

$^{16}\text{O}(\text{n},\alpha)$ cross section investigation using LENZ instrument at LANSCE

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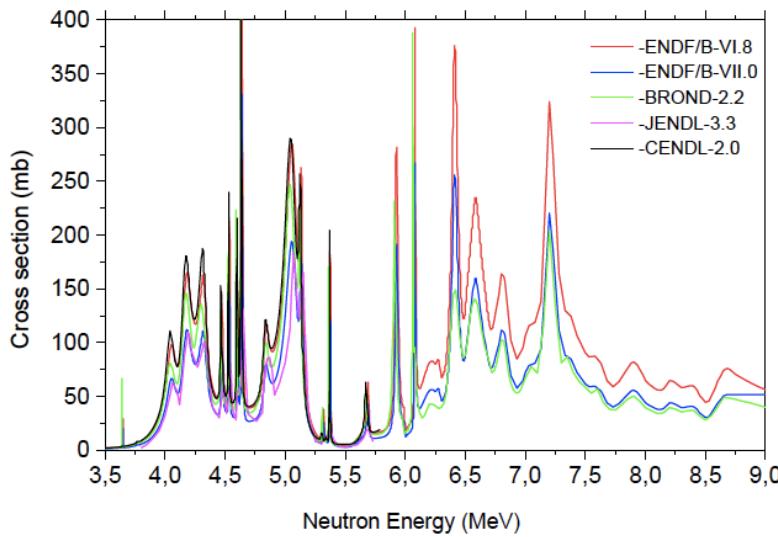
Abstract. Importance of studying the $^{16}\text{O}(\text{n},\alpha)$ reaction is motivated by multiple nuclear applications. The Los Alamos Neutron Science Center (LANSCE) produces a white neutron spectrum ranging from thermal to several hundreds of MeV energies. We have recently developed the LENZ (Low Energy NZ-neutron induced charged particle detection) capability to measure high-precision (n,α) cross sections. In order to provide more reliable data, we have enhanced solid angle coverage, and improved signal-to-noise ratios and time-of-flight resolution by implementing digitizer waveform analysis. The LENZ was commissioned by studying the $^{59}\text{Co}(\text{n},\alpha)$ reaction with neutron beams in early 2015. For the $^{16}\text{O}(\text{n},\alpha)$ reaction, we investigate solid oxygen targets and make a relative measurement to a better known cross section, such as the $^{6}\text{Li}(\text{n},\alpha)$ reaction in order to further reduce systematic uncertainty. We will discuss the progress of the $^{16}\text{O}(\text{n},\alpha)$ study at LANSCE and the outlook for improving Hauser-Feshbach prediction on (n,p) reaction cross sections.

1 Introduction : New Nuclear Data needs for the $^{16}\text{O}(\text{n},\alpha)$ reaction

Oxygen is present in many materials - water, oxides, concrete and elsewhere - and the uncertainties in its nuclear data can have a significant impact on many applications. Several nuclear applications require precise nuclear reaction cross sections on water. Naval reactors are light water reactors for cooling and moderating, which require neutron reactivity input for reactor designs. Data testing with “solution critical”, which contains a lot of water, is to benchmark critical assemblies. Experiments that use a manganese bath measure the total number of neutrons through the activation of manganese, therefore precise knowledge on neutron-induced reactions on water is necessary. Radio-biology takes advantage of better understanding of reactions related to oxygen, due to the bio-system’s oxygen content. In particular, neutron-absorption reactions cause reduction of available neutrons in applications. Therefore, improving the precision of the evaluated $^{16}\text{O}(\text{n},\alpha)$ reaction cross section is in order and for that we need to provide a new and independent set of experimental data.

This has been recognized and is the motivation for its inclusion as part of the Collaborative International Evaluated Library Organization (CIELO) project [1] looking at improving the nuclear data for key isotopes. At the heart of the questions on the oxygen-16 data is a 30-50% discrepancy between various (n,α) cross section measurements. Reconciling these discrepancies and settling on a best value are key deliverables for the CIELO project and requires new measurements for confirmation.

