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1. Introduction

Protection and control of nuclear fuels is paramount for nuclear security and safeguards; therefore, it is important to develop fast and robust controlling mechanisms to ensure the safety of nuclear fuels. Through both passive- and active-interrogation methods we can use fast-neutron detection to perform real-time measurements of fission neutrons for process monitoring. Active interrogation allows us to use different ranges of incident neutron energy to probe for different isotopes of uranium. With fast-neutron detectors, such as organic liquid scintillation detectors, we can detect the induced-fission neutrons and photons and work towards quantifying a sample's mass and enrichment.

A. Nuclear Safeguards

In order for the International Atomic Energy Agency to verify treaty agreements with countries utilizing nuclear technologies for peaceful and productive uses, instrumentation must exist to measure and confirm a country's nuclear material declarations. Future developments in nuclear safeguards' instruments include the use of different neutron detectors. Traditional systems rely on thermal neutron detection, while there are many benefits to using detectors that can measure fast neutrons directly from fission. The primary benefit used in this work is the preservation of the fission neutron's timing for detection in a fast neutron detector. Organic liquid scintillators are the subject for this study as they allow the discrimination between photons and neutrons measured from fissile material and they have the excellent timing properties that are required by this specific form of data analysis.

B. Active-Interrogation of Fissile Materials

Materials of interest in the field of nuclear safeguards are fissile materials that help provide power across the world, but can also be used illicitly in nuclear weapons. Methods of verifying the peaceful use of these materials rely on measuring the presence of fissile material and confirming that no significant quantities have been diverted. When it comes to measuring plutonium, the material's spontaneous fission probability is quite high allowing passive measurements for material characterization. When working to quantify uranium, the problem is less straight forward, considering the spontaneous fission cross-section of all uranium isotopes is quite low, and we must therefore rely on measuring induced fission. As a result, active-interrogation techniques are required for safeguarding nuclear fuels containing only uranium, as is common in many nuclear facilities around the world.

C. Measurement Campaign for Characterizing Uranium-Oxides with Liquid Scintillators

Using the simulation tool MCNPX-PoliMi [1], a system was designed to measure induced-fission neutrons from U-235 and U-238. Measurements were then performed in the summer of 2011 at the Joint Research Centre (JRC) in Ispra, Italy. Low-enriched uranium (LEU) samples were interrogated and induced fission neutrons were measured to characterize the samples in terms of its uranium mass and enrichment. The purpose of the measurement campaign was to investigate the potential applicability of using organic liquid scintillators with active interrogation techniques to characterize uranium containing materials. Additionally, MCNPX-PoliMi simulation results will be compared to the measured trends to validate the MCNPX-PoliMi code when used for active-interrogation simulations.

2. Low-Enriched Uranium (LEU) Samples

Three well-known LEU samples were available to measure at the JRC. Table 1 outlines the variation of these samples in terms of their uranium mass and enrichment. Through the use of active interrogation, we see the differences in uranium mass and U-235 enrichment by inducing fission in these three materials. The neutron-induced fission cross-sections for U-235 and U-238 are shown in Fig. 1 [2]. Based on these cross-sections; a varying induced fission response is seen by probing the three LEU samples separately with both slow and fast neutrons.

Table 1. Material specifications for the three LEU samples studied at the JRC.

Sample	Uranium Mass [g]	Uranium-235 Mass [g]	Enrichment [%]
LEU-1	1684.18	16.84	1
LEU-2	2363.76	73.28	3.1
LEU-3	2363.76	118.19	5

