Accelerator-Based Nuclear Data Measurements for Nuclear Science and Technology

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ABSTRACT

Accelerator-development and new experimental techniques at accelerator-based neutron sources are at the heart of improvements of nuclear data for science and technology. An important driver is the new interest for safety estimates based on full uncertainty propagation as opposed to the exclusive use of safety margins based on expert judgment. Large scale modeling facilitated by the massive growth of computing power emphasize the importance of nuclear data uncertainties which are often the main contributor to performance uncertainty. Advanced reactor development with its emphasis on resource economy,

minimization of high-level waste and improved safety is another important driver. Accelerator driven systems may prove essential new systems for closing the fuel cycle. Global security, emergency preparedness and safeguards change the focus towards fission and neutron- and gamma-induced yields that help forensics and detecting illegal activities
The challenges for nuclear data lie in the systematic provision of uncertainties and covariances, the substantial improvement of well-known key quantities such as those addressed in the CIELO initiative (Collaborative International Evaluated Library Organisation) and addressing some of the elusive nuclear data for minor actinides which are essential in addressing the high-level waste issues. In the field of fission measurements spectacular new techniques provide a wealth of data that opening new avenues. An overview of recent developments is presented from leading facilities around the world with a short outlook of where these developments may take us.

KEYWORDS
Nuclear science, Accelerator-based nuclear measurements, nuclear safety, security and safeguards

1. INTRODUCTION

The experimental advances and facility upgrades described in this paper contribute to a common international objective to improve our knowledge of essential nuclear cross section data. Numerous studies over the years have shown that the relatively good integral performance of our current evaluated data libraries, in criticality, transport, and safety studies, provides an overly positive perspective, because of the presence of compensating errors. For example, the neutron transport and criticality impacts of neutron inelastic scattering and fission neutron spectra reactions can work in complementary ways. If one uses the existing evaluated databases in applications that closely resemble the critical assembly validation benchmarks to which they have been calibrated, their present accuracy levels may be adequate. But for applications that extend to neutronics regimes further away from these calibration points, the existing databases (with their compensating errors) need improvement, for it is here that extrapolation errors become larger. This will be important for future nuclear energy, criticality safety, and nuclear waste disposition studies.

An additional important reason for these efforts is to better define our understanding of nuclear cross section uncertainties, so called covariance data. The evaluation projects across the world have been devoting significant attention to this topic, with a goal of developing more credible covariance data that are needed in nuclear technology applications, for determining performance margins and uncertainties. Measurements play a role in determining both the statistical component to uncertainty assessments, as well as systematic components (which, we believe, have often been underestimated). Measurements reported from different experimental techniques, employing complementary detection methods, are especially useful in guiding our understanding here.

2. THE JRC-IRMM NEUTRON SOURCES

The Joint Research Centre of the European Commission operates two neutron sources for neutron data measurements at its Institute for Reference Materials and Measurements. GELINA, the Geel electron linear accelerator provides a pulsed white spectrum neutron source for neutron time-of-flight measurements with a repetition rate of 40 to 800 Hz and a pulse width at half maximum of 1 ns. Time-of-flight measurements are made at flight paths of 8 to 400 m for the total cross section using the transmission technique, for capture and fission reactions, for elastic and inelastic scattering and for light charged particle emission reactions. At the Van de Graaff accelerator cross sections are measured for fission. Fission yields measurements combined with neutron or gamma spectra or multiplicity measurements are carried out with either a ^{252}Cf(SF) source, or with Van de Graaff based or GELINA based neutron-induced fission.

3. LOS ALAMOS NEUTRON SCIENCE CENTER (LANSCE)

LANSCE provides pulsed, intense sources of neutrons over 16 orders of magnitude in neutron energy, from ultra-cold neutrons to neutrons in the 100 MeV range. Experiments from sub-thermal to the highest neutron energies use time-of-flight techniques so that there are no gaps in energy for the reactions studied. Recently the LANSCE facility has been described as part of a white paper on nuclear data capabilities. [1]. Here we focus on the unique instruments and measurement possibilities.

Total cross sections: The unmoderated neutron source at LANSCE, called the Weapons Neutron Research Facility (WNR), is ideal for measurements of neutron total cross sections in the range from a few hundred keV to hundreds of MeV. Data on total cross sections were obtained with absolute uncertainties of 1% or less over a large range of nuclei [2]. Recently, with the use of waveform digitizers for data that are free of dead time, total cross sections of small isotopic samples are being studied [3] for insights into the optical model. For total cross sections in the resonance and unresolved resonance region, a new facility is being developed at the moderated neutron source at the Manuel Lujan Jr. Neutron Scattering Center (MLNSC). Small samples of isotopes including radioactive isotopes will be studied here.

