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SAND2019-12626
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Printed October 2019

Final 2019 Report of IWO 444118

Development of eXternal Nuclear Reaction Analysis (XNRA) Detection Technique for Quantifying Light Isotope Concentrations

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Abstract

The National Nuclear Security Administration's Tritium Sustainment Program is responsible for the design, development, demonstration, testing, analysis, and characterization of tritium-producing burnable absorber rods (TPBARs) and their components used to produce tritium for the nation's strategic stockpile. The FY19 call for proposals included the specific basic science research topic, "Demonstration and evaluation of advanced characterization methods, particularly for quantifying the concentration of light isotopes (^{1}H, ^{3}H, ^{3}He, and ^{4}He, ^{6}Li and ^{7}Li) in metal or ceramic matrices". Last year the same language appeared in the call for proposals, and a project IWO-389859 was awarded to the Ion Beam Lab (IBL) at Sandia-NM which was successful using Elastic Recoil Detection, but in the future could have resulted in tritium contamination that jeopardized other equally important NNSA projects. An alternative approach using deuterium nuclear reaction analysis was proposed and funded in FY2019 which was also successful and eliminated any possibility of contaminating the Ion Beam Laboratory with tritium, and will be described in this report.

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1. INTRODUCTION

The National Nuclear Security Administration's Tritium Sustainment Program, led by Pacific Northwest National Laboratory (PNNL), is responsible for the design, development, demonstration, testing, analysis, and characterization of tritium-producing burnable absorber rods (TPBARs) and their components used to produce tritium for the nation's strategic stockpile. Since 2006, the program has conducted a series of large-scale in-reactor and ex-reactor experiments designed to quantify irradiation effects on individual components used in TPBARs. While these experiments have provided valuable data that contributed to improvements in TPBAR performance and modeling, the experiments have not been capable of easily measuring the concentrations of all the light isotopes present in TPBAR materials such as the $LiAlO_2$ pellets and zircalloy getters following neutron exposures in the reactor.

In FY19, the tritium science research program is participating with laboratories outside PNNL for the fourth year. One of the funded projects, selected in a competitive peer review process, was submitted by SNL, entitled "Development of eXternal Nuclear Reaction Analysis (XNRA) Detection Technique for Quantifying Light Isotope Concentrations in Irradiated TPBAR Materials." This report details the successful project we developed for this purpose.

2. THE EXTERNAL NUCLEAR REACTION ANALYSIS (XNRA) SYSTEM

The ceramic $LiAlO_2$ pellets in Tritium Producing Burnable Absorber Rods (TPBARs) will contain ^{1}H, ^{3}T, ^{3}He, and ^{4}He produced by nuclear reactions, e.g. the $^{6}Li(n,\alpha)^{3}T$ reaction, in addition to the already present ^{6}Li and ^{7}Li, after being irradiated in the Watts Bar nuclear power plant. Our project was to develop an advanced characterization technique in the Ion Beam Laboratory (IBL) to quantify the concentrations of these light isotopes in irradiated pellets. The IBL has a reputation for developing new and advanced ion beam analysis techniques [1]. One such technique used regularly in the laboratory is Nuclear Reaction Analysis (NRA) applied to components of weapon neutron tubes. This technique works by exposing a material to high energy (MeV range) protons, deuterons or ^{3}He nuclei, subsequently causing light atoms embedded inside the material to undergo nuclear reactions. Protons, alpha particles or gamma rays that are produced are subsequently energy analyzed in a detector.

As mentioned above, we were successful last FY in the development of, a tritium-compatible Heavy Ion Elastic Recoil Detection (HI-ERD) system that was employed in the Limited Security Area of the IBL which succeeded in measuring all the light isotopes in $LiAlO_2$ pellet samples, but it became clear that performing HI-ERD in the IBL would probably result in an unacceptable T contamination of both the HI-ERD endstation and beamline, and possibly up to and including the Tandem Accelerator, in addition to lab areas around the HI-ERD system that includes additional beamline-endstations critical to other NNSA weapon-related measurements. The IBL simply could not take that chance. This is why we proposed and developed an alternative, that can also measure all these light isotopes, except ^{4}He, with virtually no chance of any T contamination to the IBL. It's called eXternal Nuclear Reaction Analysis, XNRA, into a portable tritium-tight chamber holding the target samples and detector.

XNRA is not a new IBA technique in the IBL and we have experience with such in-air analyses since the 1980s [2]. Our development of an external nuclear microbeam system even won an IR-100 (now R&D 100) award in 1987.

A schematic of the XNRA system we developed this year is shown in Figure 1. The MeV energy beam of deuterium

1. exits Pelletron beamline through foil, passes thru air,
2. enters a foil into the T-tight He-filled transportable Pellet Chamber that is a 2-3/4 6-way conflat cross, with all but the target holder flange sealed with Cu gaskets,
3. passes through an annular surface barrier particle detector,
4. hits the pellet at the end of the chamber,
5. interacts with the atoms in the pellet,
6. producing protons and alphas from nuclear reactions,
7. that are detected using the annular detector.

The He gas stops the backscattered D which would provide extremely high count rates.

