

MCNPX Simulations of UF₆ Neutron Spectroscopy Measurements using Liquid Scintillator Detectors for ²³⁵U Enrichment Determination

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Abstract

When safeguarding UF₆ cylinders, the determination of the ²³⁵U enrichment and the ²³⁵U mass of the solidified UF₆ is paramount. Current enrichment meter technology relates the ratios of various characteristic gamma-ray peak areas to the level of ²³⁵U enrichment. However, due to strong self-shielding effects, such instruments can only assess the outer-most layer of UF₆. Thus, little information can be gained on the composition of the inner layers of UF₆ or on the bulk ²³⁵U mass. Therefore, there are concerns that more highly enriched UF₆ could be concealed inside a shell of low enriched UF₆. The spontaneous fission of ²³⁸U and especially the (alpha, neutron) reaction of ²³⁴U alpha particles on ¹⁹F provide a significant neutron emission spectrum. Neutrons, compared to gamma rays, are significantly less affected by self-shielding in the UF₆. Consequently, there has been increased interest in developing neutron-based enrichment measurement devices. Organic liquid scintillators possess properties favorable towards fast timing and large detector volume applications. The fast timing arises from the liquid's very short, ns-scale fluorescence decay time. Using a relatively low cost liquid phase material also grants flexibility in designing the detector's shape and volume. Also, pulse shape discrimination of gamma rays and neutrons is possible. Therefore the feasibility of using organic liquid scintillator detectors is being investigated using MCNPX and MCNPX-PoliMi simulations. Specifically, the possibilities of coupling measured liquid scintillator UF₆ neutron energy spectra or measured neutron coincidences to algorithms to estimate the UF₆'s ²³⁵U enrichment are being explored. This paper includes results from the extensive simulation study on neutron coincidences, which are found to correlate with uranium enrichment and appear to provide counting rates that will allow cylinder assays to be performed in minutes.

Introduction

Background

As nuclear power continues to spread to new markets, the International Atomic Energy Agency will be obligated to apply safeguards to the mounting number of fuel cycle facilities being constructed in non-nuclear weapons states. Of these fuel cycle facilities, uranium enrichment facilities garner heightened attention by international safeguards efforts. In modern enrichment facilities, uranium

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hexafluoride (UF_6) is guided through a series of centrifuges in order to increase the isotopic concentration of ^{235}U from naturally occurring 0.72 at% to enrichments of 3-5 at% required for conventional light water reactor fuel. Material identification and accountancy techniques are required to ensure that no UF_6 is being enriched to higher than disclosed ^{235}U concentration, and that no UF_6 is being diverted from the facility. The production of highly enriched uranium (> 90 at% ^{235}U) or the diversion of UF_6 for non-disclosed purposes would both lead to suspicions of illicit military applications of said materials.

In the United States, enriched UF_6 is stored in 30B cylinders, while natural UF_6 (feed) and depleted UF_6 (tails) are stored in 48Y cylinders. The simulations discussed in this paper pertain only to 30B cylinders, but could easily be applied to 48Y cylinders as well. 30B cylinders have an outer radius of 38.1 cm (15 in), a length of 193.04 cm (6 ft 4/12 in), a steel wall thickness of 1.27 cm (0.5 in), and can hold up to 2,277 kg (5,020 lb) of UF_6 [1].

Current UF_6 safeguarding techniques

Measurement techniques must provide information on UF_6 mass and ^{235}U enrichment. Currently, separate techniques are used to measure these two parameters, and both techniques leave much room for improvement.

Portable load-cell-based systems (LCBS) currently are commonly used to ascertain UF_6 mass [2]. Apart from being cumbersome and impractical to use in storage facilities containing dozens of closely packed cylinders, using an LCBS requires the inspector to have blind faith in the facility-provided empty weight of the cylinder. The inspector must also assume that the contents of the cylinder are only UF_6 . If a facility were diverting material, and replacing it with dummy material to match its weight, an inspector using LCBS data would be none the wiser.