Neutron capture cross sections and radiative decay mechanisms: The Detector for Advanced Neutron Capture Experiments (DANCE), a 160 element array of BaF2 scintillators covering close to 4-pi solid angle, is producing new, accurate and detailed information on neutron capture at the MLNSC. The high segmentation allows measurements of the multiplicity of gamma rays following capture which then is a window into the capture mechanism. Identification and quantification of the M1 strength in radiative decay have already led to better models to predict capture cross sections [4]. Further, the high segmentation offers a reliable method to separate neutron capture from neutron-induced fission when both are possible. This instrument has recently been enhanced by the addition of fission fragment and neutron detectors near the sample for even more detailed studies of the fission process.

Time-projection chamber for fission studies: As a joint project between Los Alamos and Lawrence Livermore National Laboratory (LLNL), a time-projection chamber (TPC) is used for fission studies including neutron-induced fission cross sections and angular distributions of fission products. Precision fission cross section measurements need information on the angular distributions of the fission products to correct for events where the fission products are emitted close to 90-degrees to the sample and therefore are stopped before they enter the counting gas. The present TPC consists of nearly 6000 readout channels to give good angular and energy loss data [5].

Fission product properties: The mass, charge and kinetic energy distributions in fission are studied with the SPIDER spectrometer and Frisch-gridded ionization chambers. SPIDER [6, 7] is a velocity and energy spectrometer that provides high mass resolution for fission products, and has been used to study thermal neutron-induced fission yields. Frisch-gridded ionization chambers [8] provide higher detection efficiency with lower mass resolution, and are used to measure the change in total kinetic energy (TKE) release in fission as a function of incident neutron energy.

Prompt fission neutron spectra: The Chi-Nu arrays of neutron detectors are producing data on the spectra of neutrons emitted in neutron-induced fission of actinides at WNR. Fission neutrons below 1 MeV are detected by an array of 22 lithium-6 glass detectors and, non-concurrently, neutrons above 0.5 MeV are detected by an array of 54 liquid scintillators. [9]. Fission is detected in a parallel-plate avalanche detector made by colleagues at LLNL. Investigations are in progress also for detection of neutron emission in general, such as in (n,xn) reactions either with a gamma-ray trigger or in a triggerless mode. Understanding of neutron scattering in the experimental environment, which is critical to assigning systematic uncertainties, is based on neutron transport codes such as MCNP.

Light-charged particle production: Reactions producing charged particles, (n,xp), (n,d), (n,xα), etc., previously studied at LANSCE, are now being measured with a new detector setup that has a much larger solid angle for detecting the charged particles. LENZ (Low Energy NZ) consists of ionization detectors for charged-particle identification and large, segmented silicon surface barrier detectors for angle and energy measurement. At present, this capability uses neutrons from the WNR neutron source.

Neutron-induced gamma-ray production: The Germanium Array for Neutron-Induced Excitations (GEANIE), with 26 HPGe detectors, used for many years for productive measurements of (n,xγ) reactions at WNR (e.g. ref. [10]) has been disassembled. The detectors still exist however and could be used for further studies of compound and pre-equilibrium reactions and nuclear structure where gamma rays in the product nuclei are measured.

Lead slowing-down spectrometer (LSDS): An LSDS, driven by protons from the Proton Storage Ring, produces very high neutron fluxes for measurements on very small samples (10's of ng) especially for experiments on neutron-induced fission [11] and (n,α) reactions below 100 keV.

4. N_TOF FACILITY AT CERN

Since 2001, a wealth of neutron capture and neutron-induced fission reactions has been measured at n_TOF, providing an important contribution to a wide variety of research fields. The experimental activity is driven mostly by two motivations: on the one hand, capture reactions are studied with the aim of improving current models of stellar nucleosynthesis of heavy elements, on the other hand fission reactions are studied applications in energy. A review of the needs related to nuclear astrophysics and the contribution of the n_TOF program can be found in [12, 13].

At n_TOF, cross-sections are measured as a function of the neutron energy, as reconstructed from the neutron time-of-flight. The pulsed neutron beam is produced by spallation of 20 GeV/c protons, from the PS accelerator, impinging on a massive Pb target surrounded by water for cooling and neutron moderation purposes. An additional layer of borated water is placed on the exit surface of the spallation target for further moderation. For the first 15 years of operation, measurements have been performed in an experimental area located at the end of a 185 m long horizontal tunnel. The main features of the neutron beam in the first experimental area (EAR1), are the high instantaneous intensity of 10^6 neutron/bunch, the wide energy range, from 25 meV to over 1 GeV, the low repetition rate of <1 Hz, and the high resolution in neutron energy ($\Delta E/E = 10^{-4}$ for most of the energy range). While high-quality data can be collected in EAR1 for radioactive isotopes with relatively long half-life, of more than a few thousand years, and on samples of masses above a few tens of mg, for short-lived radioisotopes and for smaller samples the count rate starts to be dominated by the background, in particular the one related to the natural radioactivity of the sample.