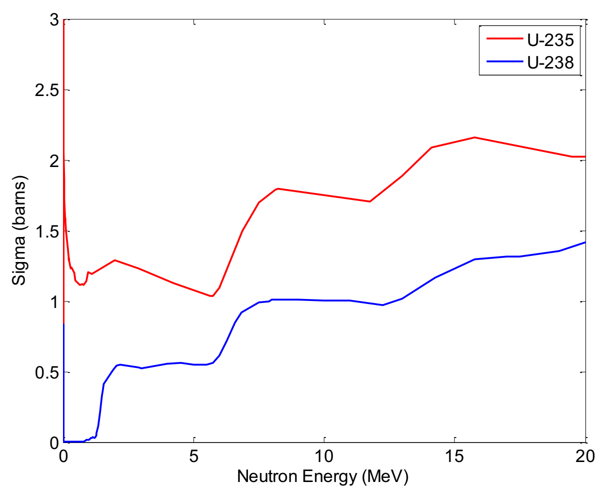


Figure 1. Neutron-induced fission cross-sections for U-235 and U-238.The U-235 cross-section increases all the way above 30,000 barns at thermal neutron energies while the U-238 cross-section is less than 1 barn.

3. MCNPX-PoliMi Active-Interrogation Simulations

Using MCNPX-PoliMi, a system was designed to measure induced-fission neutrons from U-235 and U-238. The system needed to be defined in a way to accommodate the use of a deuterium-tritium fusion (DT) neutron generator for the inducing of fission in the uranium. The liquid scintillators then had to measure the emitted fission neutrons while minimizing the measurement of transmitted and scattered DT neutrons. As shown in Fig. 1, the DT neutrons (at 14.1 MeV) will induce fission in both U-235 and U-238, providing information on the overall amount of uranium present. To learn about the enrichment of the uranium, we must probe the source at very low neutron energies to study only the U-235 presence in the LEU. To do this, a high-density polyethylene moderated Am-Li radionuclide source was used as an additional interrogation technique.

A. Neutron Interactions in Low-Enriched Uranium

MCNPX-PoliMi output includes a detailed history of all of the interactions that happen within a volume of interest, including all of the histories of subsequent particles that are created. By simulating active-interrogation cases and specifying the LEU sample as the volume of interest, we can gauge the usefulness of different active sources in learning the information that we are seeking. Fig. 2 depicts the types of interaction a DT neutron source and an Am-Li neutron source induce within the three LEU samples that were measured.