If the enrichment detection technique were capable of screening the entire cylinder volume, some of the above concerns would be significantly mitigated. However, that is not the case. Current safeguarding techniques for UF_6 cylinders heavily rely upon gamma-based non-destructive assay (NDA) techniques. Commercially available products [3] use high-resolution gamma ray detectors, generally high purity germanium (HPGe), to obtain a detailed gamma emission spectrum. Enrichment meters use the net area of the ^{235}U predominant 185.7 keV gamma peak to infer ^{235}U enrichment. Software algorithms also correct for the attenuation of gamma rays in the cylinder wall. If the cylinder wall thickness is not known precisely, the calculated ^{235}U enrichment may have an uncertainty upwards of 5% [4].

Another concern pertains to the high degree of self-shielding associated with high Z materials such as uranium. This means that the mean penetration depth of the 186 keV gamma rays is on the order of millimeters, so that any enrichment measurement only pertains to a tiny volume adjacent to the cylinder's inner wall. Therefore every measurement represents merely a snapshot of the much larger total UF_6 volume. In essence, an inspector must assume that any material that is not directly adjacent to the cylinder wall is of the same enrichment as the outermost thin layer of UF_6 that can actually be measured.

New UF₆ safeguarding techniques

The mean penetration depth of fast neutrons in UF₆ is much greater than that of gammas [5]. Various decay types contribute to the overall neutron signature. Three isotopes are the predominant constituents of natural uranium: 0.0055 at% ²³⁴U, 0.72 at% ²³⁵U and 99.2745 at% ²³⁸U. At low enrichments, the enrichment of ²³⁴U is proportional to the enrichment of ²³⁵U [6]. All three of these isotopes can undergo alpha decay. These α particles can then interact with the fluorine in UF₆ and undergo the ¹⁹F(α,n)²²Na reaction. These reactions produce neutrons of energies up to approximately 2.5 MeV. Spontaneous fission of ²³⁸U produces on average 2.1 neutrons per fission, and these neutrons have energies up to 10 MeV. Induced fission of ²³⁵U is an additional source of neutrons.

Organic liquid scintillators possess properties favorable towards fast timing and large detector volume applications. The fast timing arises from the liquid's very short, ns-scale fluorescence decay time. Using a relatively low cost liquid phase material also grants flexibility in designing the detector's shape and volume. Also, pulse shape discrimination of gamma rays and neutrons is possible.

A neutron scatter camera (see Figure 1) consists of two planes of such liquid scintillator detectors. A neutron may scatter off of a hydrogen or carbon atom in the first plane of detectors, thus depositing some energy that is measured as light by a photomultiplier tube. The scattered neutron then may undergo an additional scattering in the second plane of detectors. Since the fast timing property of liquid scintillator allows for a precise time of flight measurement, this information along with the known flight path distance between the two detectors, as well as the energy deposited in the first plane can be used to calculate the incident neutron energy (see Figure 2).

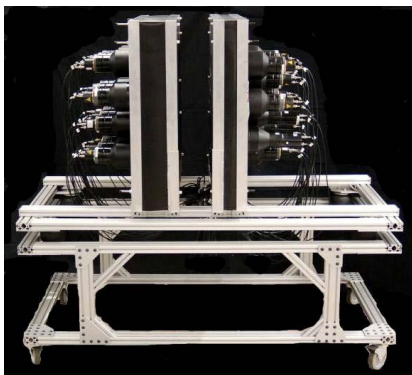


Figure 1: Neutron Scatter Camera [7]