To address the needs of data for short-lived radioisotopes, or for stable isotopes available in extremely small amounts, the n_TOF Collaboration proposed the construction of a new experimental area (EAR2) at a shorter distance from the spallation target to exploit a much higher neutron flux [14, 15]. Since the spallation target is placed underground at approximately 20 m under the surface, a very convenient solution was to build the new experimental area just on top of the pit hosting the target, in the vertical direction. The large gain in the neutron flux, of about a factor of 30 relative to the first experimental area, allows one to perform measurements with samples of correspondingly smaller mass or in a shorter time. Most importantly, the combination of the higher flux and shorter time-of-flight, a factor of 10 relative to EAR1, is particularly convenient when measuring radioactive isotopes, as it results in an increase of the signal-to-background ratio of more than two orders of magnitude for the background related to the radioactive decay of the sample. As a consequence, in EAR2 [14, 15]. it becomes feasible to perform challenging measurements with isotopes of half-life as short as a few tens of years, offering the unique opportunity to address some open questions in nuclear astrophysics, in applications related to energy production, both from fission and fusion, in nuclear medicine, in fundamental nuclear physics, and in related fields.

The first of such physics measurement was performed in the fall 2014. It concerned the ^{240}Pu(n,f) cross-section, of importance for energy production and waste transmutation projects. Due to the large specific α-activity of ^{240}Pu, this is a very challenging measurement. Previous attempts to measure the cross-section below the fission threshold, made in various facilities around the world, had failed. This successful measurement demonstrates the enormous potentiality of the new beam-line, paving the way to challenging fission measurements of short-lived actinides.

Another very challenging measurement planned for the near future in EAR2 is the ^{7}Be(n,α) reaction relevant to the so-called "Cosmological Lithium Problem." This reaction has never been measured before at neutron energies of astrophysical interest, while different theoretical models predict completely different trends of the cross-section as a function of neutron energy. The difficulty of this measurement is related to the very low reaction cross-section and to the short half-life of ^{7}Be (56 days), which results in a huge activity, of the order of tens of GBq, even for samples of a few mg mass. In this respect, the new experimental area at n_TOF is rather unique, as it offers the possibility to measure this reaction in a wide neutron energy range with even a small amount of ^{7}Be, thanks to the extremely high instantaneous neutron flux.

5. THE GAERTTNER LINAC CENTER AT RPI

The Gaerttner LINAC Center at RPI uses a 60 MeV pulsed electron LINAC to produce neutrons that span the energy range from sub-thermal to 60 MeV [16]. Different types of experimental setups are used to cover different parts of this energy range. These setups combine a neutron production target optimized for a specific energy range and a detector setup. Measurements are done using the time of flight (TOF) method; several setups to measure neutron induced reactions are available at the facility and are listed in Table I. In addition to conventional TOF, the facility also houses a 67 metric ton lead slowing down spectrometer (LSDS) that is used for fission and capture measurements and research on assay of spent nuclear fuel.

Table I. Experimental capabilities at the RPI LINAC

Flight Path distance [m]	Type of Experiment	Detector Type	Useful Energy Range
15	Transmission	Thin Li-glass	Thermal to 20 eV
25	Transmission and capture	Li-glass and NaI multiplicity	Thermal to several keV
30	Fast and epithermal neutron scattering, Prompt fission neutron spectrum	Liquid scintillator array, plastic and Li-glass	Few keV to 20 MeV
35	Transmission	Li-glass	Few eV to several tens of keV
45	Capture	Four C_6D_6	Few eV to 2 MeV

100	Transmission	Modular Li-glass	Few eV to 0.5 MeV
250	Transmission	Modular liquid scintillator	0.5 to 20 MeV
LSDS	Fission, fission fragments spectroscopy, capture, assay of spent fuel	Fission chambers and YAP scintillators.	0.1 eV to 100 keV

Recently the activity was focused on fast neutron scattering [17] and total cross sections, keV neutron capture [18] and prompt fission spectrum measurements [19].

