NUCLEAR DATA MEASUREMENTS WITH THE OAK RIDGE ELECTRON LINEAR ACCELERATOR (ORELA) FOR SUPPORTING NUCLEAR FUEL CYCLE APPLICATIONS

M. E. Dunn and K. H. Guber
Oak Ridge National Laboratory, P. O. Box 2008, Oak Ridge, TN 37831-6170 USA; dunnme@ornl.gov

For more than 30 years, the Oak Ridge National Laboratory (ORNL) has performed research and development to provide accurate nuclear cross-section data in the resonance region. Overall, the Nuclear Data (ND) Program is tightly coupled with nuclear fuel cycle analyses and radiation transport methods development efforts at ORNL. As a result, resonance region measurements and evaluations are performed in concert with nuclear science and technology needs and requirements. At the heart of the ND program is the Oak Ridge Electron Linear Accelerator (ORELA) that is used to perform high-resolution neutron cross-section measurements in the resonance region. Recently, an accelerator refurbishment effort has been initiated at ORELA to improve operation reliability. As a result, new cross-section measurements have been performed at ORELA, and ORELA is readily available to address current and future nuclear data needs in the resonance region.

I. INTRODUCTION

The Oak Ridge National Laboratory (ORNL) Nuclear Data (ND) Program is tightly coupled with nuclear methods and analysis work to support fuel cycle applications. Figure 1 provides a flow diagram showing how ND work activities support radiation transport modeling and simulation work at ORNL. As shown in Fig. 1, the ORNL ND Program consists of four complementary areas of research: (1) cross-section measurements at ORELA; (2) resonance analysis methods development with the SAMMY R-matrix analysis software; (3) cross-section evaluation development for dissemination by the Evaluated Nuclear Data File (ENDF/B) System that is maintained by the National Nuclear Data Center at Brookhaven National Laboratory; and (4) cross-section processing methods development with the AMPX software system. Moreover, the cross-section data with covariance data are provided to support computational modeling development and nuclear application analyses. As a result, nuclear fuel cycle analyses and radiation transport methods development efforts are directly supported by resonance region measurement and evaluation efforts, thereby ensuring nuclear science and technology needs and requirements are addressed by the basic nuclear data. Since the mid-1990s, many of the resonance region advances have been driven by needs within the Nuclear Criticality Safety Program (NCSP) of the U.S. National Nuclear Security Administration (NNSA). For example, assessments of previous nuclear data measurements and evaluations have revealed deficiencies in nuclear data (e.g., missing resonances, high neutron sensitivity, etc.) that are important for criticality safety applications.[1] As a result, new measurements and evaluations have been performed to address the nuclear data deficiencies. Recent advances in each component of the ORNL ND Program have led to improvements in resonance region measurements, R-matrix analyses, cross-section evaluations, and processing capabilities that directly support radiation transport research and development. Details concerning all of these research components are beyond the scope of the paper. The objective of the paper is to describe the ORNL cross-section measurement capabilities and show how ORELA provides basic nuclear data that support the three additional complementary areas of the ND program and nuclear fuel cycle analyses.

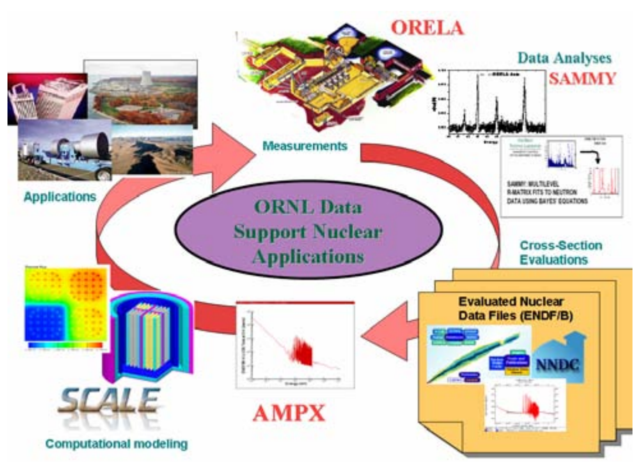

Fig. 1. ORNL Nuclear Data Program.

