Practical considerations for radioactive target fabrication for neutron-induced reaction studies at LANSCE

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Abstract. High quality nuclear reaction data for radionuclides is lacking, which can be best rectified by performing direct measurements of neutron-induced reactions on the target nuclei of interest. Two instruments at the Los Alamos Neutron Science Center, the Low Energy (n,Z) (LENZ) instrument and the Device for Indirect Capture Experiments on Radionuclides (DICER), can perform such measurements on radioactive targets. This manuscript covers the target requirements for such experiments and provides examples of how defects in the targets manifests in the measured data.

1 Introduction

Acquiring high fidelity nuclear reaction data is essential for improving the accuracy of neutron reaction network models, which leads to more reliable stellar nucleosynthesis models and validations in nuclear applications. High quality nuclear reaction data have been obtained primarily for stable nuclei to date. Thus, nuclear data for nearly all radionuclides rely entirely on theoretical data and require experimental data [1]. Direct measurements of neutron-induced reactions on solid targets provide the highest quality of nuclear data [2]. Some instruments, such as the Low Energy (n,Z) (LENZ) instrument and the Device for Indirect Capture Experiments on Radionuclides (DICER), at the Los Alamos Neutron Science CEnter (LANSCE) can be utilized to perform such measurements on radionuclides. Specifically, direct measurements of neutron-induced charged particle reactions and the overall neutron transmission have been performed for experiments on ^{56,59}Ni, ^{44}Ti, and ^{40}K with LENZ [3–5] and ^{88}Zr with DICER [6, 7], respectively. These targets had specific requirements that needed to be met, which included those necessary for any target used in neutron-induced reaction studies [2, 8]. Specifically, they needed to be high in purity while having good uniformity in density, composition, and morphology. How important each requirement is depends on the instrument and measurement method being utilized. Understanding target requirements for these experiments will increase the likelihood that high quality radioactive targets can be fabricated for neutron-induced reaction studies with LENZ and/or DICER.

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2 Device for Indirect Capture Experiments on Radionuclides

2.1 Instrument overview

DICER was designed to address the challenging task of measuring neutron capture (n,γ) cross-sections of short-lived radionuclides. Instead of directly measuring the (n,γ) cross-sections by detecting prompt γ-ray emissions, which would be difficult to perform using short-lived radionuclides due to the high background contributions to the γ-ray spectra, DICER measures the neutron transmission of the target nuclide. From this data, nuclear information including the nuclear level spacings, total resonance widths, and neutron resonance widths for the target nuclide can be directly determined, while the (n,γ) cross-sections can be indirectly and accurately determined through Nuclear Statistical Model calculations [9]. Furthermore, the usage of a binocular collimator and sample holder minimizes the required target sample mass and decreases the measurement time, both of which are advantageous when performing direct transmission measurements with short-lived radionuclides and are an improvement from typical transmission measurement setups [10].

Early work with the DICER instrument utilized enriched solid pucks or disks as targets for benchmarking the instrument [10, 11]. After those successful benchmark tests, radioactive targets were utilized to perform neutron transmission measurements on ^{88}Zr [6, 7] and ^{239}Pu [12]. Both targets in these experiments were liquid targets sealed in a 1.2 mm diameter capillary tube with lead spheres pressed into the ends [7, 12, 13]. Although these experiments were successful at obtaining transmission data on these radionuclides for the first time, the targets were larger than the minimum required for the instrument. To realize the full potential of DICER, targets need to be fabricated as small as a 0.1 mm diameter by 15 mm in length cylinder. These targets will need to have well-defined lengths and be excellent in uniformity and purity.

2.2 Target requirements

2.2.1 Length

It is difficult to make highly enriched targets of relatively short-lived radionuclides. For measurements of these radionuclides to be feasible with DICER, the minimum target size was selected to be 0.1 mm in diameter by 15 mm in length. It may appear that, as long as the target is homogeneous and uniform in density and composition, fabricating a 0.1 mm diameter by 15 mm long target or a slightly larger target would be sufficient. However, imperfections in the target geometry, particularly the length of the target over the beam spot, can lead to significant errors in the measurement if corrections are not made [14]. As such, these effects should be minimized by ensuring DICER targets are well-characterized in geometry.

2.2.2 Uniformity

In addition to well-characterized geometries, DICER targets need to have excellent uniformity in both composition and density. A completely homogeneous target with no air gaps or substantial pores of various sizes is preferred. The inclusion of such void spaces leads to a target that is non-homogeneous in density, which results in inaccurate measurements of the dip depths (as demonstrated by Becker *et al.* [15]).