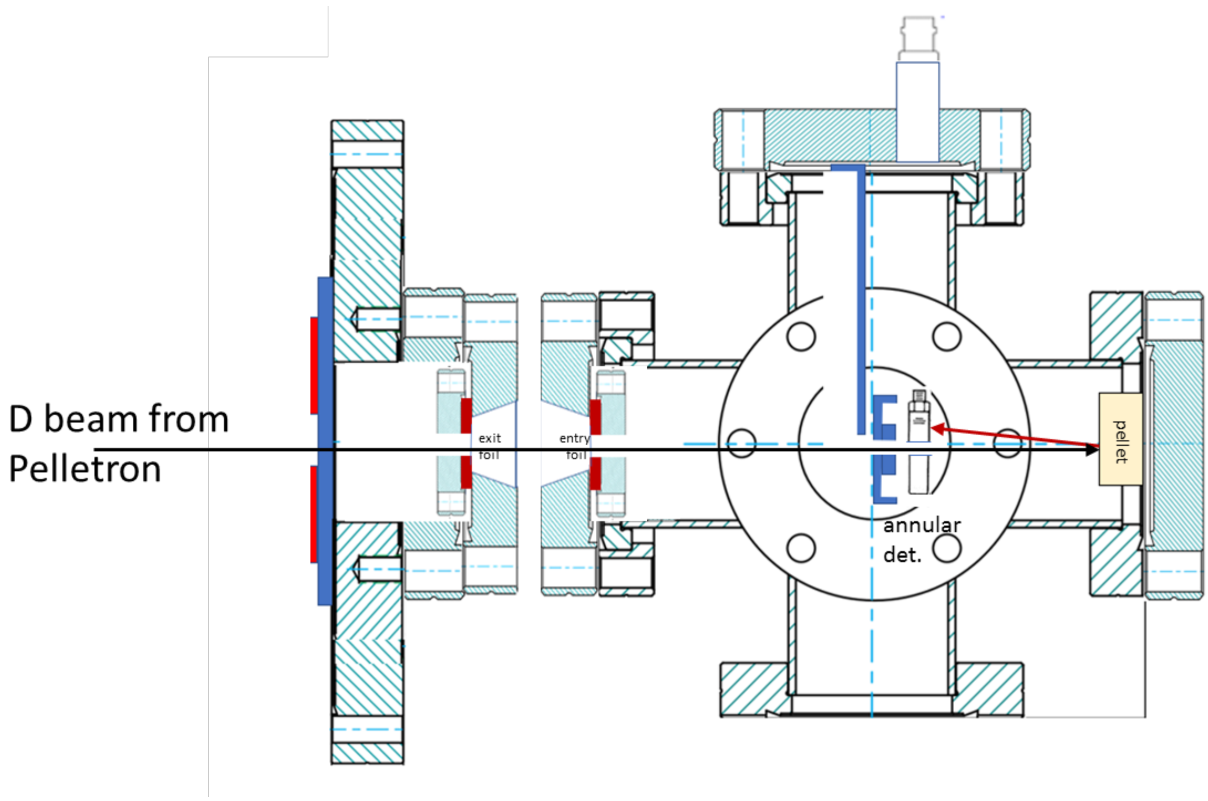

Figure 1 Schematic drawing of the XNRA system developed in the IBL to detect and quantify the composition of light isotopes in TPBAR components.

This XNRA system was utilized to provide an avenue for simultaneously measuring ^{6}Li and ^{7}Li areal densities and in the future ^{3}H, and ^{3}He as a function of exposures and processing of TPBAR $LiAlO_2$ pellets. The XNRA system is not expected to be able to measure high resolution depth concentration profiles of the TPBAR light isotopes because the highly exothermic nuclear reactions produce protons and alpha particles with such high energies making difficult energy based depth profiling. But the possibility of having moderate depth resolution still may be realized at the micron level.

3. THE ANALYSIS OF UNIRRADIATED LIALO$_2$ AND OTHER SAMPLES

The XNRA system was optimized for the proposed experimental parameters. Initially, while the system was being prepared, $LiAlO_2$ pellets enriched with ^6Li that were previously obtained from PNNL were obtained. These unirradiated but ^6Li enriched $LiAlO_2$ pellets were initially measured using the system to determine concentrations of ^6Li and ^7Li.

3.1 The XNRA system

The XNRA system is in the High Radiation Room (HRR) of the IBL where most of the experiments involve the generation of 14 MeV neutrons with the T(d,n) reaction using the HVEE implanter. This has complicated the experiment compared to the HI-ERD we developed last FY which was not in the HRR. The DT neutron experiments supersede our XNRA experiments due to their importance to other time-critical NNSA radiation effects projects. Better coordination of future measurements in the HRR should not be a problem.

A photograph of the XNRA system is shown in Figure 2.