II. FACILITY OVERVIEW

ORELA is equipped with a pulsed, gridded, electron gun (pulse width 2–30 ns, repetition rate 1–1000 Hz), a four-section radio frequency (RF) LINAC, and a water-cooled tantalum target with beryllium housing to generate short neutron pulses via (γ,n) reactions. The RF power for the accelerator sections is provided by four 24-MW, 1300-MHz klystrons that can accelerate electrons up to 180 MeV and yield a neutron production rate of 1014 n/s at 50-kW beam power.

ORELA has 10 flight paths ranging in length from 9 to 200 m with 18 underground flight stations. The combination of long flight paths and short neutron pulses leads to an excellent neutron energy determination using the time of flight (TOF) method. The water-cooled tantalum target is 15 cm in diameter and generates high-intensity pulses of neutrons with a comparatively small moderation time compared to other neutron facilities. Because of the relatively high flux and brightness of the source, a small sample size can be used for measuring neutron-induced cross sections. A layout of the ORELA facility is shown in Fig. 2. Over the past 30 years, ORELA measurements have contributed to ~80% of U.S. ENDF/B evaluations.[2]

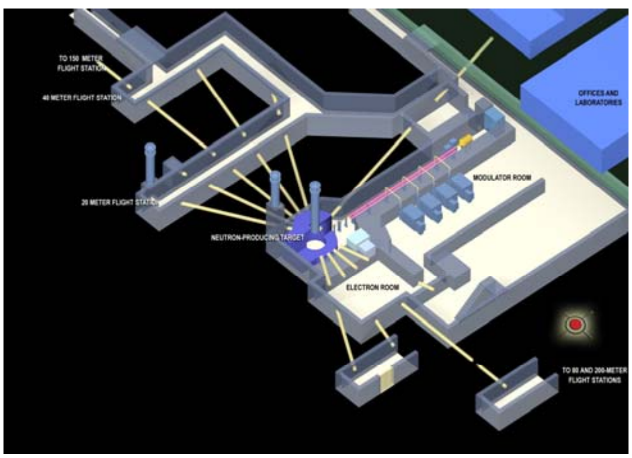

Fig. 2. Layout of the ORELA facility.

The ORELA accelerator,[3] modulator, and control systems were designed and built by Varian in 1968 (Varian model V7727). The accelerator consists of four sections with an active length of 16.5 m, which leads to an average accelerating gradient of ~10 MeV/m. Each section is a coupled cavity TM01 transmission line operating in the 2/3 π mode and is terminated with an internal RF load. Twelve water-cooling lines are distributed evenly around each section to maintain a constant operating temperature. A WR650 waveguide SF_6 gas-cooled window is used to feed up to 24 MW of microwave pulses into each section. Each accelerator section is equipped with two 60-L/s Vac-ion pumps. In total, 10 Vac-ion pumps are used to maintain a high vacuum. The electron gun has incorporated a specially designed 35-L/s Vac-ion pump.

III. FACILITY STATUS

Between December 2001 and April 2006, ORELA experienced operational difficulties resulting in unplanned outages. The main operational problems were directly attributed to electron gun malfunctions and the inability to maintain adequate vacuum conditions in the accelerator. In 2006, the NNSA NCSP provided refurbishment support to address the immediate problems and re-establish reliable operation. Since April 2006, ORELA has operated reliably for ~1000 h, and the following discussion provides additional details concerning key accelerator components and the status of recent refurbishment efforts.