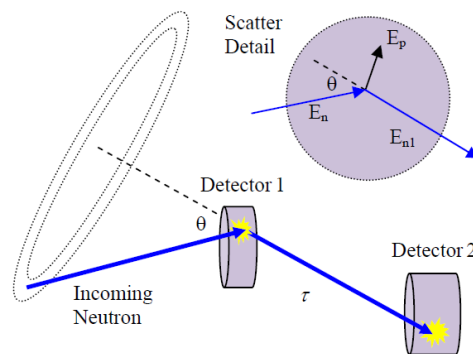


Figure 2: Neutron Scatter Camera Physics [7]

Such a neutron scatter camera system has been developed at Sandia National Laboratories [7]. Using ratios of carefully chosen neutron energy spectrum regions, algorithms are being designed to compute UF₆ enrichment from these spectrum area ratios. This may be possible since the ¹⁹F(α,n)²²Na reactions and fission reactions produce two distinctive neutron energy spectra. While (α,n) neutrons dominate the lower energy spectrum, fission neutron energies cover a much wider range of possible energies. As will be shown later, (α,n) neutrons can be predominantly attributed to the high alpha decay activity of ²³⁴U, and to a much lesser extent the alpha decay activity of ²³⁵U and ²³⁸U. Assuming that ²³⁴U enrichment is proportional to ²³⁵U enrichment, one could choose a low neutron energy region of the

spectrum to represent ^{235}U content. At higher neutron energies, spontaneous fission neutrons from ^{238}U dominate (see Figure 3).

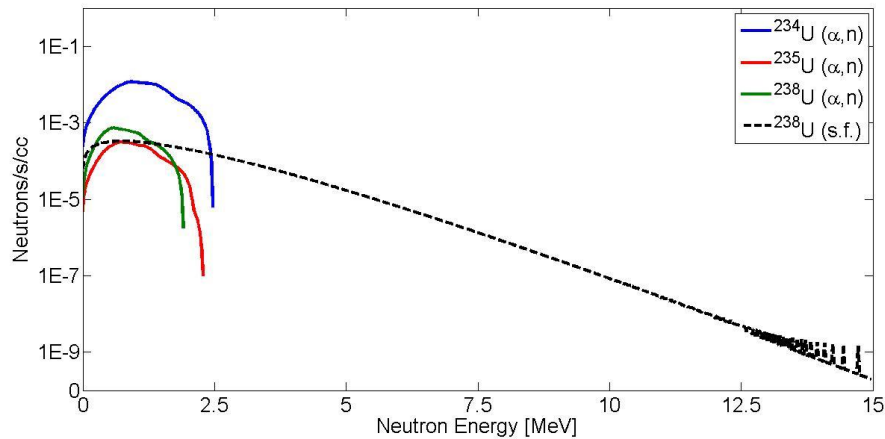


Figure 3: SOURCES-4C Neutron Spectrum of 5% Enriched UF_6 (4.6 g/cc)

The Passive Neutron Enrichment Meter (PNEM) (see Figure 4) is another UF_6 safeguarding tool being developed at Los Alamos National Laboratory [2]. It utilizes pods containing a dozen ^3He tubes and the polyethylene necessary to thermalize fast neutrons emitted by the UF_6 (see Figure 5). Thermalizing neutrons is necessary to take advantage of the much higher cross-section of the $^3\text{He}(n,p)^3\text{H}$ reaction at those energies. Although the energy signature of the neutrons is lost, multiplicities are used as a signal instead. It has been shown that the measured singles rate increases linearly with UF_6 mass, while the doubles-to-singles count rate can be related to ^{235}U enrichment [2]. However, ^3He supplies are assumed to be severely constrained for the near future.