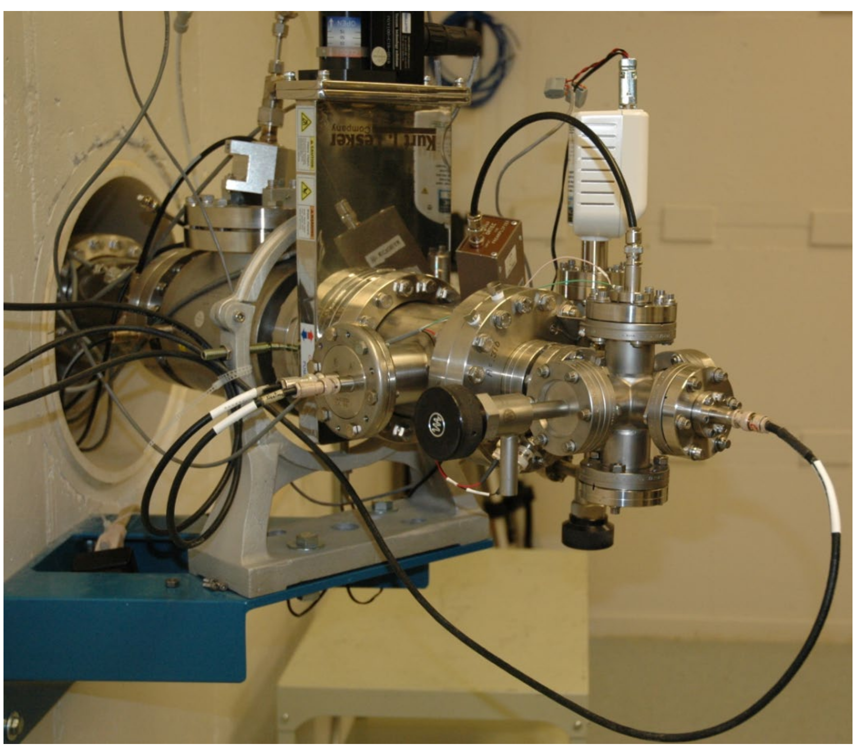

Figure 2 Pellet Chamber attached to eXternal beam line of Pelletron in High Rad Room of the IBL

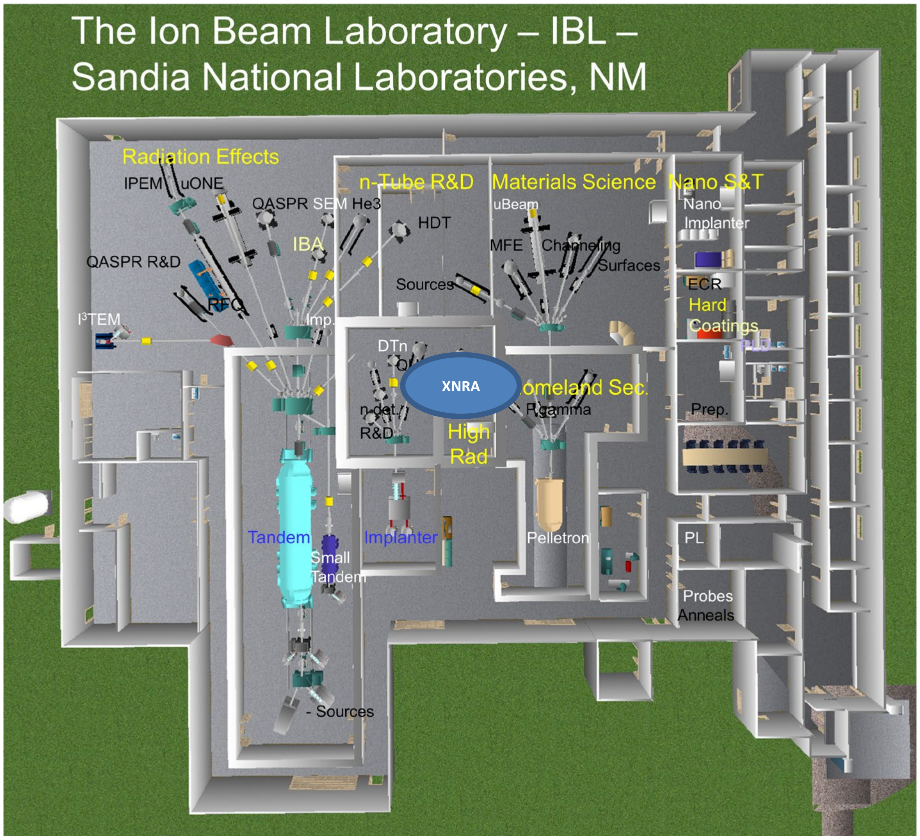

Figure 3 Floor plan of the Tandem Lab in the IBL at SNL. Near the center see the High Radiation Room (HRR) where the XNRA using the Pelletron Accelerator is performed. Also note the Implanter with its beamlines into the HRR where ion implantation and the use of the T(d,n) reaction is used to make fusion neutrons for radiation effects testing.

3.2 XNRA Kinematics and Cross Sections

The most important thing about nuclear reaction analysis experiments is the selection of the incident D energy. This energy must be high enough to produce significant numbers of the reaction particles of the isotopes of interest for n-exposed $LiAlO_2$ pellets, but low enough not to produce nuclear reactions with isotopes not of interest that make particles of energies that interfere with the ones of interest. An Excel program was therefore developed to calculate these secondary particle energies and is shown below in Figure 4.