III.A. Accelerator Vacuum System

Over the years, a number of vacuum issues have been a problem, including small leaks from the water cooling lines into the accelerator high vacuum. Unfortunately the leaks are in locations that are difficult to access and detect with a leak checker but could be roughly localized by observing relative responses on the various Vac-ion pump currents. Also, a residual gas analyzer (RGA) was installed for better detection and identification of the leaks. For many years the leaks were controlled with a "stop leak" mixture circulated in the cooling water. The leak sealing material does eventually harden and block smaller water passages. In the last decade, this method of controlling leaks became less and less effective until the vacuum became too poor to effectively run the accelerator. By isolating the individual accelerator sections, two braze joints in the main water inlet of Sect. 4 and cracks in threads of tapped water fittings on many of the outlets were found to be the major leak points.

During the original manufacturing process, the tapered water fitting threads were overcut, leading to small cracks between water-cooling and high vacuum systems. Eventually, the additives to the cooling water failed to work reliably, so a differentially pumped guard vacuum system was designed and installed on all outlet fittings of the cooling lines. This was achieved by installing O-ring sealed inserts that isolate the cracked threads from water. During the refurbishment period, this system was overhauled and improved by replacing the aging O-rings and installing new pumps with automatic valves to prevent oil-backstreaming. Because the leaks for two fittings were rather large, individual pumps were used to maintain vacuum on those lines.

At the water inlets of accelerator Sect. 4, leaks were detected at a stainless steel to copper braze joint in the accelerator body. Several attempts to seal this leak with epoxy and other sealants were unsuccessful. Therefore, a mechanical seal with a compressed rubber gasket across the braze joint was designed and installed; and this seal has maintained reliable vacuum for more than a year.

All ten Vac-ion pumps were replaced, because they were at the end of their expected lifetime and had been exposed to a large gas load as a result of the recent vacuum problems. Also, the high-voltage cables and controls for the Vac-ion pumps were debugged and refurbished in the past year. In addition, a new 300-L/s turbo pump was purchased to facilitate startup of the Vac-ion pumps. The turbo pump also was very helpful to maintain a good vacuum for leak checking and operating the RGA.

III.B. Klystrons

Four high-power klystron tubes and associated modulators provide the microwave power pulses to drive the accelerator. The tubes used at ORELA are Litton model L-5081 1.3-GHz klystrons that produce up to 24-MW 2–μs pulses with up to -250 kV, 250 A of beam power. A pulse repetition rate of up to 1000 Hz can be used; however, 525 Hz is typical for most experiments. Each klystron has a modulator unit that consists of an adjustable 0- to 25–kV high-voltage direct current (HVdc) power supply and a thyratron-driven pulse-forming network (PFN). The thyratron/PFN system is setup with selectable pulse width and uses either a single or double thyratron depending on the pulse length. EEV CX7815 thyratrons are now used. A master oscillator, attenuator/phase shifter and traveling wave tube (TWT) amplifier system provides drive power for each klystron. A WR650 waveguide transmits power to each accelerator section. Flowing SF_6 gas inside the waveguide is used to cool the klystron and accelerator vacuum windows and also to protect against breakdown at the windows.

The klystrons themselves have been quite reliable over the years and typically last several thousands of hours. A couple of tubes have been rebuilt and spares purchased. The modulator systems, especially the interlocks, cause the most down time. Water flow and air flow, under current monitors are intermittent at times. A high-speed trip system was used in the early days but disconnected due to excessive false trips. Also, voltage breakdown in the thyratron drive grid bias circuit during HV turn-off is currently an issue.

III.C. Electron Guns

The ORELA electron gun shown in Fig. 3, is a triode structure in Pierce geometry.[4] It was developed and is built in house, because commercially available guns were not able to produce the required currents. The grid is a hexagonal web of molybdenum that is pulsed to about 1.5 kV to generate a ~24-A pulse at ~ 110 kV. Grid pulses are generated by an optically coupled thyratron-driven PFN pulser. The grid drive system and high-frequency cathode heater system are mounted in an SF_6-filled gun tank. The pulse length can range from 2–30 ns, with 8 ns and 6 A being typical operating values.

Fig. 3. Exploded view of the ORELA electron gun. The guns are built in-house.