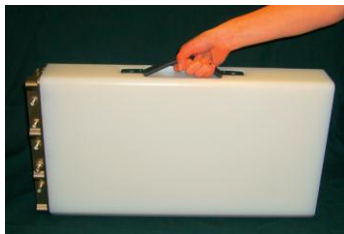


Figure 4: PNEM Detector Pod [2]

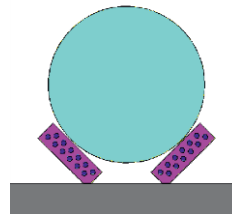


Figure 5: Two PNEM Pods In Use With 30B Cylinder [2]

Proposed UF_6 safeguarding technique using liquid scintillator detectors

The Detection for Nuclear Nonproliferation Group (DNNG) at the University of Michigan is looking to develop a system that incorporates aspects of both of the aforementioned designs. An array of four 12.7 cm diameter EJ-309 liquid scintillator detectors is to be used for multiplicity measurements similar to those performed with ^3He tubes for the PNEM. Singles and doubles-to-singles count rate analysis would be performed analogously, albeit with a less expensive and more abundantly available detector material.

Spectral unfolding and other spectral analysis could also be performed using the spectrum measured in a single liquid scintillator, similar to the analysis discussed for the neutron scatter camera.

Simulations

Source

All particle transport simulations were performed with MCNPX-PoliMi. The mixed source option was used to sample ^{234}U (α,n), ^{235}U (α,n), ^{238}U (α,n) and ^{238}U spontaneous fission sources. As these particular (α,n) reactions on fluoride did not exist yet in MCNPX-PoliMi, the development of custom sources was required.

SOURCES-4C [8] was used with an assumed UF_6 density of 4.6 g/cc [9] to produce neutron energy spectra for various ^{235}U enrichments (see Figure 3 as an example). Dr. Enrico Padovani of Politecnico di Milano used these (α,n) energy spectra to create custom PoliMi sources for the $^{19}\text{F}(\alpha,n)^{22}\text{Na}$ reactions for ^{234}U , ^{235}U and ^{238}U alpha particles.

The user inputs the atom density of the uranium isotopes in atoms/cc into SOURCES-4C. This is the only input that will vary when considering UF_6 of varying enrichments. The code will compute the number of neutrons emitted per second per cubic centimeter of UF_6 individually for all three considered (α,n) reactions and the ^{238}U spontaneous fission. After correcting this value for $\bar{\nu}$, in the case of ^{238}U spontaneous fission, these four values were used as relative probabilities for sampling the four PoliMi sources. An additional source of neutrons arises from induced fission caused by neutrons traveling through the UF_6 , and could be found in the neutron production tables in the MCNPX output file. Spontaneous fission of other uranium isotopes, as well as (n,xn) reactions were negligible ($\ll 1\%$ of total neutron production).

The relative contributions of these five neutron sources are shown for UF_6 of 0.7% and 5% ^{235}U enrichment in Figure 6.

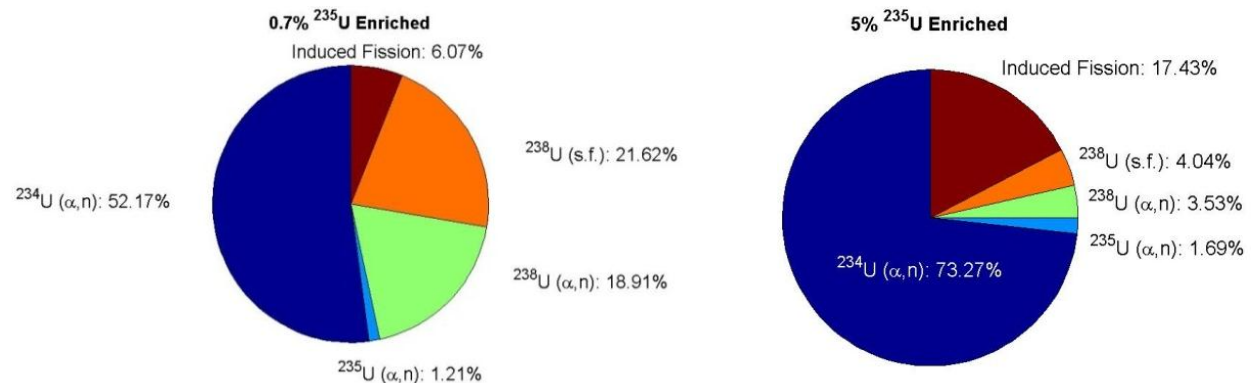


Figure 6: Percentage of Neutrons Produced From Various Reactions For 0.7% and 5% Enriched UF_6

Looking at Figure 6, one clearly sees how increased enrichment enhances the ^{234}U (α,n) neutron contribution and decreases the ^{238}U spontaneous fission contribution.