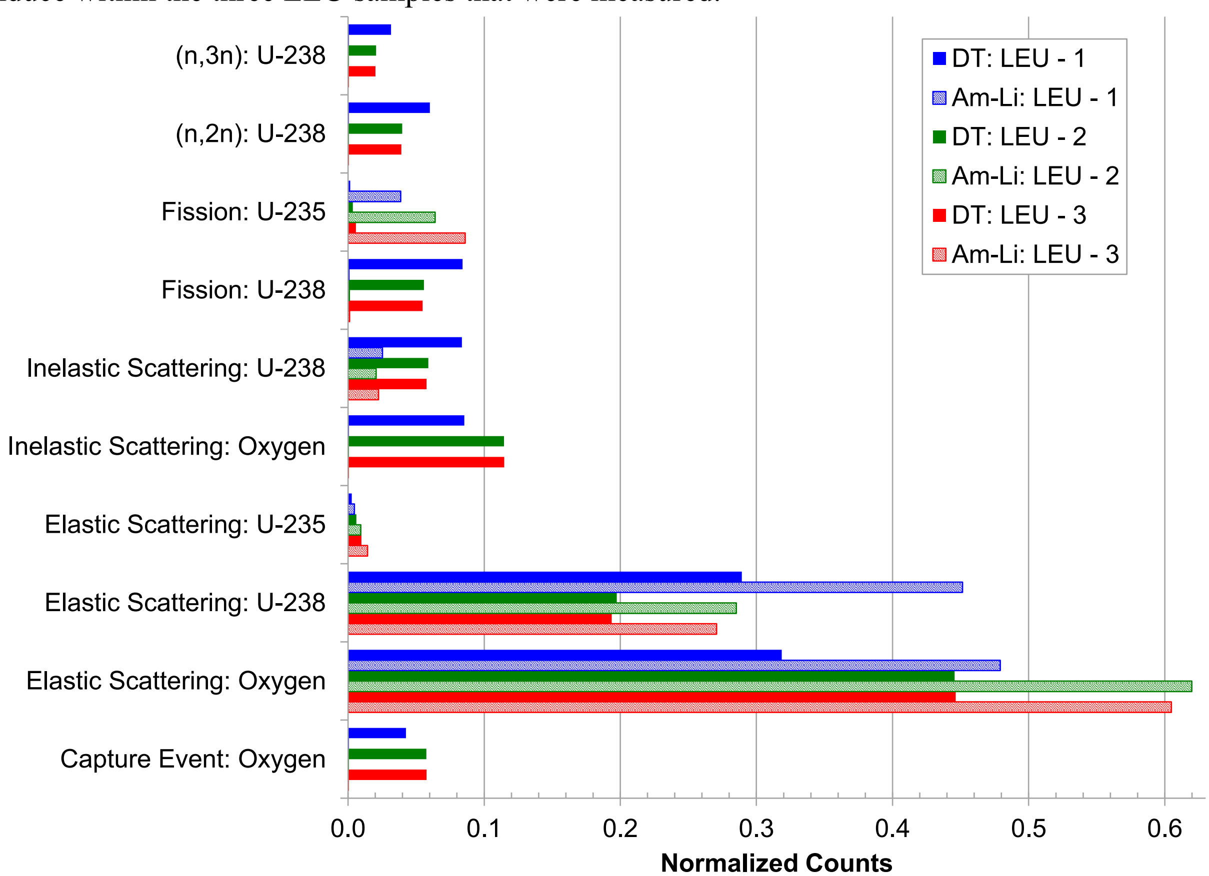
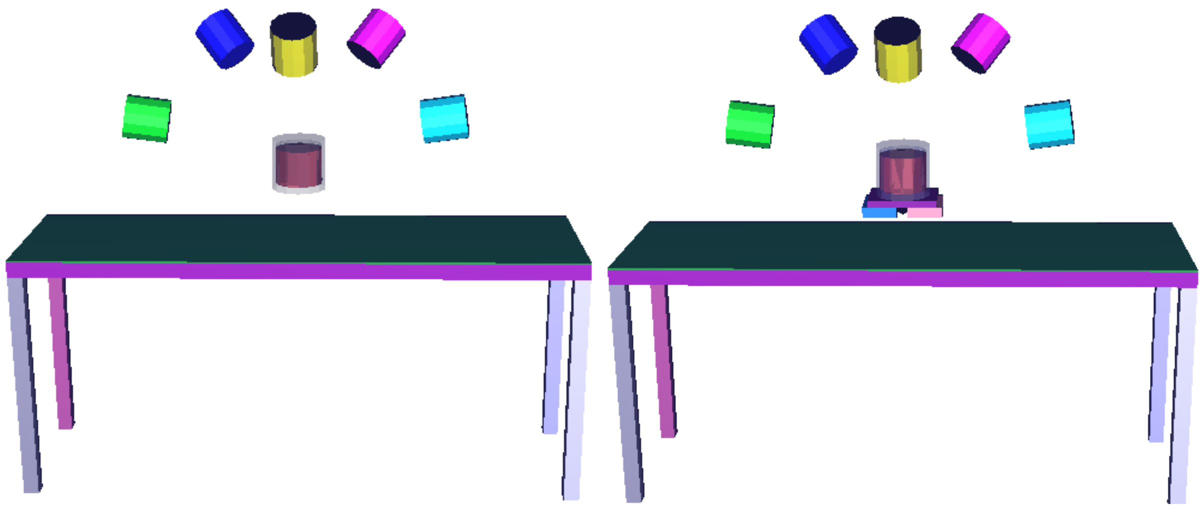


Figure 2. Simulated neutron interaction probability from the three interrogated LEU samples.