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Figure 1. Summary of available evaluations on the $^{16}\text{O}(\text{n},\alpha)$ reaction taken from the NEMEA-7/CIELO meeting presentation [2]

2 Status of the $^{16}\text{O}(\text{n},\alpha)$ cross section evaluations : experimentalist point of view

Evaluated cross sections on the $^{16}\text{O}(\text{n},\alpha)$ reaction have shown discrepancy on normalizations and energy-dependent shapes, as shown in Figure 1. Six different evaluations are shown and there are no evaluations with good agreement in the whole energy range. In particular, the ENDF/B-VII.0 evaluation [3] was released in 2006 after including the experiment result from Ref. [4] and the whole normalization was reduced by about 30 % compared with the previous evaluation of ENDF/B-VI.0 [5]. Since then, a new normalization factor to apply to the ENDF/B-VII.0 evaluation has been suggested, based on additional new measurements and the recent development of the LANL R-matrix study on ^{17}O -nucleus system [6].

Previously measured cross sections (The subsets are shown in Figure 2) were performed by two different approaches . One is to measure the $^{16}\text{O}(\text{n},\alpha)$ reaction directly [2, 7–10]. Gas detectors like a Frisch-gridded ionization counter or a proportional counter were used in a gas mixture, which contained oxygen in order to measure $^{16}\text{O}(\text{n},\alpha)^{13}\text{C}$ reaction cross section. Since the target gas plays as a counting gas as well, identifying α signals from background signals is crucial in data analysis. Most measurements were performed by thorough data reduction in order to remove background contribution, however there could be incomplete reduction of non-linear behaviors to the foreground. Since neutrons are produced from charged particle beams impinging on a gas target, there was in-built significant energy broadening, which will be discussed in detail in the following Section 4.

The other is the time-reversed reaction of $^{13}\text{C}(\alpha,\text{n})^{16}\text{O}$ using the principle of detailed balance [4, 11, 12]. It is measured by impinging an alpha beam on a ^{13}C target and followed by neutron detection. In order to compensate decreasing cross sections at low energy, a thick target is used. However, self absorption of alpha particles in a thick target has to be corrected in order to disentangle an effective

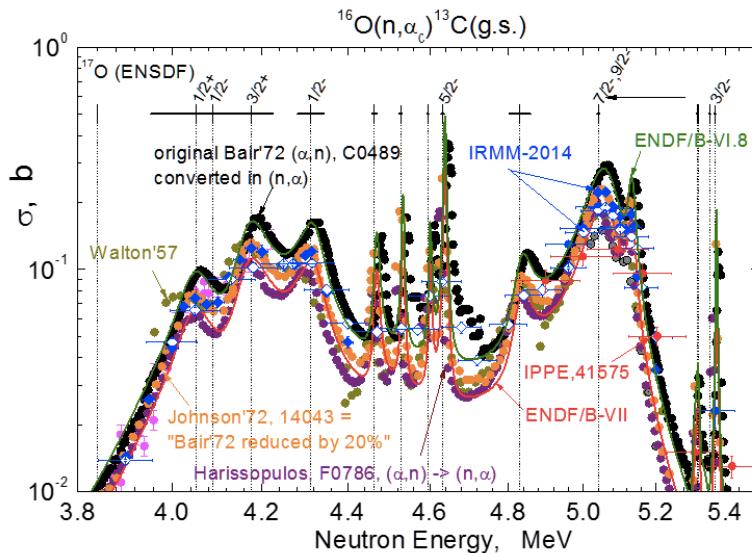


Figure 2. Subset of available experimental data on the $^{16}\text{O}(\text{n},\alpha)$ reaction taken from the NEMEA-7/CIELO meeting presentation [2]

energy using a realistic stopping power for straggling particles. The neutron detector efficiency is determined by simulating neutron transport in the detector system, which is then calibrated with the measured yield relative to a known reference. Bair and Hass [12] used a Sb-Be source to calibrate their neutron detectors. Harissopoulos *et al.* [4] used a ^{252}Cf spontaneous fission source, which has a shape of Maxwellian distribution with the temperature of 1.42 MeV and the peak around 1 MeV. The prompt fission neutron spectrum might not be optimal for calibrating neutrons above 4 MeV, where the number of available neutrons from fissions drops rather quickly. On other hand, the neutron energy populated from this $^{13}\text{C}(\alpha,\text{n})$ reactions ranges from 3.5 MeV to 9.0 MeV. The neutron detectors used here, like BF_3 counters or ^3He counters, can not provide neutron angular information, which has to be corrected from a Monte Carlo simulation using predicted angular distribution as input.