The cathode material is a mixture of lacquer and Ba, Sr, and Ca carbonates that are converted into oxides. The material is electroplated on a nickel button.[5] This oxide type cathode was chosen because of its high emissivity at relatively low temperature (~850°C) to reduce the “dark” current thermally emitted by the grid. Depending on the accelerator vacuum quality, the electron gun lasts typically more than 1000 h. But a number of recent gun failures have been attributed to pin-hole vacuum leaks in the side of the insulator ceramic. It appears that this problem was caused by scattered electrons from the anode region, so the design recently was changed to protect the ceramic. EGUN modeling of the gun potential distribution and emission was performed to investigate the effect of extending the cathode ring or the high-voltage bushings. One gun with an extended ring was built and performed acceptably. A second gun with an extended high-voltage bushing performed on the test stand above specification (see Fig. 4) and showed very good high-voltage behavior. Also, the gun magnet alignment was improved to maximize coupling of electrons to the accelerator beam line and reduce anode overheating as a potential source of dark current emission.

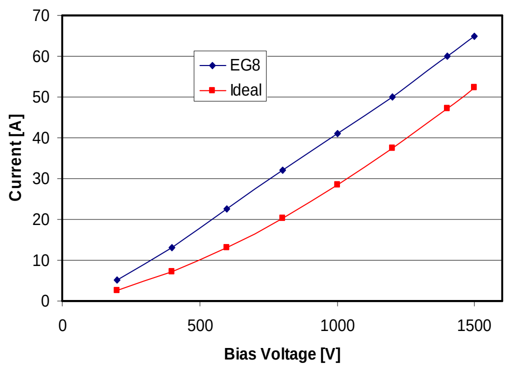

Fig. 4. Performance of electron gun EG8 cathode at 100-W heater power and a repetition rate of 100 Hz.

Improved cathode processing and emitter materials have been investigated to increase gun current. The use of a new filament was investigated because the original failed quite often during operation at high temperature while converting the cathode. High-voltage processing up to 160 kV is used to maintain stable operating conditions at the normally much lower operating voltage of about 110 kV. In addition, this procedure helps to minimize dark current.

IV. RECENT MEASUREMENTS

In the past year, new neutron cross-section measurements have been performed at ORELA to support nuclear criticality safety applications in the DOE complex. In particular, neutron capture and total cross-section measurements have been performed for ^{39,41}K and ^{55}Mn to improve the cross-section data in the resonance region. With regard to nuclear applications, ^{55}Mn is a key constituent of steel that is needed to improve nuclear criticality safety analyses of fissionable systems. Primary DOE applications involving ^{55}Mn include spent nuclear fuel (SNF) operations at the Savannah River and Hanford sites (e.g., K-Reactor cleanout, Plutonium Finishing Plant clean-up, tank farm operations, SNF storage, etc.). Operations involving ^{39,41}K include fissionable material operations at the Oak Ridge Y-12 plant.

For the cross-section measurements, ORELA was operated at a repetition rate of 525 Hz, a burst width of 8 ns, and a power of 5 to 6 kW. The combination of short pulse width and long flight path resulted in a neutron energy resolution superior to any previously reported measurements for these nuclides.

Neutron capture cross sections for ^{39,41}K and ^{55}Mn were measured on flight path 7 at a distance of 40 m from the neutron-producing target using a pair of deuterated benzene (C^6D^6) detectors by employing the pulse-height-weighting method. This apparatus[6] has been improved in several ways, compared to that used in previous measurements at ORELA for these nuclides. First, the original C^6F^6–scintillator was replaced by C^6D^6, which is much less sensitive to backgrounds from scattered neutrons. Second, much of the structural material surrounding the sample and detectors was removed to reduce the neutron sensitivity even further. For example, the massive Al-sample changer and beam pipe were replaced by a thin carbon fiber tube. Additionally, the massive detector housings were replaced with reduced-mass detector mounts. Third, the appropriate detector weighting function for each experiment is now calculated using the more sophisticated computer codes EGS4 and MCNP. All structural materials within 30 cm of the detectors, including the sample, are incorporated into these calculations. More details about these improvements can be found in the papers by Koehler et al.[6, 7]