B. *Experimental Configuration Models for Mass and Enrichment Studies*

The generator emits neutrons in a time tagged cone at the side of the LEU canister. Therefore, the best configuration to place the detectors, where they will have the least opportunity to be affected by neutrons from the DT generator, was directly above the LEU sample. Five detectors were used to maximize the measurement statistics. Figure 3a shows the MCNPX-PoliMi model for the DT interrogation case. For the Am-Li interrogation case, the radionuclide source was surrounded by polyethylene moderator and placed under the LEU sample in order to minimize the direct measurement of Am-Li neutrons and primarily measure photons and neutrons that are created in the LEU from the incident Am-Li particles, as shown in Figure 3b.



(a) (b)

Figure 3. MCNPX-PoliMi model of the five liquid scintillators measuring the LEU samples for the DT interrogation case (a) and the moderated Am-Li interrogation case (b).

When using the associated particle DT generator, a collimated alpha-detector provides a signal for when a DT event is emitting a neutron in the direction of our LEU sample. By using this signal as a trigger, a time-of-flight (TOF) measurement can be performed, measuring the arrival time of particles created in the LEU sample. The distance between the LEU sample and the five detectors (~35 cm) must be chosen to provide separation between the arrival of the photons and the arrival of the fast neutrons. The TOF simulation results showing this separation are included in Fig. 4a. The photon signal can be eliminated and we can focus on the change in the neutron TOF curves for the three LEU samples. Fig. 2 showed that the mass of the LEU sample would trend with the amount of induced fission events, (n,2n), and (n,3n) events. Therefore, the integrals of these neutron TOF curves should provide information on the sample mass, shown in Fig. 4b. For the study of the U-235 enrichment, Fig. 2 showed that the moderated Am-Li neutrons (average energy of ~0.4 MeV) will induce fission primarily in the U-235. The U-235 fission neutrons' TOF (shown in Fig. 4c) will be measured in the liquid scintillators by triggering on the photons produced during the nuclear interactions. The relationship between the enrichment and these three curves is shown in Fig. 4d.