The program takes account the D beam passing through a 6 micron Al foil into air, an airpath, and the passage through another 6 micron Al foil into the Pellet Chamber filled with an atmosphere of He gas, then 12.7 cm of the He gas to the target, producing in the example shown, the (d,α) reaction with ^{6}Li and then through 4 cm of He gas to an optional foil (not used here) and finally to the detector. In the case shown in the figure the alphas strike the detector with an energy of 9.57 MeV. This was then done for all the relevant isotopes in TPBAR $LiAlO_2$ pellets with the results plotted in Figure 5. See the extended caption of this figure for an explanation of what is being plotted.

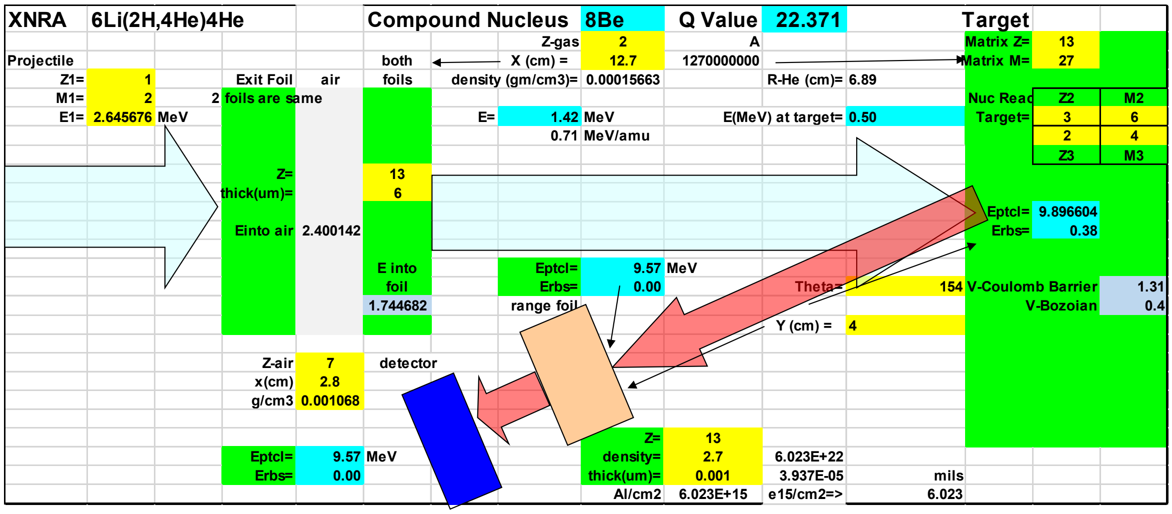

Figure 4 2.65 MeV D XNRA of $LiAlO_2$, looking at the kinematics of the $^6Li(d,\alpha)$ nuclear reaction when the deuterons strike the sample with an energy of 0.5 MeV.