For the multiplicity simulations, the source was time distributed from 0 to 600 seconds, and the NPS card (the desired number of simulation histories) was computed appropriately based upon the source strength computed with SOURCES-4C.

Geometry

Only a 30B cylinder, its UF₆ content and four liquid scintillator detectors were modeled in MCNPX. The 30B cylinder dimensions were described previously [1]. The 30B cylinder was assumed to be constructed out of stainless steel of 7.86 g/cc density. The distribution of the UF₆ depends upon how the UF₆ is filled into the 30B cylinder, as well as on the environmental conditions during storage [4]. A uniform cylindrical shell of 12.23 cm thickness was chosen together with a UF₆ density of 4.6 g/cc to ensure that the 30B cylinder was filled to its maximum limit of 2,277 kg.

Four EJ-309 detectors were placed in a line along the side of the 30B cylinder, parallel to its axis. The active volumes of the cylindrical detectors had diameters and heights of 12.7 cm (5 in). The EJ-309 material was modeled as 55.54 at% ¹H and 44.46 at% C at 0.935 g/cc density. The detector casings were 0.15 cm thick and made of stainless steel. The front faces of the detectors were placed 5 cm from the 30B cylinder and the gap between individual detectors was 2 cm. The entire setup is illustrated in Figure 7.

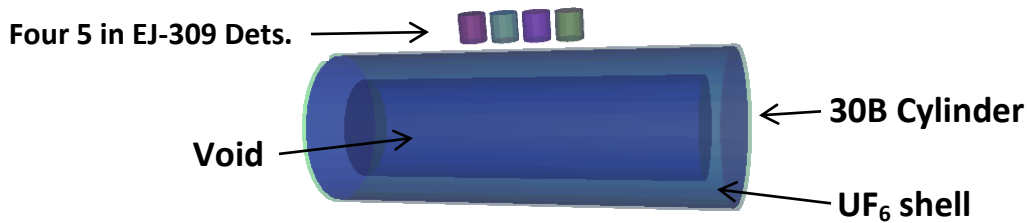


Figure 7: MCNPX Geometry For UF₆ Multiplicity Studies

Post-processing

DNNG developed a post-processing script (MPPost [10]) that takes the collision list file from the output of MCNPX-PoliMi and applies a detector response. For these simulations the appropriate detector response for liquid organic scintillator detectors was selected. A 70 keVee lower threshold and a 2.14 MeVee upper threshold were applied. Light production for neutron collisions on hydrogen was modeled using equation (1); collisions on carbon were modeled using (2):

$$H: \text{Light Energy [MeVee]} = (0.74787E_d - 2.4077(1 - \exp(-0.29866E_d))) \quad [10] \quad (1)$$

$$C: \text{Light Energy [MeVee]} = 0.02 E_d \quad [10] \quad (2)$$

where E_d [MeV] is the energy deposition in the detector written to the PoliMi output data file. MPPost then produces pulses for all interactions recorded in the detector during the simulation.

Additional modules were used to extract neutron and photon multiplicities. For the scintillator multiplicity module of MPPost a digitizer window length of 400 ns was selected. The digitizer dead time

was chosen to be 16 ns. Finally, cutoffs at 80 ns and 220 ns were chosen as the range between which pulses would be accepted.