In order to enhance the quality of a new measurement, the requirements are (a) a large number of target atoms and a detection system with high efficiency, due to small cross sections, (b) a large signal-to-background ratio and low detection threshold, in order to capture all the low energy alphas from the reaction, (c) a good energy resolution, since some resonances have their resonance width less than 100 keV, (d) an improved systematic uncertainty for resolving the discrepancy among current data sets, and (e) additional angular distributions would improve data analysis for using advanced R-matrix codes.

3 LENZ (Low Energy NZ) capability

At LANSCE, neutrons are produced in the energy range of thermal and several hundreds MeV [13]. A 800 MeV proton impinges a bare tungsten target through spallation process, then produces neutrons in white spectrum to be delivered to 6 different flight paths at Weapons Neutron Research (WNR). Protons are pulsed to have the time structure of 1.8 μs repetition, so neutron energy is determined by

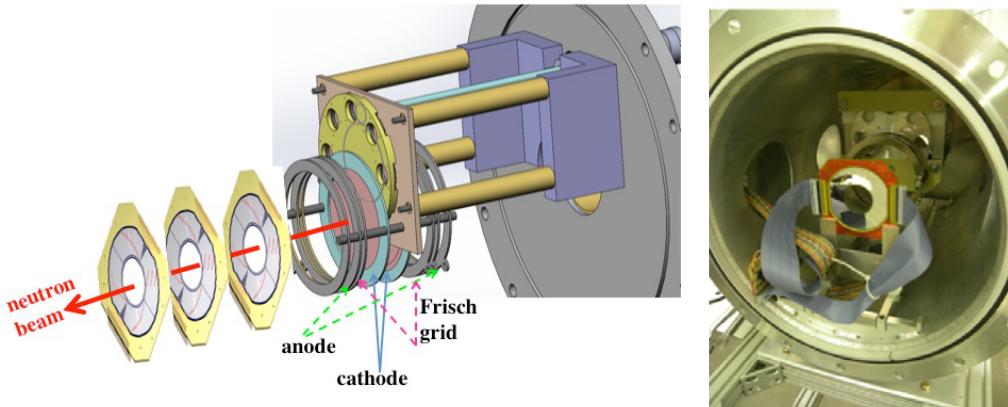


Figure 3. (left) The drawing of LENZ detector diagram (right) The picture of LENZ chamber

measuring the time stamp at the location of neutron detection relative to the proton time stamp right before bombarding the tungsten target. This broad neutron energy range and neutron flux at LANSCE are ideal to study the double differential cross sections for these (n,p) and (n,α) reactions.

The LENZ instrument is an update of the preceding NZ chamber [14] at LANSCE, to measure cross sections of neutron-induced charged particle reactions. The LENZ instrument is designed to have a large solid angle and low detection threshold in particular measuring low-energy alpha particles. It is composed of twin Frisch-gridded ionization counter [15] as a delta-E detector and double-sided silicon strip detectors (DSSD) as E detector to be used as “telescope”, as shown in Figure 3.

All the electrodes in an ionization counter are 63.5 mm in radius and the distances for grids and anodes relative to cathodes can be easily adjusted, in order to be flexible for detecting different types and/or energy range of charged particles. The Frisch grid signal allows to correlate emitting angles through the arrival time at the grid and the fast timing of the cathode signal is specifically optimized to provide a good neutron energy resolution, which was deduced from the time of flight method. The target wheel can hold up to 8 different targets, including a backing material, targets of interest, a calibration target, a normalization target, etc. The DSSDs include two of 1000 micron thick S1 type detectors (an active area is defined by a disc of the 48 mm outer radius with a hole of the 24 mm inner radius) and one 1500 micron thick S3 detector (an active area is defined by a disc of the 35 mm outer radius with a hole of the 11 mm inner radius). The location of these DSSDs can be configured to optimize each experiment’s goal, depending on either largest solid angle coverage or largest proton energy coverage. For the maximum solid angle coverage, the combination of the twin ionization counter and DSSDs can cover up to 70 degrees. The gas of P10 was chosen due to expected neutron-beam induced backgrounds on gas elements. Based on previous studies in the literature [16], the prominent background peaks in the energy spectrum are reported, due to the reactions like $O(n,\alpha)$, $Ar(n,\alpha)$, and recoil protons. Therefore, depending on the reaction of interest, meaning the potential overlap of the pulse heights with these background reactions, the beam-induced background reactions on gas elements need to be investigated case by case.