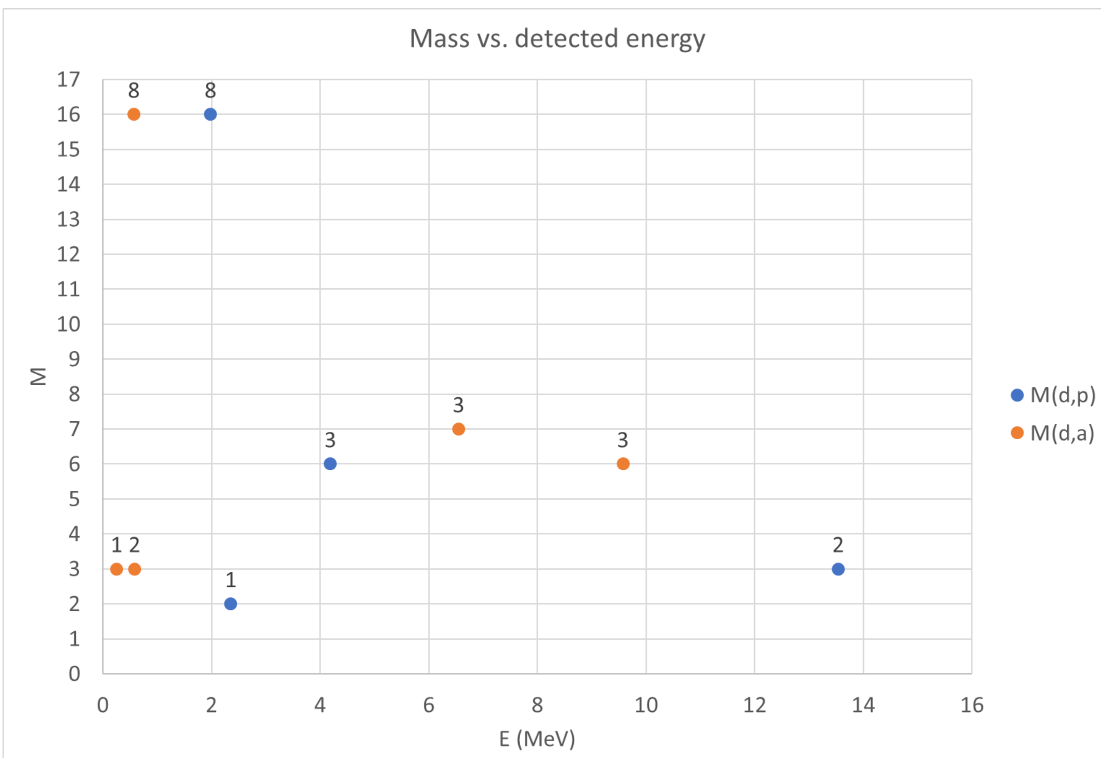

Figure 5 Excel calculation of the detected alpha and proton energies produced by all of the ground state nuclear reactions of 0.5 MeV deuterons on the isotopes than can be in the $LiAlO_2$ pellets after neutron exposure. The blue dots are for (d,p) reactions while the red are for (d,α) reactions. The data labels associated with each dot are the atomic numbers of the isotope in the target, and the y axis indicates the atomic weights. For example the point calculated in Fig. 4 is for the $^6Li(d,\alpha)$ reaction and the point is therefore red, labeled 3, and plotted at M=6 on the y axis, and at 9.57 MeV on the x axis.

Figure 5 shows that for the highly exothermic reactions with ^{6}Li, ^{7}Li, ^{3}He and ^{2}D that the energy separation will be very good and their identification easy. On the other hand, separation of the ^{3}T(d,α), ^{3}He(d,α) from the nuclear reactions of deuterons with ^{16}O could be problematic. This issue can potentially be solved by having the deuterium's energy high enough to create high count rates for the isotopes of interest, but low enough to reduce the count rates for the ^{16}O nuclear reactions. In Figure 6 we plot the cross sections for these nuclear reactions to the ground states of the residual nuclei as a function of incident deuterium energy. This data was found in the Ion Beam Analysis Nuclear Data Library (IBANDL) maintained by the IAEA [3].

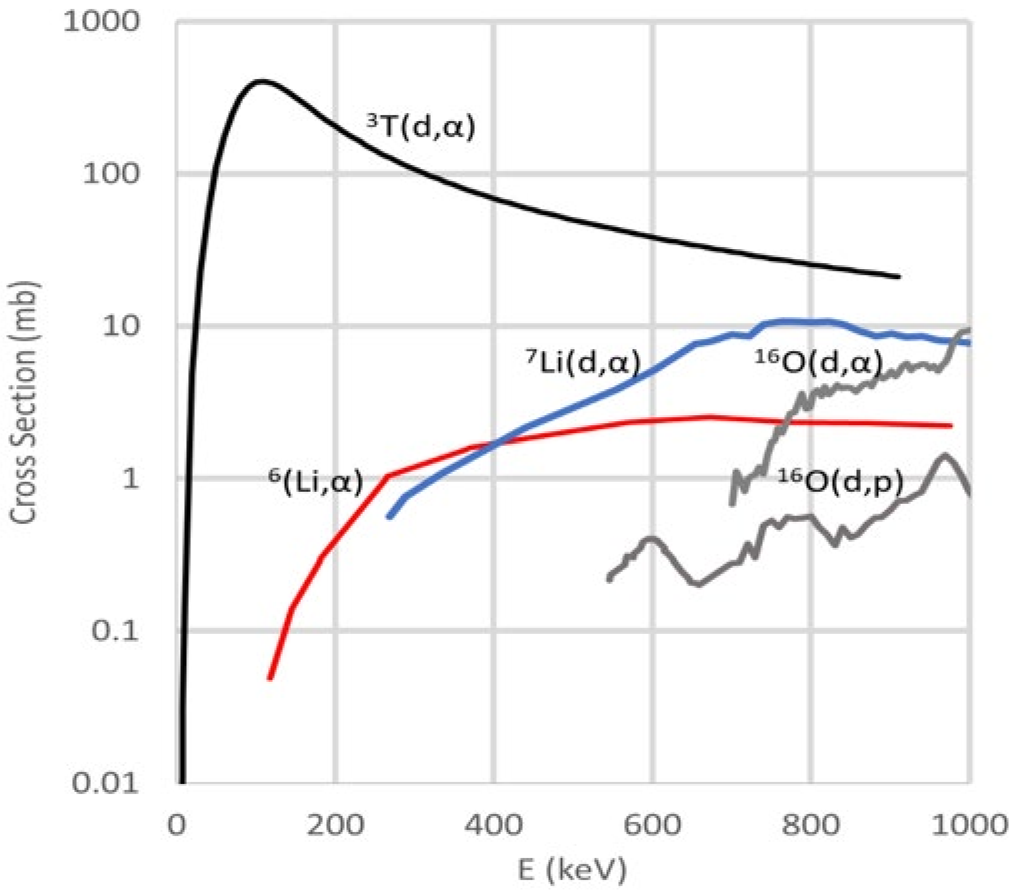

Figure 6 Selected nuclear reaction cross sections for X(d,y) for detection angles between 150-155° and to energies up to 1 MeV. 0.5 MeV D has high cross section (XS) for T (and ^{3}He not plotted), fairly high XSs for ^{6}Li and ^{7}Li, but very low XSs for ^{16}O, which is not of interest and would provide backgrounds to the T and ^{3}He signals.