2.2.3 Purity

Because transmission measurements are examining the total interaction of incident neutrons with the target material, the observed spectrum reflects the total absorption or reflection of neutrons from all nuclides present in the target material. Impurities that interact strongly with neutrons, such as ^{1}H, will increase the background in the measurement and may increase the error in the measurement [16]. Although there are techniques that can be used to determine the isotopic composition of the target material, such as neutron resonance analysis, these techniques are limited in the case of nuclei containing large resonances as the transmission signal can saturate if the target contains enough of these nuclei [17]. Reducing impurities, such as ^{1}H, and using highly enriched material reduces the background and improves the accuracy of these transmission measurements.

3 Low Energy (n,Z) (LENZ) instrument

3.1 Instrument overview

The LENZ instrument was designed to be an improvement of the preceding NZ chamber [18] by allowing for better charged particle detection and larger solid angle coverages of neutron-induced charged particle emitting reactions with small sample sizes [19]. If the target nuclide is a relatively short-lived radionuclide, then a verision of the LENZ instrument, called hotLENZ, can be used. The hotLENZ configuration allows for remote precision alignment of a radioactive target and LENZ silicon detectors to the neutron beam axis. Additionally, an advanced collimation system was developed to reduce beam induced backgrounds while still maximizing the flux over a 6 mm diameter spot comprising of the sample target material [3].

The hotLENZ configuration was used to provide the first direct measurements of (*n*,*p*) reactions on ^{56}Ni and ^{56}Co and confirmed the successful implementation of the advanced collimation system for optimizing the neutron flux through the radioactive target [3]. More experiments have been performed with the standard LENZ configuration to study neutron-induced charged particle reactions, with both stable and longer-lived radioactive samples (*e.g.* ^{40}K, ^{44}Ti, ^{59}Ni)[4, 5, 20–25]. The results from these studies provide examples of how the target quality can impact the measurement accuracy and precision.

3.2 Target requirements

3.2.1 Thickness

Ideal LENZ targets will have enough of the target nuclide to obtain good measurement statistics while being as thin as possible. Thicker targets result in worsened spectroscopic resolution due to energy loss and straggling effects. An example of this can be seen by comparing the neutron-induced charged particle spectra obtained for microjet printed sodium chloride (NaCl) [24] and evaporated silver chloride (AgCl) targets [20] (see Figure 1). Despite both targets being exposed to nearly the same neutron flux, having a similar number of Cl atoms, and being located about the same distance from the DSSD, the thinner evaporated targets resulted in spectra with narrower kinematic bands (better channel resolution) than those obtained with the microjet printed targets, which allow for better background characterization and to reduce the measurement uncertainty. The background from a thicker target backing or inclusion of stable material can be subtracted, as demonstrated by Kuvin *et al.* [22] with a ^{59}Ni sample on a 25 μm thick Pt backing. This ^{59}Ni sample was highly enriched and uniformly deposited as a thin film through electrodeposition, resulting in good spectroscopic resolution to isolate the reaction channels of interest.

3.2.2 Uniformity

If targets are uniform in morphology, then the measurement uncertainty can be minimized. The reasons for this are similar to how imperfections in the length of a DICER target affect the acquired data. If these local differences in target thickness that consistently appear as some pattern that can be well-characterized, then the effects these differences have on the resulting spectrum can be minimized with the application of the appropriate corrections. However, if the variation in local target thickness is great