The relevant output of MPPost is a table of the number of occurrences of a number of neutron and photon multiplicities. These simulations were performed for UF_6 of 0.7, 1, 2, 3, 4, and 5 % ^{235}U enrichment, always using the same geometry and MPPost settings. However, for the analysis only the neutron multiplicities will be considered. Since many of these may in fact be attributed to crosstalk, they will be merely considered as coincidences and not true multiplicities.

Results and Discussion

Neutron pulse height distributions are shown for all simulations in Figure 8. Up to this point, only limited spectrum unfolding analysis has been performed. When plotted on a linear scale, major variations with ^{235}U enrichment are observed at lower energies of the pulse height distributions, a signature being investigated in a similar effort [5]. This trend is also visible in the detector incident neutron spectra which show large variations at lower energies (see Figure 9).

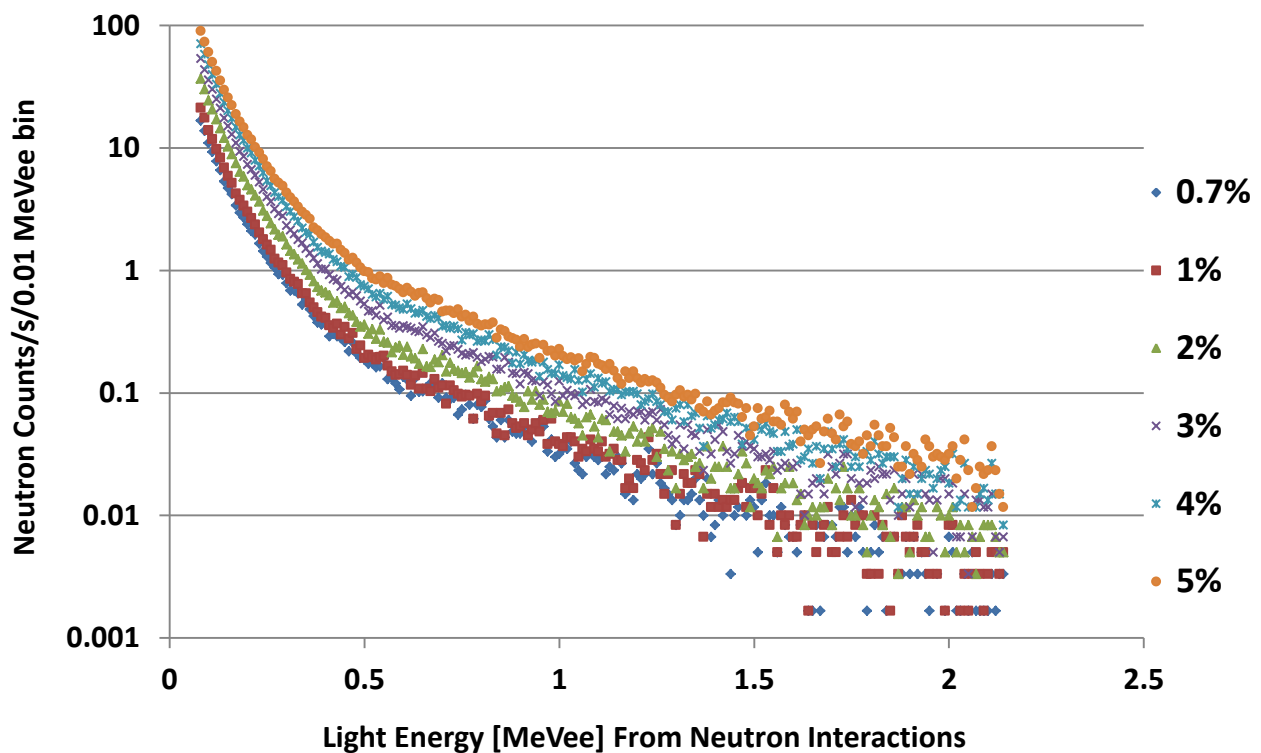


Figure 8: Simulated Neutron Pulse Height Distributions For Liquid Organic Scintillators Measuring 30B Cylinders With UF_6 Of Six Different Enrichments