The neutron flux was measured using a 0.5-mm-thick ^{6}Li glass scintillator at a distance of 39.695 m from the neutron target. The final normalization of the capture efficiency was carried out in a separate measurement using the “saturated resonance” technique by means of the 4.9-eV resonance from a gold sample.[8]

The neutron capture cross section of ^{55}Mn was measured using a solid metallic ^{55}Mn sample with dimensions of 2.54 x 5.08 x 0.317 cm. The experimental data are shown in Fig. 5 and compared to the cross section calculated using SAMMY[9] with ENDF/B VI resonance parameters. This calculation included experimental effects such as self-shielding and multiple scattering. There are substantial differences between ENDF and the data, including missing resonances in the evaluation.

For ^{39}K we used a natural potassium carbonate sample with a thickness of 0.0088 at/b. The ^{41}K neutron capture experiments using ^{41}KCl were performed with a sample enriched to 99.17% in ^{41}K and a thickness of 0.007972 at/b. Even though the sample contained chlorine, it will be possible to extract reliable resonance parameters for ^{41}K using SAMMY by including the most recent chlorine resonance parameters in the analysis.[10] The neutron capture cross sections obtained from this experiment show that there are discrepancies compared to the cross section calculated using the resonance parameters from the JENDL-3.2 library. Figure 6 shows a small part of the data from this measurement, illustrating that many resonances for ^{41}K are missing from the evaluation and that several resonances in the evaluation have been overestimated. The calculated cross section included experimental effects such as self-shielding, multiple scattering, and Doppler and resolution broadening.

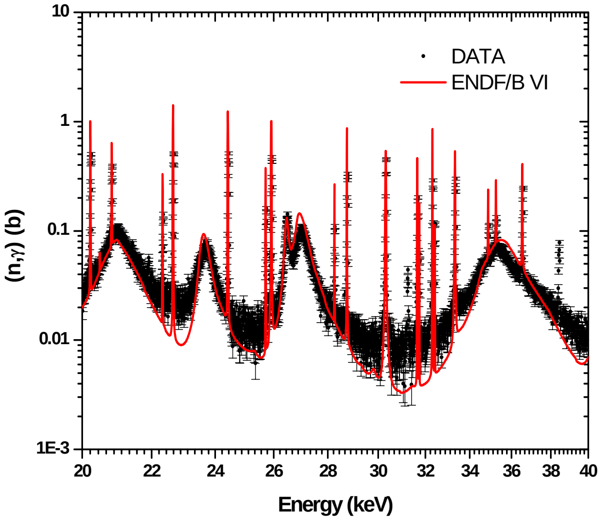

Fig. 5. Neutron capture data for Mn compared to the calculated cross section using the ENDF/B VI parameters, including all experimental effects.