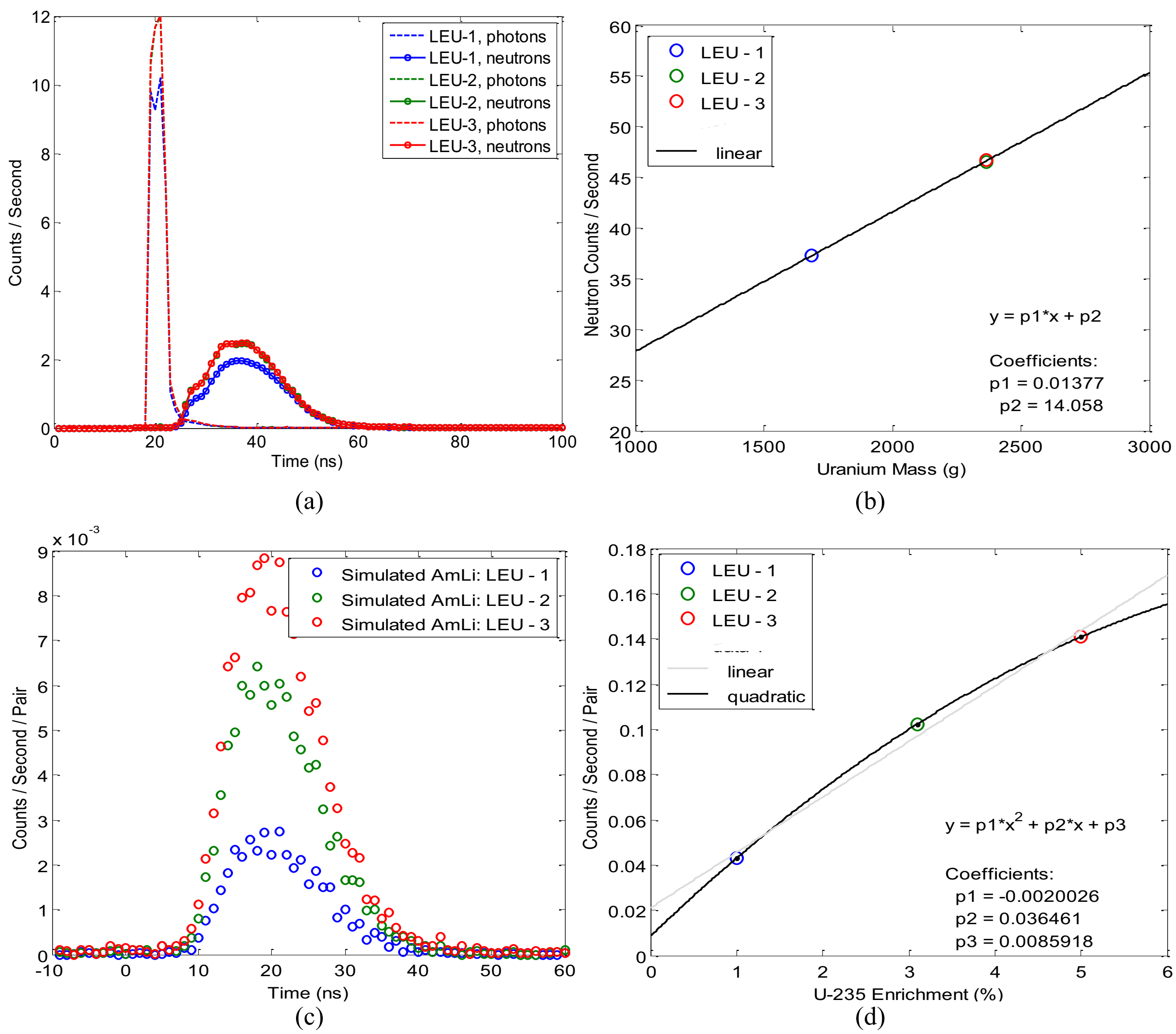


Figure 4. a) Simulated photon and neutron TOF curves for time-tagged DT interrogation of the three LEU samples, b) trend of the total neutron counts with uranium mass, c) simulated photon triggered neutron TOF curves for the moderated Am-Li configurations, and d) trend of the total photon-neutron correlations with U-235 enrichment.

4. Active-Interrogation Experimental Campaign

A. Experimental Configuration and Data Acquisition and Analysis

Fissions were induced with an associated particle D-T generator and a moderated radionuclide Am-Li source. The fission neutrons, as well as neutrons from (n, 2n) and (n, 3n) reactions, were measured with five 12.7x12.7 cm^3 EJ-309 organic liquid scintillators. The D-T neutron generator was available as part of a measurement campaign in place by Padova University. Fig. 5 displays the described measurement configuration.