6. A BRIEF VIEW OF ANNRI

The Accurate Neutron-Nucleus Reaction measurement Instrument (ANNRI) was constructed by the collaboration of Hokkaido University, Tokyo Institute of Technology and JAEA. ANNRI is located on Beam Line No. 04 of the materials and life science experimental facility (MLF) in the Japan Proton Accelerator Research Complex (J-PARC). Neutrons are supplied from a coupled moderator that provides the most intense neutron beam among the moderators[20].

Two detector systems are installed in ANNRI. An array of large germanium (Ge) detectors is the main detector of ANNRI and located at a flight-length of 21.5 m. The array of Ge detectors is composed of two cluster-Ge detectors, eight coaxial-Ge detectors with anti-coincidence BGO detectors. Each cluster Ge detector consists of seven Ge crystals. The peak efficiency of the spectrometer is 2.28 ± 0.11 % for 1.33 MeV γ rays [21]. A NaI spectrometer located at a flight-length of 27.9 m. The NaI(Tl) spectrometer is composed of two anti-Compton NaI(Tl) scintillators located at 90 degree and 125 degree with respect to the neutron-beam line [22]. The measurement using the NaI(Tl) spectrometer has two objectives. One is to perform the complementary measurement to that using the Ge array. The other is to extend the upper limit of neutron energy region in the measurement.

Fig.1 shows a comparison of the neutron intensity at the 21.5-m sample position under proton beam power of 17.5 kW with those of DANCE at LANSCE, and n-TOF [23]. Current proton beam power is 500 kW. The present neutron intensity under the 500 kW operation and that under the future 1-MW operation are also shown in Fig. 1. As seen from Fig. 1, the present and future neutron intensities are higher than those of the other facilities by about one order of magnitude. Neutron-capture cross sections are deduced by using ANNRI with a small amount (less than 1 mg) of a high radioactive sample.

Currently, analyses for ^{244}Cm, ^{246}Cm [24], ^{241}Am [25], and ^{237}Np [26] have been finished, analyses for ^{129}I, ^{107}Pd, ^{99}Tc, ^{93}Zr and some stable isotopes are in progress.

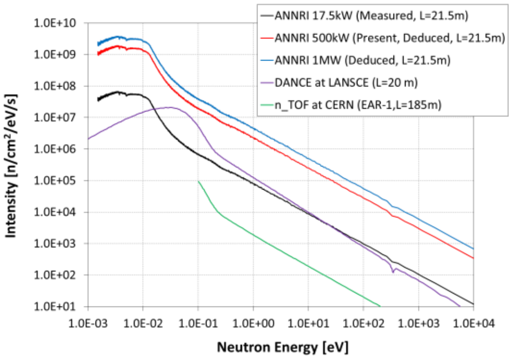

Figure 1. Neutron intensities per second at the 21.5-m sample position of ANNRI comparison to those of DANCE at LANSCE, and n TOF at CERN [23].

7. NEUTRONS FOR SCIENCE AT SPIRAL-2

The “Neutrons For Science” (NFS) facility is one of the experimental areas of SPIRAL-2 on the GANIL site at Caen (France). SPIRAL-2, designed for the production of very intense radioactive beams, is mainly composed of a high-power superconducting driver linear accelerator. The accelerator, today under commissioning, will deliver high-intensity beams (up to 5 mA) of proton, deuteron and heavy ions.
NFS is composed of two rooms: a converter cave where neutrons are produced and a large time-of-flight hall (30 m x 6 m). The two rooms are separated by a thick wall containing the collimator defining the neutron beam. A continuous energy distribution extending up to 40 MeV can be generated by the deuteron break-up reaction on a thick converter made of carbon or beryllium. Quasi-mono-energetic spectrum up to 33 MeV will be produced by the ^{7}Li(p,n)^{7}Be reaction on a thin converter.

The expected flux, with the maximum authorized primary beam intensity, 50µA of deuteron at 40 MeV, and a thick converter, is presented on figure below and compared to other major TOF facilities, namely n_TOF at CERN, WNR at Los Alamos and GELINA in Geel. Between 1 and 35 MeV, NFS is very competitive in terms of average flux in comparison with these facilities. However the instantaneous flux (by burst) will be less intense at NFS. The energy of the incident neutron can be measured by the TOF technique with a rather good resolution thanks to a small time spread at the converter point (shorter than 1 ns) as well as to the length of the experimental area.

The ion beam line in the converter cave is also equipped with an irradiation box for the measurements of ions induced reactions cross-section by the activation technique. The energy range extend up to 33 MeV for proton, 20 MeV/A for deuterons or He4 beams and 14.5 MeV/A for heavier ions. The irradiation by very intense neutrons flux will also be possible by putting the sample very close to the converter. For these experiments NFS will be equipped of a pneumatic system to transfer the sample from the irradiation point to the spectrometry chain and reverse.