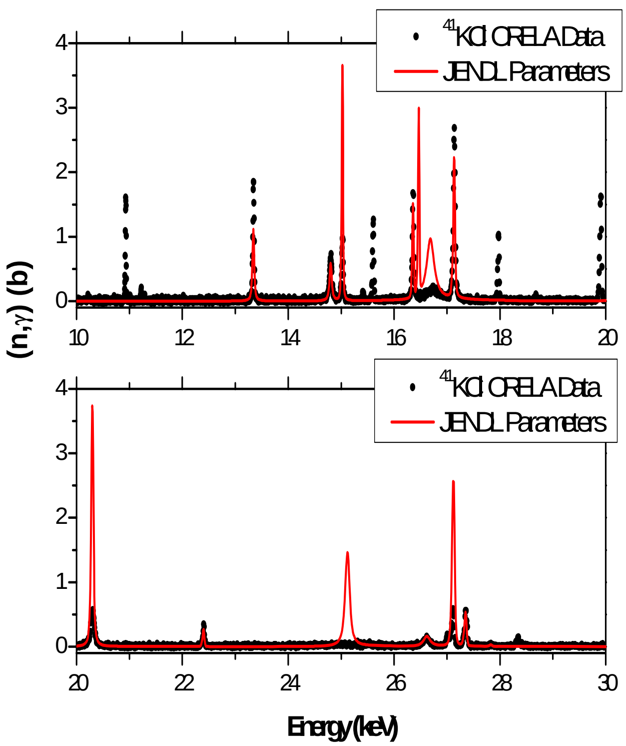

Fig. 6. Neutron capture data for ^{41}KCl compared to the calculated cross section using the JENDL 3.2 parameters, including all experimental effects.

Usually neutron capture experiments are performed with a fairly thick sample; therefore, the corrections for self-shielding and multiple scattering can be sizeable. To apply the most accurate corrections for these experimental effects, corresponding total cross-section measurements were performed at ORELA. These data also can be useful in making isotopic assignments as well as for observing some resonances not visible in the (n,γ) data. For the total cross-section measurements on ^{55}Mn, a metallic powder sample was used with a thickness of 0.119 at/b. It was mounted in a computer-controlled sample changer positioned at about 10 m from the neutron target in the beam of ORELA. The natural potassium transmission measurement was carried out using two metallic samples (0.013367 and 0.10517 atom/b). For ^{41}K, the same ^{41}KCl sample from the capture measurements was reshaped to yield a thickness of 0.0155 at/b. A presample collimator limited the beam size to 2.54 cm at the sample position and allowed only neutrons from the water-cooled part of the neutron source to be seen by the detector. As a neutron detector, an 11.1-cm-diameter, 1.25-cm thick ^{6}Li-glass scintillator was used in the beam at the 80-m flight station at a distance of 79.815 m from the neutron source.

The total cross section was measured using a modernized data acquisition apparatus. Two channels of an Acqiris digitizer were used to digitize signals from the ^{6}Li-glass detector and determine both TOF and pulse height for each event. The data were written to disk in event mode as well as displayed online in histograms. By using the digitizer many of the previous data acquisition electronics were removed; therefore the new system with fewer components should be much more reliable. A small portion of the two-dimensional data, TOF vs pulse-height, is shown in Fig. 7 for the ^{41}KCl transmission measurements.

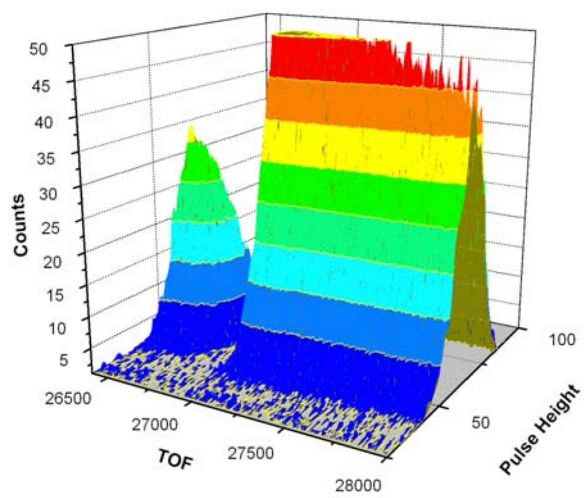

Fig. 7. Transmission data for ^{41}KCl acquired with the new data acquisition system, which records pulse-height vs TOF.

Efforts are in progress to analyze the measured cross-section data for $^{39,41}K$ and ^{55}Mn and prepare new resonance evaluations for inclusion in future ENDF/B cross-section evaluations.

V. OUTLOOK AND FUEL CYCLE DATA NEEDS

Currently, ORELA is readily available to address current and future nuclear data needs in the resonance region. The United States has initiated efforts to develop an advanced nuclear fuel cycle (AFC) to address global energy needs. The AFC initiative will present nuclear data challenges for deploying novel nuclear systems. Moreover, basic science R&D will be needed to support closing the fuel cycle [e.g., SNF reprocessing, transportation, handling, etc.]. Establishing the safety basis for licensing applications for AFC operations will require the verification and validation of existing and new radiation transport modeling software and associated nuclear data with benchmark critical and/or subcritical experiments. Moreover, current and future work efforts are devoted to developing improved modeling and simulation capabilities; and commensurate improvements in the underlying nuclear data will be needed to support the higher fidelity modeling and simulation capabilities.