Since LANSCE provides a white neutron energy spectrum, the beam energy information is deduced from a time of flight at a target, in this case the time stamp measured in cathode. For reducing the energy resolution, the timing information needs to be preserved as precise as possible. We used waveform digitizers, which allowed us store partial wavelet information to obtain the best timing and

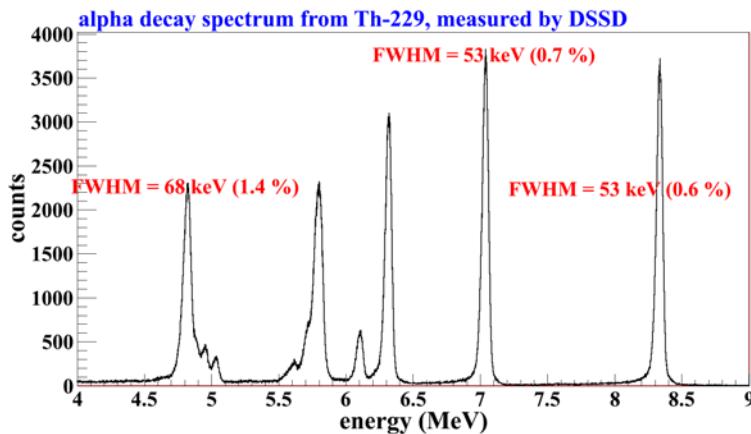


Figure 4. Energy of a DSSD detector is calibrated with ^{229}Th α -emitting source using a CAEN V1724 digitizer.

energy resolution, via post-processing with the use of various digital filters. Digital waveform analysis was implemented to achieve better signal-to-noise ratios, lower alpha detection threshold, and better timing resolution of the cathode signal. In order to match the characteristics of each detector preamplifier signal, two different types of CAEN digitizers [17] were tested. The preamplifier outputs of ionization counter signals have about 500 ns rise time with about 50 μs fall time, and the pulse height is up to 200 mV, with a 550 Torr. of P10 gas pressure. The preamplifier outputs of DSSD signals have about 40 ns rise time with about 35 μs fall time. When the ionization counter signals were used with the CAEN 1720 digitizer, which is a 12 bit and 250 MS/s sampling rate digitizer, the

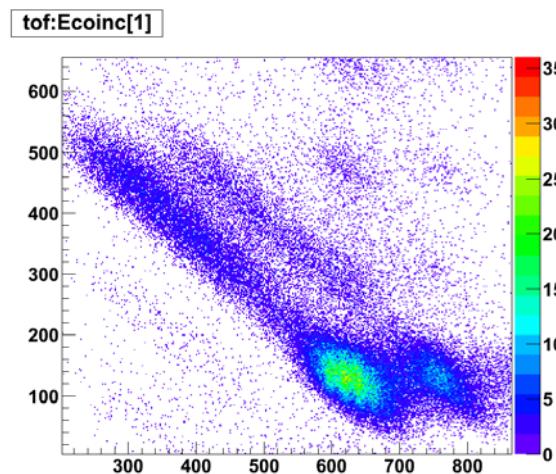


Figure 5. 2d plot of Time of Flights between T0 - Cathode (x-axis) and DSSD - Cathode (y-axis). Both axes are in arbitrary units.

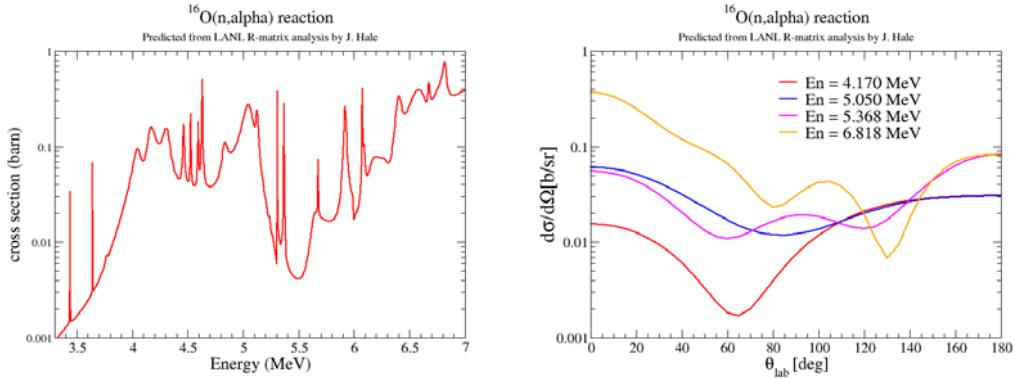


Figure 6. (left) The calculated reaction cross section of $^{16}\text{O}(\text{n},\alpha)$. (right) Calculated angular distributions for several strong resonances between 4 and 7 MeV of neutron energies. Both are performed using the Los Alamos developed R-matrix analysis by G. Hale [18].

time information was taken by using a double-derivative filter and peak height was charge integration. For DSSD detector signals, the CAEN 1724 digitizer with a 14 bit and 100 MS/s sampling rate was used to preserve most peak height information by fitting to an exponential decay from saved wavelets.