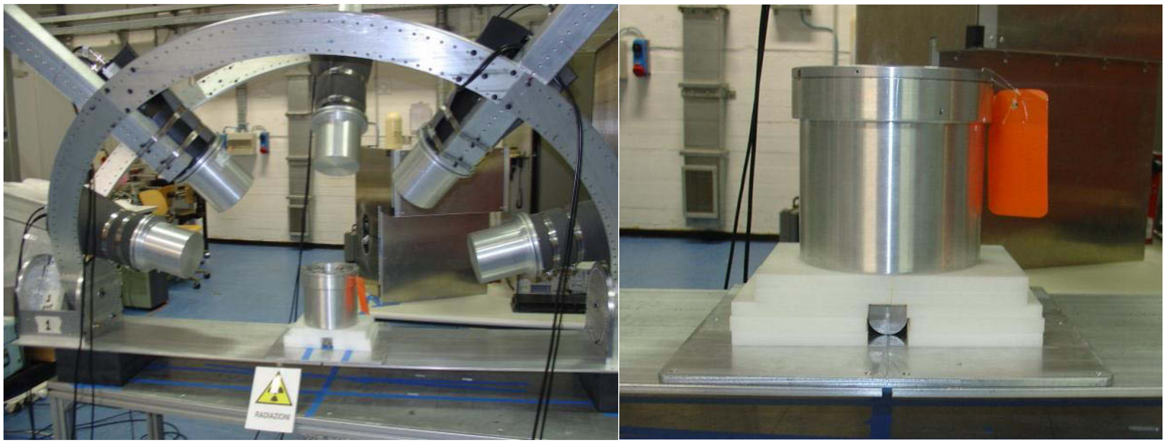


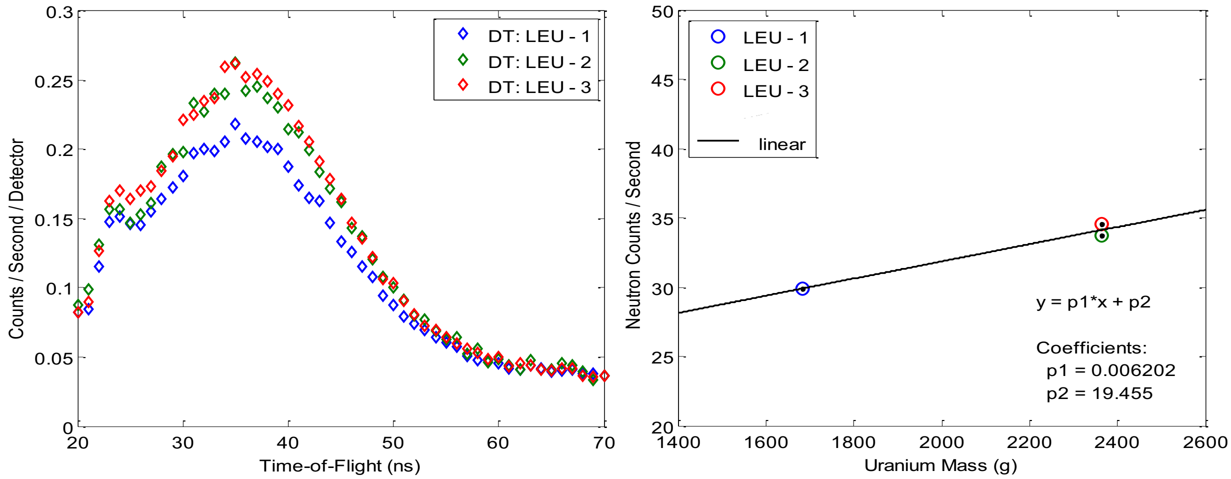
Figure 5. The five-detector geometry positions the liquid scintillator faces at approximately 35 cm from the top of the LEU canister. The associated particle tagged DT neutrons are emitted in the direction coming out of the page from the red dot placed on the picture. Also shown is the moderated Am-Li source placed under the LEU canister.

The measurement and data-acquisition systems were developed at the University of Michigan utilizing a CAEN V1720 digitizer and pulse-shape discrimination (PSD) algorithms to differentiate neutron and photon detections. The CAEN digitizer has eight channels, six of which were used: one for the DT generator's associated alpha detector and the remaining five for the liquid scintillators). The three LEU samples of varying mass and enrichment, defined in Table 1, were interrogated separately with the high energy and low energy neutron sources. Acquired time-of-flight curves were then analyzed to draw relationships between detected neutrons and sample mass and enrichment.

The PSD algorithm, applied above a 0.07 MeVee threshold (~0.7 MeV neutron energy), is important to isolate the neutron signal that comes from the induced fission, (n,2n), and (n,3n) neutrons. In the DT interrogation case it allows the removal of photon accidentals in the neutron TOF distribution. In the Am-Li interrogation case, PSD is more important as it allows us to identify the photon-neutron correlations that are studied here as a pseudo-TOF method.