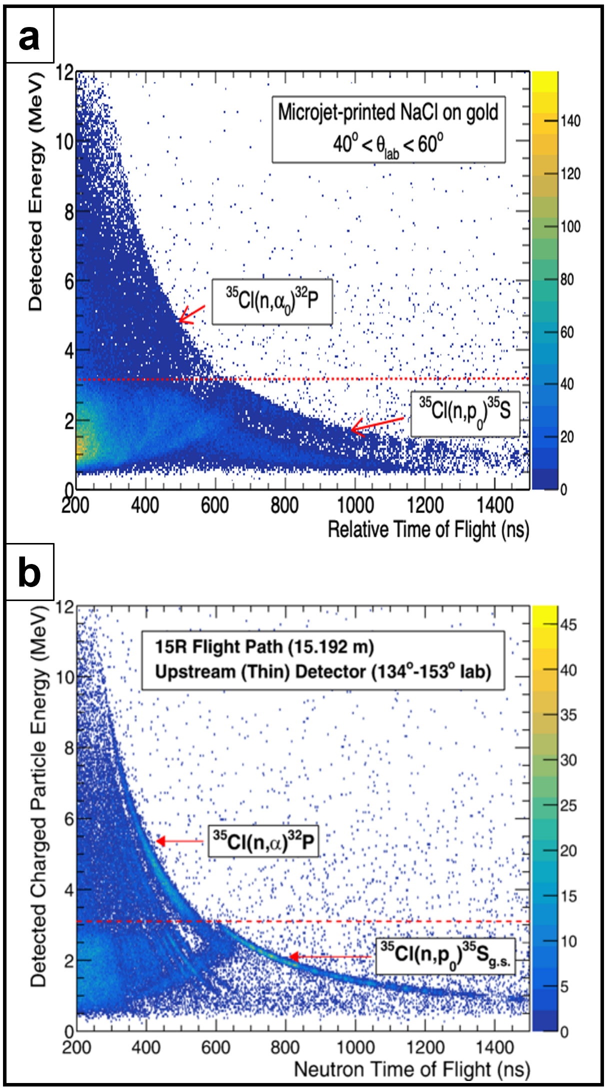

Figure 1. Neutron-induced charged particle spectrum obtained with a downstream DSSD and a 330(30) μg/cm^2 microjet printed NaCl target described in Essenmacher *et al.* [24] (a), and a spectrum obtained with an upstream DSSD and a 631(22) μg/cm^2 thermally evaporated AgCl target (plot reprinted with permission from Hanselman *et al.* [20]) (b).

enough and the neutron beam spot is smaller than the sample target area, then it is possible to obtain results for a different thickness than the average target thickness. Essenmacher *et al.* [24] demonstrate this with microjet printed NaCl and V_2O_5 targets. Meanwhile, Dede *et al.* [25] demonstrate how such local differences in target thickness can be masked to some extent if the beam spot is larger than the sample target area. It is important to note that such variations in thickness still increase the uncertainty in the direct measurements because of the broadening of the kinematic bands in the resulting neutron-induced charged particle spectrum.

3.2.3 Purity

The LENZ instrument measures the outgoing angle, energy, and detection time of the outgoing charged particles produced from neutron-induced charged particle reactions on the sample target. Identifying the target nuclide requires reconstructing the Q-value of the reaction from the detected energy, angle and time-of-flight for the neutron (derived relative to the timing of the LANSCE proton beam) involved in the reaction. These values are characteristic for (n,p) and (n,α) on a specific target nuclide. However, these values for target impurities or backing materials can be very similar to those of the reactions of interest. In these cases, such nuclides need to be removed or minimized during the target manufacturing process.

Additional issues with target impurities can arise if the impurities interact strongly with incident neutrons in the energy range of interest or if the presence of such impurities are difficult to replicate between sample targets. An example of the latter case is shown in Figure 2, where a foil backing of 1 μm thick Ti was used for molecular plating of an Al film onto the Ti surface. The foil had not been light tight tested during the manufacturing process, which meant the molecular plating solvent could seep through the microscopic pores within the foil during the plating process and interact with the copper electrode, causing the target to contain copper contamination. The small amount of copper was not uniformly distributed over the target area and, because the pores are not always present in each piece of foil, were not always observed in Al molecular plated target samples. Although, the Al layers in these samples have good uniformity, the presence of copper in the foil backing would cause complications when performing any neutron-induced charged particle reaction measurements. As such, the fabrication procedure was adjusted to eliminate this copper impurity from the target samples. Similar adjustments that eliminate or minimize impurities during the target development process are always preferred to minimize the uncertainty in direct neutron reaction measurements with the LENZ instrument.

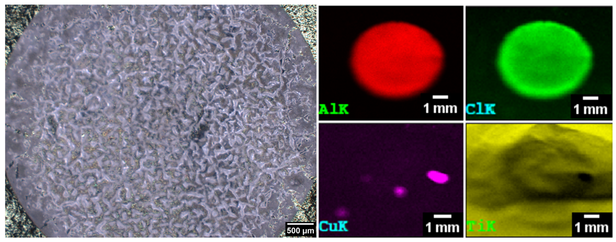

Figure 2. A 2.5x magnification optical image of a molecular plated Al target captured using a Keyence VK-X3100 laser microscope alongside elemental mapping images acquired using an Orbis X-ray flourescence (XRF) analyzer with a 40 keV and 400 A X-ray source.

4 Conclusions

DICER and LENZ/hotLENZ are two instruments available at LANSCE for performing measurements on neutron-induced reactions with radioactive targets. To minimize the uncertainty in these measurements, fabricated targets need to meet certain requirements. For DICER, these requirements are well-characterized in length or areal density, having good uniformity in density and composition, eliminating or minimizing detrimental impurities, and being highly enriched in the target nuclide of interest. These requirements for LENZ include minimizing the target thickness, having good uniformity in target morphology, and eliminating or minimizing impurities that would interfere with or obfuscate the measurement. By producing high quality targets, the nuclear data on target nuclides can be discerned from direct neutron-induced reaction measurements at LANSCE with high fidelity; ultimately leading to more reliable modeling of stellar nucleosynthesis and validations in nuclear applications.

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