The parameter optimization to be used in these digital filters was performed for both detector signals using a ^{229}Th source. Figure 4 demonstrates the energy resolution in Full Width Half Maximum obtained for DSSD detectors in ~ 30 mTorr. vacuum pressure, without P10 gas being present. The energy resolution is determined to be as good as 53 keV above 6 MeV, which is better than the specification, 75 keV, provided by the Microsemiconductor manufacturer [19].

The in-beam commissioning of the LENZ instrument at LANSCE was performed by repeating the $^{59}\text{Co}(\text{n},\text{p})$ and (n,α) reaction, which were measured previously using the NZ chamber [20]. This is in particular to investigate the beam-induced background reactions on gas elements and to determine the timing resolution of detectors relative to the incoming neutron time stamp. For this run, the P10 gas pressure at 550 Torr. was chosen to generate enough energy loss for protons and alphas in the gas volume and one S1 DSSD was coupled to the ionization counter at forward angles. As shown in Figure 5, the different time of flight groups were measured to correlate the information on different reaction channels. With higher incoming neutron energy, different types of charged particles were populated through energetically allowed different reaction channels. The detailed analysis, including optimization of best timing resolution in ionization counter signals, is still on going.

4 The $^{16}\text{O}(\text{n},\alpha)$ measurement at LANSCE

Regarding the discrepancy on the $^{16}\text{O}(\text{n},\alpha)$ cross sections, which is known to be 30-50 % among currently available experimental data sets, we plan to improve the quality of this measurement by achieving the uncertainty to be less than 20 %. For absolute cross section measurements, the major uncertainty is caused by estimating the detection efficiency including a dead time in data acquisition system, the beam flux, the amount of target material, and the target uniformity and the beam profile if beam has a sizable area. A relative measurement to the reaction with a better known cross-section knowledge, called a “ratio method”, could be performed to reduce the uncertainties associated with

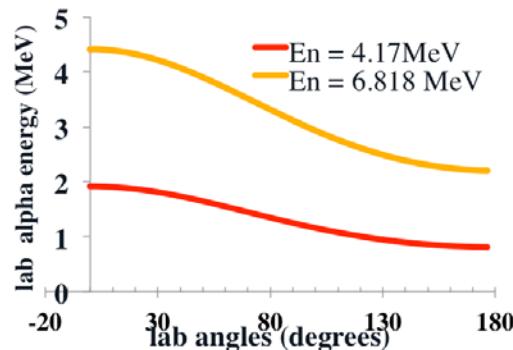


Figure 7. Calculated kinematics of alpha from the $^{16}\text{O}(n,\alpha)$ reaction. For the resonances of 4.17 MeV and 6.818 MeV, the forward angles in LAB system are preferable for detecting alphas due to higher LAB energies than that at the backward angles.

beam intensity and detection efficiency estimation. For (n,α) reactions, the $^6\text{Li}(n,\alpha)$ reaction can be used as a reference reaction. Target characterization still needs to be taken into account for the uncertainty estimation, although this ratio method is used.

As Figure 6 (left) shows multiple resolved resonances, improving the neutron energy resolution, i.e. timing resolution of the cathode signal in the ionization counter is critical. R-matrix analysis in Figure 6 (right) shows larger angular distributions at forward angles, where more DSSDs are positioned to cover larger angles. Based on the kinematics calculations shown Figure 7, it confirms to locate more DSSDs at forward angles for detecting kinematically favored alpha energies. In order to detect alpha above 1.5 MeV, the gas pressure in the ionization counter and the preamplifier parameters are optimized for small pulse-height signals. The gas type is chosen to be P10, because any oxygen contained gas mixture is excluded for potential background. The pressure and the distance between the cathode and anode are set to be 300 Torr. and 12 mm to generate least energy loss and energy straggling from low-energy alphas in the gas volume. For the same reason, the Frisch grid is not utilized and the angle information is taken from the DSSDs, which has annular segmentations. The DSSDs is located in such a way to provide the angular resolution of about one degree.