B. Experimental Results

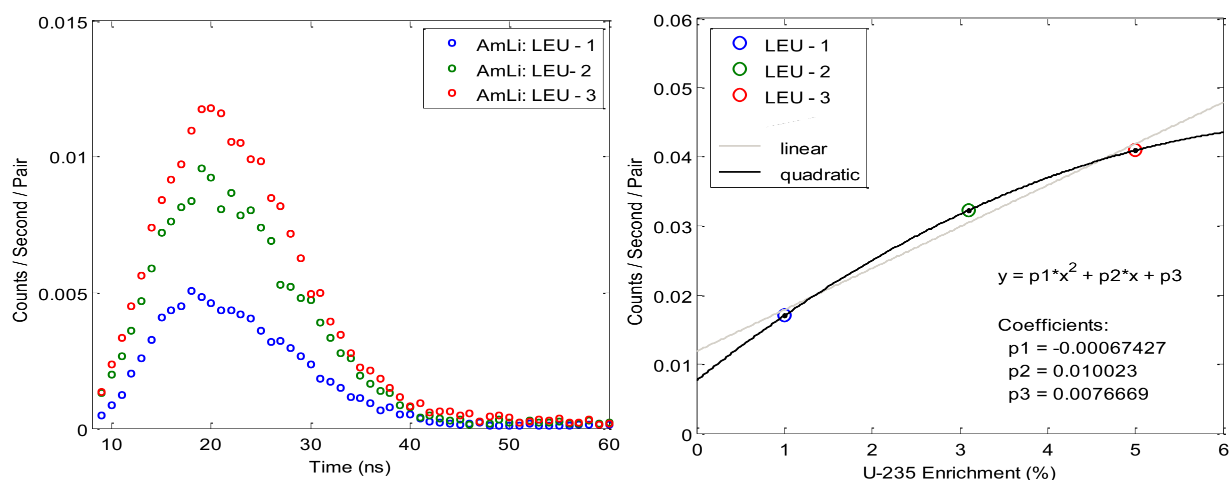
Fig. 6a shows the measured neutron TOF curves for the DT interrogated LEU samples. Fig. 6b shows the trend in the total neutron counts with uranium mass. The neutron counts trend appropriately with uranium mass, with the two canisters of equal mass having roughly the same neutron TOF response for the DT interrogation case.



(a) (b)

Figure 6. a) Measured neutron TOF curves for time-tagged DT interrogation of the three LEU samples and b) the trend of the total neutron counts with uranium mass.

Fig. 7a shows the measured photon-neutron correlations for the Am-Li interrogated LEU. Fig. 7b shows the trend of correlations with LEU enrichment. It is difficult to directly compare the LEU-1 sample to the LEU-2 and LEU-3 samples, as the mass is not consistent, although the general trend agrees with what is expected. The relationship between neutron counts and both uranium mass and enrichment follow the MCNPX-PoliMi predicted trends.



(a) (b)

Figure 7. a) Measured photon triggered neutron TOF curves for the moderated Am-Li configurations, and b) trend of the total photon-neutron correlations with U-235 enrichment.

5. Summary and Conclusions

The objective of this year-long project was to investigate the use of liquid scintillation detectors coupled with active interrogation techniques to characterize uranium-containing materials. Active interrogation methods including a DT generator and an Am-Li source were studied to measured neutron induced fission in LEU powder samples. MCNPX-PoliMi was used for the system design and experiments took place at the JRC in Ispra, Italy in the summer of 2011.

It was seen that high-energy (14.1 MeV) neutrons induced fission in U-235 and U-238 isotopes, allowing the relative amount of mass to be determined from neutron TOF measurements. Then, the supplemental use of very low energy neutrons from a moderated Am-Li source to induce fission in primarily U-235 alone, allowed conclusions as to the relative U-235 enrichment.

Standard charge integration PSD methods were sufficient to discriminate photon detections from neutron detections in the liquid scintillators. This allowed the thorough analysis of neutron TOF distributions with the ability to eliminate photon accidentals. It also allowed TOF distributions to be formed from the Am-Li interrogation cases by triggering on the photons that are emitted from the nuclear reactions in the LEU.

During the 2011 experimental campaign, measurements were also taken on a fuel assembly with the Am-Li source and liquid scintillation detectors. Future work will include the analysis of this data with MCNPX-PoliMi simulations to investigate the use of liquid scintillators in neutron coincidence collar type applications.

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