3.3 Theoretical Calculations of the Spectrum

The deuterium nuclear reaction cross sections above were then used with the SIMNRA 7.1 program developed at the Max-Planck-Institut fur Plasmaphysik in Garching, Germany by Matej Mayer [4]. The annular detector had in ID of 5mm and an OD of 13mm. It was place 4cm away from the targets. This resulted in a scattering angle of ~ 175° and a solid angle of .071 sr. It is important to note here that the cross sections used were at angles from 150-155 because these were the best experimental measurements we could find in the literature. There would therefore be some difference between our experimental results and the theoretical calculations shown below in Figure 7.

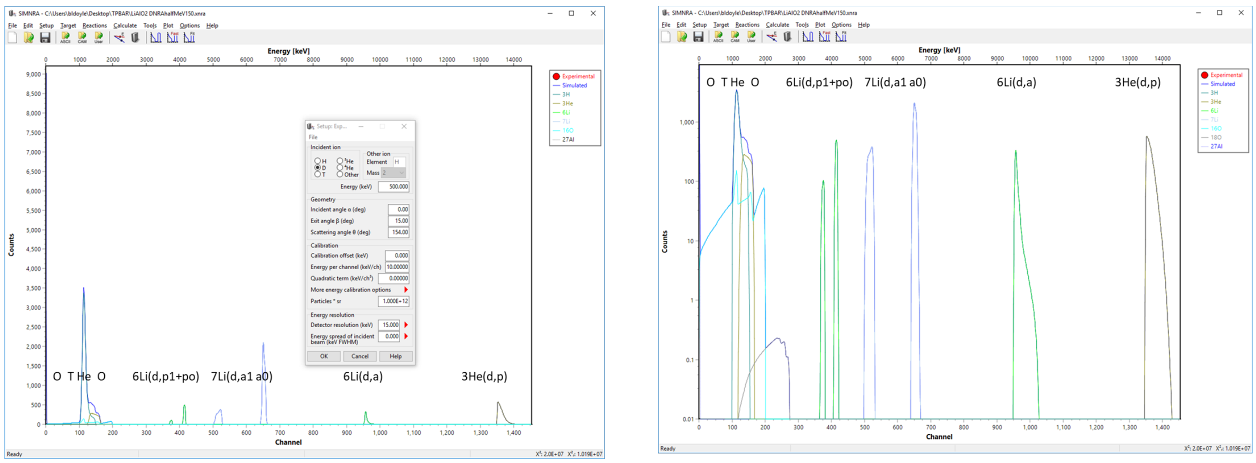

Figure 7 SIMNRA calculations of the XNRA spectrum we expect for having 0.5 MeV deuterium strike a $LiAlO_2$ pellet enriched in ^6Li to 30 atomic % and with 1% ^3T and ^3He. The figure on the left is linear, while that on the right is semi-log.

This calculation was done with 10^{12} deuterium particles – sr (the fluence unit used by SIMNRA), and if we assume a beam current of 1nA, this spectrum would be collected in 0.6 hours. The anticipated statistics for the XNRA in this case would seem adequate.

4. INITIAL RESULTS

The first XNRA run was held in September 2019 after all the safety paperwork was completed, i.e. a Technical Work Document (TWD), a Preliminary Hazard Screening (PHS), and a Pressure Safety Data Package (PSDP), and a Readiness Review (RR) was held with staff from the ES&H and Radiation Protection organizations and upper management of the IBL. The first beam of deuterium was selected to be 2.8 MeV, which was considerably higher than the calculations done at 2.65 MeV. There were several reasons for this:

1. The accelerator settings for a focused beam into the XNRA system already existed
2. The higher energy would provide a worst-case study of the neutron radiation around the Pellet Chamber.
3. The signals from ^{16}O and perhaps even ^{27}Al would possibly be collected to assess the magnitude of future detected particle energy interferences caused by nuclear reactions with these isotopes.
4. And the idea was that subsequent runs would start with this energy and march down to lower energies monitoring (hopefully) a high signal strength of the nuclear reactions of the isotopes of interest, i.e. mainly ^3T, ^6Li and ^7Li, and rapidly decreasing signals from ^{16}O.

A unirradiated $LiAlO_2$ pellet enriched with 25 atomic% ^6Li (natural abundance of ^6Li is only 7.5%) was inserted in the Pellet Chamber where it was mounted on a structure that was insulated so the current of the deuterium beam could be measured. The chamber was then purged with one atmosphere of ^4He gas, closed up and mounted on the Pelletron beam line "L1" that goes into the High Radiation Room of the IBL.

The beam had a positive current of 2.5μA in a Faraday cup just upstream of the Pellet Chamber, but a negative current on the pellet sample was measured. This was unanticipated and most likely due to a large secondary electron current made by the deuterium as it passed through the 1 atmosphere of He gas. This is a problem that will need to be solved in the future because very little of the beam measured on the Faraday cup actually gets to the pellet target. This is because of the small 1.6mm ID of a shield and tube that was used in this experiment to both keep deuterons from penetrating the annular detector from the back (i.e. upstream side), and minimize the scattering of the deuterons from the He gas back into the detector's front side. If we assume the beam area on the Faraday cup was 100 mm^2, then this current would be reduced to 63nA, and probably a lot less than that due to small angle scattering in the He gas. So for now, we estimate the deuterium beam current on target to be ~10nA.

