beam transport ($12M), hot cell and external shielding ($11M), target assembly services ($8M), and target assembly fabrication ($7M). Annual operating costs for the MTS are estimated to be between $6M and $10M.

The MTS project schedule is subject to timely approvals of DOE critical decisions. If this process proceeds smoothly, construction could commence in FY08, with commissioning taking place in FY11.

The MTS will provide a critical domestic irradiation capability to the DOE, allowing fast-spectrum irradiations of new MA-bearing fuels to commence well before a new fast reactor can be expected to start operation in the USA. The neutron spectrum in the fuel irradiation region is similar to that of a fast reactor, and fast flux levels in the irradiation region are about one-third that of the peak fast flux in the Phénix reactor, which currently produces the most intense fast flux in the world.

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