Over the past few years through the DOE/NE Advanced Fuel Cycle Initiative (AFCI) and Global Nuclear Energy Partnership (GNEP) programs, nuclear data needs have been established and prioritized by the Transmutation Physics Working Group. This Working Group has developed and updated a priority list of nuclear data needs based on advanced reactor sensitivity/uncertainty analyses. As a result, significant research has been performed by DOE/NE to identify reactor nuclear data needs; however, the nuclear data needs (i.e., including accuracy requirements) for the entire fuel cycle have not been clearly defined. Although a detailed data needs study for the entire AFC has not been performed, R&D recommendations can be made based on knowledge of existing measurement capabilities and the current state of nuclear data in the evaluated databases. The following materials have been identified as needing improved cross-section data with corresponding uncertainty (i.e., covariance) information.[11]

- ^{232}Th, ^{233}U, ^{234}U, ^{235}U, ^{236}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu,
- ^{241}Am, ^{242m}Am, ^{243}Am, ^{242}Cm, ^{243}Cm, ^{244}Cm, ^{245}Cm,
- Pb, Bi, ^{56}Fe, ^{57}Fe , ^{58}Ni, ^{52}Cr, Zr, ^{15}N, Si, C, O, Na, ^{10}B ^{1}H ,
- Ti (5 isotopes), ^{85}Rb, ^{87}Rb

The data needs for these materials will span the complete energy range, and cross-section measurements will be needed in the resonance region and at high energies above the resonance range.

As part of the AFC plan, the fuel cycle will be closed, thereby requiring that SNF be reprocessed to produce new fuel for nuclear reactors. During reprocessing, two situations develop:

1. higher mass number isotopes of plutonium, americium, and curium build up, and
2. the transition from fluid to solid form in the fuel reprocessing involves systems, which establish intermediate- and thermal-energy neutron spectra.

The nuclear data for many of the actinides anticipated in the fuel reprocessing streams are not well known at intermediate energies that encompass the resonance region. Moreover, the safety basis for efficiently sized equipment, in terms of inventory and throughput, will require the demonstration of acceptable margins of sub-criticality. ORELA will be needed to address these data needs in the resonance region.

In addition to the reprocessing component of the advanced fuel cycle, additional operations will involve material handling and SNF transportation in approved shipping casks. The following are possible issues that should be investigated to assess differential data needs for advanced fuel cycle applications:

- Improved fuel exposure prediction of spent fuel isotopics (actinides and fission products);
- Improved prediction of spent fuel reactivity worth for criticality safety burnup credit (BUC) that is needed for transportation in addition to efficient sizing of reprocessing equipment; and
- Improved prediction of neutron radiation source terms, required neutron shielding and subsequent neutron reflection in criticality evaluations
- Improved cross-section data for isotopes acting as chemical reagents (important for neutron moderation and absorption).

For the above list of AFC issues, BUC will be a significant issue that will be important for the transportation and handling of SNF. BUC consists of taking credit for the reactivity decrease associated with the presence of fission products in the SNF. Past efforts by DOE, the Electric Power Research Institute (EPRI), and the Nuclear Regulatory Commission (NRC) have provided sufficient technical information to enable the NRC to issue regulatory guidance for implementation of pressurized-water reactor (PWR) burnup credit. However, consideration of only the reactivity change due to the major actinides is recommended in the guidance. Approximately one-third of the reactivity of a SNF cask can be attributed to fission products and minor actinides. Burnup credit could potentially save hundreds of millions of dollars in the cost of SNF transportation. Moreover, fission product cross-section data improvements will be needed to help strengthen the safety basis for BUC.