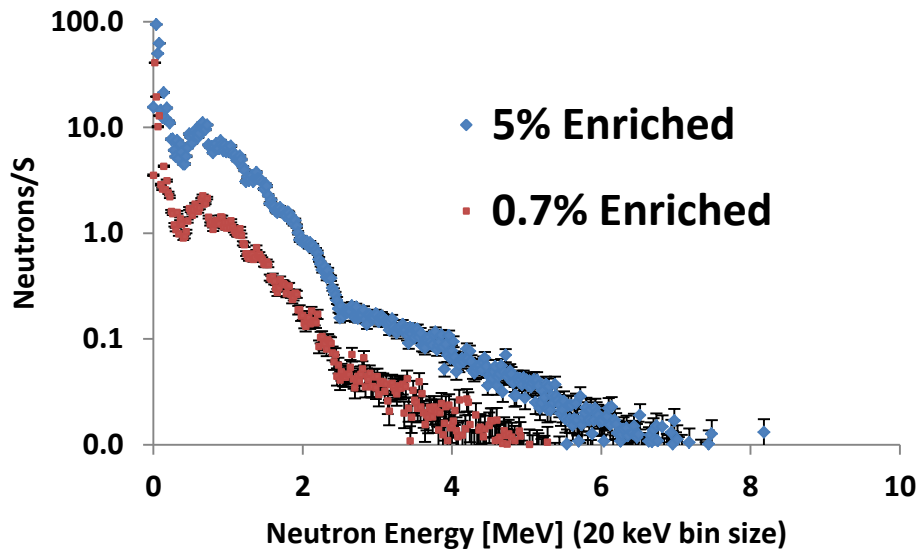


Figure 9: F1 Neutron Tally On Front Face Of Bottom Detector For Two Enrichment Extrema

The MPPost coincidences for the six simulations are shown in Table 1.

Table 1: MPPost Coincidence Occurrences For Four 5 in Liquid Organic Scintillators Measuring 30B Cylinder For 600 s

Multiplicities	0.7% ²³⁵ U	1.0% ²³⁵ U	2.0% ²³⁵ U	3.0% ²³⁵ U	4.0% ²³⁵ U	5.0% ²³⁵ U
n	69266	86903	149791	218970	293806	376356
nn	479	537	917	1466	2124	2871
nnn	4	6	4	10	21	30
nnnn	0	0	0	0	0	0