An RCT monitored the dose rate of neutrons being made during this first experiment to be >100mr at 100cm, and therefore the HRR had to be posted as a High Radiation Area and locked. This was not really a great problem because the HRR is usually a High Radiation Area anyway due to the production of 14 MeV DT neutrons using the implanter.

One of the first spectra taken on the unirradiated but enriched $LiAlO_2$ pellet is shown in Figure 8 below. The energy scale was calibrated using a ^{241}Am source that provided 5.4 MeV alpha particles to the detector in a separate experiment. Spectral peaks from the $^7Li(d,\alpha)$ and $^6Li(d,\alpha)$ are clearly identified, as are two other lower energy peaks or edges from ^{27}Al and ^{16}O, but these are only tentatively identified. Again, this was an unirradiated pellet so no nuclear reactions from ^3T would be seen. The counts stop in the low energy region of this spectrum due to the setting of a lower level discriminator (LLD) on the analog-to-digital convertor (ADC) used in the multichannel analysis (MCA) system. Very high count rates existed below this LLD that were due to back scattering of the deuterons by the He gas. This is another problem that will need to be solved in the future and is also discussed below.

A second XNRA experiment was performed replacing the $LiAlO_2$ sample with $LiNbO_3$ that had the ^6Li natural abundance of 7.5%. The spectrum resulting from that measurement is shown in Figure 9. Comparing Figures 8 and 9 it is clear that the signals from the $^6Li(d,\alpha)$ reaction decreases for the $LiNbO_3$, while the signals from the $^7Li(d,\alpha)$ reaction increases, as expected.

It can be shown that a simple thin-target yield analysis that uses the ratio of the areas of the ^6Li peak to the ^7Li peak for the $LiNbO_3$ of known 7.5 at.% ^6Li abundance for calibration, that the ratio of these peaks for the enriched $LiAlO_2$ pellet yields an experimental measurement of 23.4at.% ^6Li enrichment. This is very close to the 25% enrichment that this pellet was known to have.

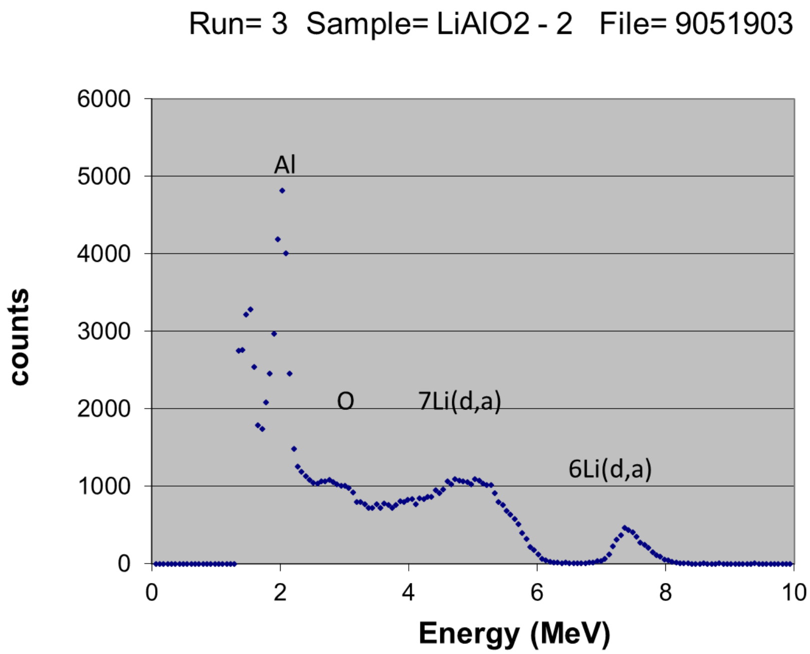

Figure 8 Spectrum of nuclear reaction products produced when 2.8 MeV D+ is placed into the XNRA system and onto a $LiAlO_2$ target. The energy of the deuterons is calculated to be 0.87 MeV on the 25% ^{6}Li enriched $LiAlO_2$ pellet.

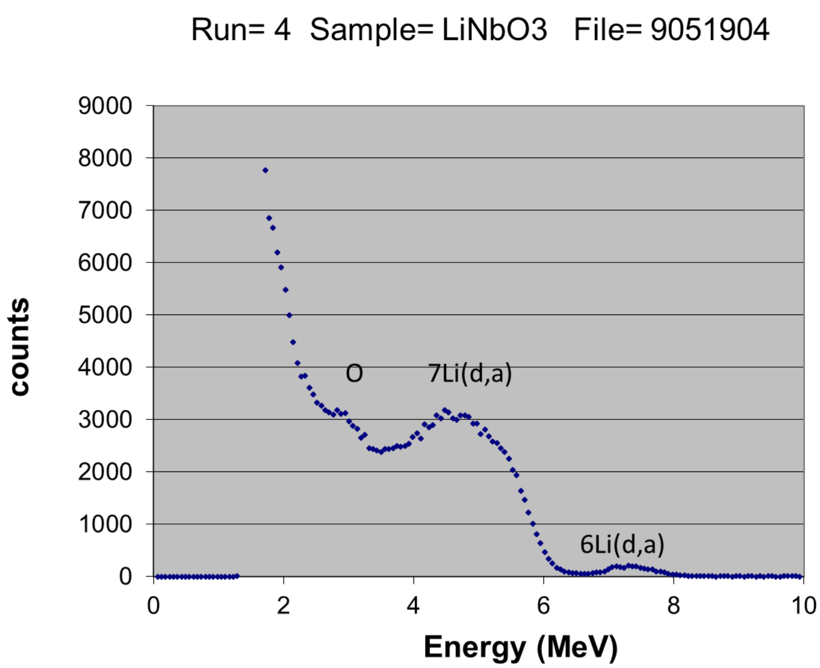

Figure 9 Spectrum of nuclear reaction products produced when 2.8 MeV D+ is placed into the XNRA system and onto a $LiNbO_3$ target. The ^{6}Li concentration in this case is at natural abundance of 7.5 atomic%.