Since the amount and uniformity of the target material was dominant contribution to systematic uncertainties in the previous measurements, solid oxygen targets were investigated for this experiment.

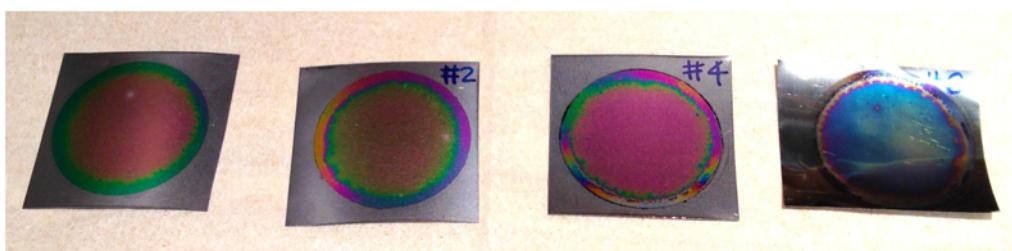


Figure 8. Picture of Ta_2O_5 targets manufactured by the anodizing technique

We have investigated two methods for manufacturing solid oxygen targets. One is the anodizing technique by applying an electric field in ^{16}O -enriched water, via etching oxygen ions onto the backing material (tantalum) surface with relatively small variation in thickness [22]. The target was manufacture by applying 250 V, which corresponds to about 4000 Å in thickness. The pictures of the Ta_2O_5 targets in different thicknesses are shown in Figure 8 and the color on the deposit could be used to identify a specific oxygen thickness. The other method was to evaporate lithium carbonate (Li_2CO_3) powder on a tantalum backing, which can be used as the in-built normalization factor correction for neutron beam intensity. The $^6\text{Li}(\text{n},\alpha)$ reaction cross section is known within 5 % uncertainty up to 1 MeV and about 10 % uncertainty above 1 MeV [23], so it can be used as a reference reaction in order to cancel out the neutron intensity uncertainty and detection efficiency uncertainty, as long as target stoichiometry is well characterized and does not change during the measurement.

For the in-beam background estimate, we plan to measure neutron-induced reactions on a blank tantalum under the same condition as the $^{16}\text{O}(\text{n},\alpha)$ reaction study. A first step is to investigate the realistic background on this reaction due to a backing material in solid target, then we can optimized our setup or different choice of backing materials. We plan to investigate the stoichiometry and the stability of the target before and after neutron irradiation.

As discussed in Section 2, the previous (n,α) measurements [2, 9] discussed the difficulty from the poor neutron energy resolution, since the neutrons were produced from $\text{D}(\text{d},\text{n})^3\text{He}$ or $^3\text{H}(\text{p},\text{n})$ reactions. The combination of the production gas cell and entrance and exit windows could add large energy straggling and broadening. At LANSCE, the neutron energy resolution is deduced from the detection timing resolution of time of flight, therefore the cathode signal is optimized to provide the best timing resolution.

5 Outlook

With the development of this multi-purpose LENZ chamber, we plan to study (n,p) reaction cross sections. Various evaluations used Hauser-Feshbach formalism to calculate neutron-induced charged particle cross sections up to 20 MeV, since experimental data sets lack in this entire energy range. Due to the limited available neutron energies, which were generated from charged-particle accelerators, data sets exist only at specific energies. However, these evaluations show significant discrepancy in the overall shape, but good agreement at around 14 MeV, which is the only energy range for existing data sets on the reaction of $^{77}\text{Se}(\text{n},\text{p})$ [21]. This similar behavior has been found in the comparison of different evaluations on the cross section of $^{75}\text{As}(\text{n},\text{p})$ reaction, which has great interest for nuclear applications. LANSCE is an ideal place to measure the (n,p) cross sections in the broad energy range of 1-20 MeV, therefore we plan to measure the reactions of $^{77}\text{Se}(\text{n},\text{p})$. This effort also can improve understanding of nucleosynthesis in astrophysics via using better predicted reaction rates on neutron-induced charged particle reactions. The survey on the nuclear input sensitivity in p-process nucleosynthesis [24] identified several (n,p) and (n,α) reactions as potential branching points to affect the final p-process abundance. The LENZ instrument at LANSCE can study on these nuclear reactions to impact on understanding of heavy element productions in various stellar environments.

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