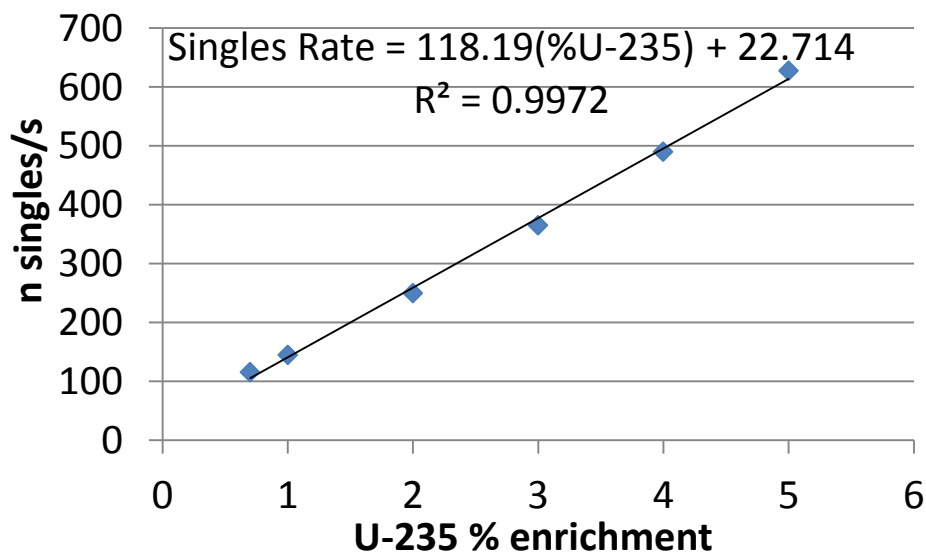


Figure 10: Neutron Singles Rate As Function Of ²³⁵U Enrichment

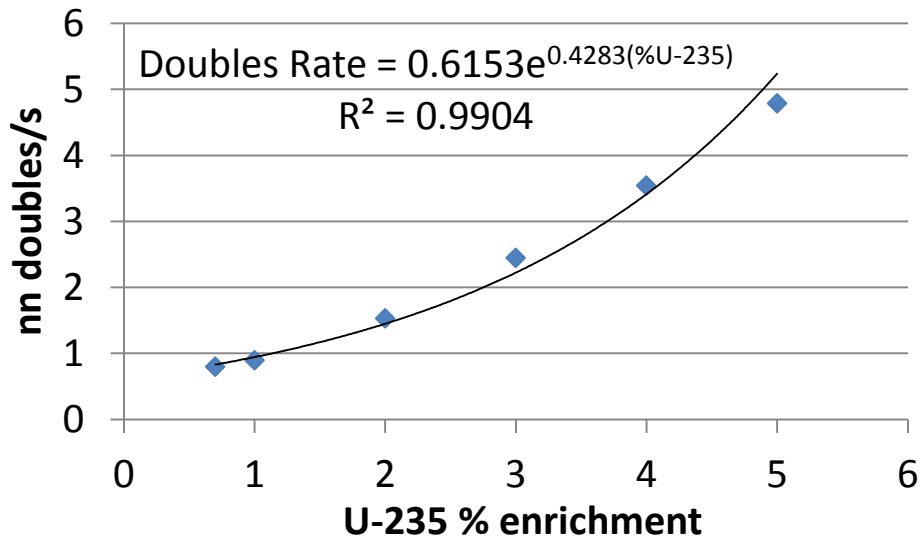


Figure 11: Neutron Doubles Rate As Function Of ^{235}U Enrichment

Figure 10 shows that the neutron singles rate is an exceedingly linear function of ^{235}U enrichment, while Figure 11 illustrates that the neutron doubles rate appears to have an exponential dependence upon ^{235}U enrichment. The singles rate appears to be rather sensitive to enrichment, with a linear slope of approximately 118 singles counts per second per percentage point change in enrichment. Due to the exponential nature of the doubles rate, its sensitivity to enrichment improves with increasing enrichment. However, further simulations will be needed to study the effects of variations in the UF_6 mass as well as the UF_6 fill profile in addition to the already simulated effects of enrichment variation.

Also, changes in the MPPost multiplicity module settings, such as the digitizer window length, may create a more linear doubles rate. Many of the doubles may be accidentals and crosstalk since, as shown in Figure 6, the majority of neutrons arise from (α, n) reactions for which multiplicities are impossible. Further research on spectrum unfolding may glean more information from the pulse height distributions.

Conclusions

MCNPX-PoliMi simulations of four liquid organic scintillator detector responses to a 30B UF_6 cylinder's neutron and photon emissions were performed. It was shown that neutron singles and doubles rates increased linearly and exponentially with respect to ^{235}U enrichment, respectively. This coincidence analysis as well as future pulse height distribution analysis show promise as a safeguarding technology that, unlike current gamma-based enrichment meters, is sensitive to a much larger volume of UF_6 .

Assuming continued positive simulation findings, field experiments on actual 30B cylinders are planned to be conducted later this year. If these studies continue to show positive results, liquid organic scintillators could be a more inexpensive and more reliable alternative to HPGe enrichment meters and

^3He multiplicity detectors for safeguarding the hundreds of UF_6 cylinders located at enrichment facilities spread around the globe.

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