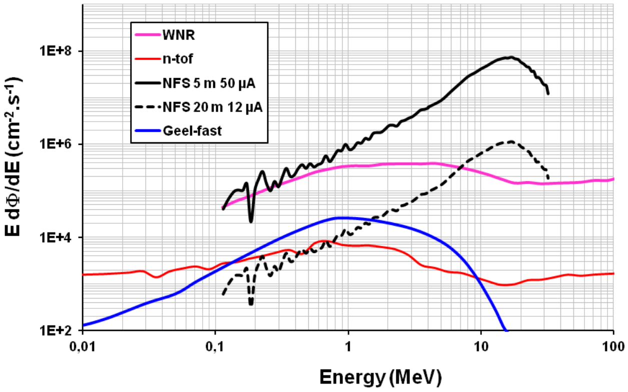

Figure 2: Comparison of the neutron flux of NFS with three other TOF facilities, WNR, n-tof and GELINA. The fluxes for NFS are shown at 5m and 20 m.

NFS will be a very powerful tool for physics, fundamental research as well as applications like the transmutation of nuclear waste, design of future fission and fusion reactors, nuclear medicine or test and development of new detectors. Letters of intents, covering a very wide domain of experiences, have been submitted to the scientific committee of SPIRAL-2. The study of n,xn reactions is of prime interest and NFS is particularly well suited for these experiments. The neutron induced fission is also a major topic of study and several experiments are foreseen to measure fragment distributions as well as neutron and gamma energy spectra and multiplicity distributions. The measurement of cross-sections by activation technique in neutron, proton and deuteron induced reactions will also give access to new data of interest for the fusion technology or the medical applications. Additional experiments dedicated to the study of single event upset or the characterization of neutrons detectors will be performed as well. The commissioning of the facility should start by the end of 2016 and the first experiments are expected for 2017.

8. CHINA SPALLATION NEUTRON SOURCE BACKSTREAMING NEUTRON BEAM

The China Spallation Neutron Source (CSNS) is a large scientific facility dedicated mainly to multidisciplinary research on material characterization using neutron scattering techniques [27, 28]. This project was officially approved by the Chinese central government in 2008, and will be in operation in 2018 for the 1st stage. In the first phase (CSNS-I), the machine will deliver 1.6 GeV protons on a tungsten target (TS1 [29, 30]) with 100 kW beam power and a pulse repetition rate of 25 Hz. A back-streaming neutron beam line based on this machine was proposed and constructed mainly for nuclear data measurements.

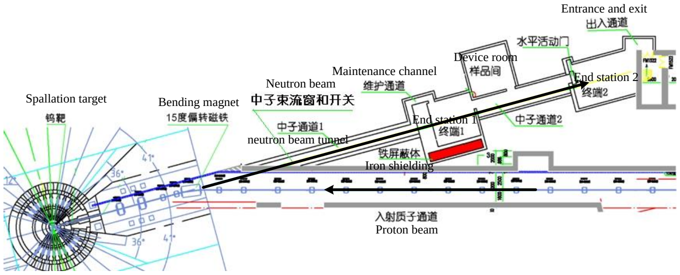

Figure 3. Layout of the CSNS back-streaming neutron beam tunnel and the experimental halls.

A detailed study of the characteristics for the back-streaming neutrons at CSNS is described in ref. [31]. Based on this study, a nuclear data measurement program using the CSNS back-streaming neutron beam was proposed. The layout of the neutron beam tunnel and the experimental halls are shown in fig.1. Two end stations were constructed, with ~50 meters flight path for end station 1 and ~80 meters for end station 2, respectively. The end station 1 will be used for higher neutron flux required experiment while the end station 2 for better time resolution required experiment. In the first stage, the neutron total cross section measurement, capture cross section measurement with C6D6 detectors and fission cross section measurement with a normal ionization chamber are planed with this back-streaming neutron beam.

9. CONCLUSIONS

This paper highlights the achievements and potential of state-of-the-art accelerator-based neutron sources and experimental facilities focused on nuclear measurements for science and applications. Of particular interest are the range of techniques available for addressing all aspects of neutron-induced nuclear reactions for the benefit of nuclear applications: energy, safety, security, medicine, and fusion. In the field of nuclear science stellar nucleosynthesis attracts wide interest. We firmly believe that upcoming accelerator developments, new measurement instruments, improved data-acquisition and data-handling will continue to provide the means to tackle challenges where data are lacking, uncertainty estimates are essential, and where correlated emissions allow new applications.

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