As the AFC program continues to evolve, additional nuclear data needs will be identified; however, some AFC analyses have been performed, and these studies have identified cross-section data needs that span energy ranges from the low eV region to the MeV region. Therefore, full-range nuclear data measurements will be needed to address current AFC data needs. ORELA can be used to address the key cross-section data needs in the resonance region.

VI. SUMMARY

ORNL has a ND program that is integrated with nuclear fuel cycle research and development activities. The ORELA measurement facility is a key neutron science facility within the ORNL ND program. As a result, ORELA neutron cross-section measurements and corresponding evaluations are performed in concert with nuclear science technology needs and requirements thereby providing a complete feedback loop between nuclear applications work and basic science measurements. In the past year, facility refurbishments have been completed to ensure reliable ORELA operation that has been further demonstrated by the completion of neutron cross-section measurements for ^{39,41}K and ^{55}Mn. ORELA is a proven, well-characterized accelerator facility that has been used for more than 30 years to address U.S. nuclear data measurements needs. Moreover, ORELA is readily available to address current and future nuclear data needs in the resonance region to support U.S. advanced fuel cycle intiatives.

ACKNOWLEDGMENTS

The authors would like to acknowledge the NNSA Nuclear Criticality Safety Program for providing operation and refurbishment support for ORELA and providing support for the ^{39,41}K and ^{55}Mn measurements. Also, the authors acknowledge the operation support by the Office of Nuclear Physics within the DOE Office of Science for ORELA. Research was sponsored by Oak Ridge National Laboratory, managed by UT-Battelle, LLC, for the U.S. Department of Energy under contract DE-AC05-00OR22725.

REFERENCES

1. K. H. Guber, L. C. Leal, R. O. Sayer, P. E. Koehler, T. E. Valentine, H. Derrien, and J. A. Harvey, "New Neutron Cross-Section Measurements at ORELA for Improved Nuclear Data Calculations," International Conference on Nuclear Data for Science and Technology (ND2004), 1706–1711, Santa Fe, NM (September 26–October 1, 2004).

2. M. E. Dunn, ed., *Proceedings of the Oak Ridge Electron Linear Accelerator* (ORELA) Workshop ORNL/TM-2005/272, July 2005.

3. N.C. Pering and T.A. Lewis, "Performance of a 140 MeV High Current Short Pulse LINAC at ORNL," *IEEE Trans Nuc. Sci* NS-16, 316 (1969).

4. J. R. Pierce, *Theory and Design of Electron Beams*, Van Nostrand, 1954.

5. O. W. Christian and T. A. Lewis, ORNL/TM-7886, Oak Ridge National Laboratory, 1981.

6. P. E. Koehler, R. R. Spencer, R. R. Winters, K. H. Guber, J. A. Harvey, N. W. Hill, and M. S. Smith, *Phys. Rev. C*, **54**, 1463–1477 (1996).

7. P. E. Koehler, R. R. Winters, K. H. Guber, T. Rauscher, J. A. Harvey, S. Raman, R. R. Spencer, J. C. Blackmon, D. C.Larson, D. W. Bardanyan, and T. A. Lewis, *Phys. Rev. C*, **62**, 055803-1 (2000).

8. R. L. Macklin, J. Halperin, and R. R. Winters, *Nucl. Instrum. Methods*, **164**, 213 (1979).

9. N. M. Larson, ORNL/TM-9179/R7, Oak Ridge National Laboratory, 2006.

10. R. O. Sayer, K. H. Guber, L. C. Leal, and N. M. Larson, *Phys. Rev. C* **73**, 044603 (2006).

11. Lee Schroeder and Ewing Lusk, ed., "Report of the Nuclear Physics and Related Computational Science R&D for Advanced Fuel Cycles Workshop," Sponsored by the U.S. Department of Energy Office of Science, Office of Nuclear Physics, and Office of Advanced Scientific Computing Research, Bethesda, MD, August 10–12, 2006.