5. DISCUSSION AND CONCLUSIONS

XNRA using (d,p) and (d,α) nuclear reactions will clearly work to measure the concentration of ^{6}Li and ^{7}Li isotopes in $LiAlO_2$ pellets exposed to neutrons at the Watts-Barr reactors in Tennessee, and released and collected at SRNL. By combining this information with the chemical determination of ^{6}Li enrichment prior to neutron exposures in the reactors, the burn-up fraction of the ^{6}Li through the $^{6}Li(n,\alpha)^{3}T$ reaction can be determined. Using a thin-target yield analysis of the $^{6}Li(d,\alpha)$ and $^{7}Li(d,\alpha)$ peaks, the initial enrichment of a $LiAlO_2$ pellet was found to be 23.4 at.%, While this is a very simplified measurement of enrichment, it gives confidence that when a true thick-target yield analysis is developed that the XNRA should be an excellent way to measure ^{6}Li burn-up when analyzing irradiated $LiAlO_2$ pellets.

The eventual measurement of tritium, ^{3}T, concentration may also be possible, if the spectral interferences of deuterium nuclear reactions with ^{16}O can be minimized, or even eliminated by using deuterons with such low energy that these cross sections nearly vanish. This can only be determined by future experiments on irradiated pellets that contain ^{3}T. We will also need to find a way to prevent the backscattering of deuterium from He into the detector which also interferes with the signals from the $^{3}T(d,\alpha)$ reaction.

While we didn't actually get to analyze irradiated pellets this FY, there will be virtually no chance of any T contamination to the IBL by using XNRA in the Pellet Chamber. This is because the Pellet Chamber is "tritium-tight" because of its design. The only chance of ^{3}T escaping from the Pellet Chamber is breakage of the albeit thin (6μm) Al window through which the MeV-energy deuterons are admitted. The IBL has had over 30 years of experience using even thinner Al windows (<1μm) without rupture, leading to confidence that this would be unlikely with the current Pellet Chamber while performing XNRA. Nevertheless, the TWD for all XNRA experiments requires a portable T Contamination Air Monitor (CAM) be positioned near the Pellet Chamber during the analysis of irradiated pellets that contain T.

A few problems with XNRA were found this year, but we think they can be fixed with minor modifications of the Pellet Chamber. We couldn't measure D current on the pellet due to high negative current caused by the generation of delta electrons in the He gas. We plan to bias the pellet at negative HV, or to identify an analysis approach that uses the ^{7}Li peak yield to normalize $LiAlO_2$ analyses. The use of H_2 gas instead of He in the Pellet Chamber may also be considered. Another problem mentioned above was that the D scattered from He gas into detector at low energies that would interfere with the T signal. To address this the tube that is inserted through the annular detector will be lengthened to go almost all the way to the pellet. D will still backscatter from the He gas between the end of the tube and the target, but the ~4cm of He gas between where this backscattering occurs and the detector will act as a filter, ranging out this high intensity background signal. The alphas from T(d,α) may still have energies too low to be detected, and require a modification of the Pellet Chamber by adding another detector at a smaller detection angle, such as ~135°. All of these modifications should be easily accomplished.

An important part of the optimization of XNRA when these problems are resolved will be to determine the optimum energy that the D needs to have when it strikes the pellet. This optimization will involve scanning the D energy down to get just enough signal for the Li isotopes, while eliminating the low energy signals from the oxygen and Al that interfere with the alpha particles from the T(d,α) reaction. The peaks in the spectra (see Figures 7 and 8) should also sharpen because of the rapidly reducing Li cross sections with energy and depth.

All of this is in our proposal to the Tritium Science Program in FY19.

6. ACKNOWLEGEMENTS

While the XNRA idea was one of the author's (BD), the design and construction of the Pellet Chamber and the modification of the beamline of the Pelletron into the High Rad Room was skillfully performed by Stuart Van Deusen (now retired) and George Burns. These two remarkable technologists also made the modifications to the Pelletron to accelerate deuterium beams and to bring a beam into air, both of which had not been done using the large accelerators in the IBL since the 1980's. MeV-energy deuterium beams can make a lot of radiation when the neutron is stripped off, and an external, in-air, beam is extremely dangerous. We also therefore acknowledge Diana Case, Walen Mickey and Steven Allen of Sandia's Radiation Protection and ES&H organizations for advising us during the planning stage of this project, the generation of safety documents, and the monitoring of the radiation produced in the HRR during the first use of XNRA.

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