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Spectrum Tailoring of the Neutron Energy Spectrum in the Context of Delayed Neutron Detection

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Abstract

For the purpose of measuring plutonium mass in spent fuel, a delayed neutron instrument is of particular interest since, if properly designed, the delayed neutron signal from ^{235}U is significantly stronger than the signature from ^{239}Pu or ^{241}Pu. A key factor in properly designing a delayed neutron instrument is to minimize the fission of ^{238}U. This minimization is achieved by keeping the interrogating neutron spectrum below ~1 MeV. In the context of spent fuel measurements it is desirable to use a 14 MeV (deuterium and tritium) neutron generator for economic reasons. Spectrum tailoring is the term used to describe the inclusion of material between the 14 MeV neutrons and the interrogated object that lower the neutron energy through nuclear reactions and moderation. This report quantifies the utility of different material combination for spectrum tailoring.

Introduction

Most of the world's plutonium (Pu) is stored in commercial spent fuel assemblies and yet, as of today, no method to directly measure the Pu mass in these assemblies exists. In an effort to assist the International Atomic Energy Agency (IAEA) in safeguarding nuclear facilities and material, Los Alamos National Laboratory is leading a multi-laboratory effort to investigate different nondestructive techniques for quantifying Pu mass in spent nuclear fuel. Since no one nondestructive technique can quantify Pu on its own, several techniques need to be integrated into a final product. The starting point for this integration is understanding how each technique functions as a standalone instrument. One technique is delayed neutron counting in which a neutron source (deuterium-tritium, deuterium-deuterium, or ^{252}Cf) creates a bursts of neutrons next to a fuel assembly causing fissions primarily in the uranium and Pu isotopes present in the fuel. Fission fragments are produced and some of these fragments emit delayed neutrons. The resulting signal can be analyzed to determine the fissile content (sum of Pu and U) of the spent nuclear fuel.

Initial results using Monte Carlo N-Particle eXtended (MCNPX) modeling of the delayed neutron instrument show that the delayed neutron signal scales well with fissile content in water[1]. However, this is not the case in air. The high energy neutrons from the neutron generator are moderated less in air then they are in water. As a result, fertile isotopes such as ^{238}U, which makes up more than 95% of the actinides in the fuel, fission. This is undesirable since the purpose of the delayed neutron instrument is to produce a signal that is propositional to the fissile content in the fuel. A spectrum tailoring cylinder in the delayed neutron instrument helps to reduce the incident neutron energy spectrum in an effort to make it useful in air. This paper describes the methods and conclusions in analyzing this spectrum tailoring ring.

Delayed Neutron Background

In designing a delayed neutron instrument, a deutrerium-tritium (DT) fueled neutron generator was the primary neutron source studied given its high neutron output and relatively low cost. Unfortunately the neutrons are born at the relatively high energy of 14 MeV. For the design researched, the neutrons are injected three hundred and sixty degrees around the assembly. Because the goal is to have the delayed neutron count be primarily dependent on fission in fissile material (^{235}U, ^{239}Pu, and ^{241}Pu) it is desirable to minimize the fissions in fertile isotopes such as ^{238}U. The fission cross section for fertile isotopes is generally very low below 1 MeV[1] as illustrated in figure 1 below.

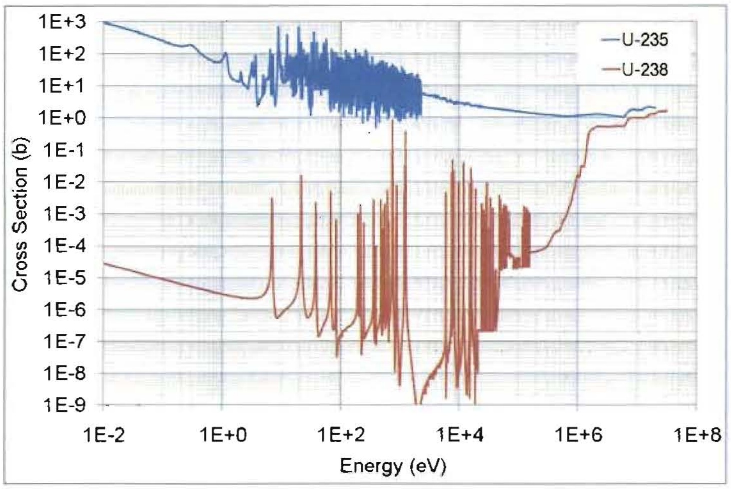

Figure 1: Fission cross section for ^{235}U and ^{238}U as a function of incident neutron energy[2].

To lower the 14 MeV neutrons below 1 MeV a 10 cm cylindrical region was inserted between the fuel and the neutron generator as illustrated in figure 2. In initial modeling this 10 cm region was composed of 8 centimeters of lead placed inside a 2 centimeters of tungsten. The tungsten-lead cylinder was then set between the neutron generator and the assembly to increase the chance of fission by causing (n,2n) reactions. These reactions 1) lowered the energy of the neutrons and 2) increased the number of neutrons. The process by which the energy of the neutrons is lowered is called spectrum tailoring which takes a particle's energy distribution and changes it.

The following tests use MCNPX, a Monte Carlo neutron transport code, to try and determine how different elements with varying thicknesses affect the neutron energy spectrum. The goal is to then use this information to see if the (n, 2n) - moderating cylinder used in delayed neutron counting can be improved.

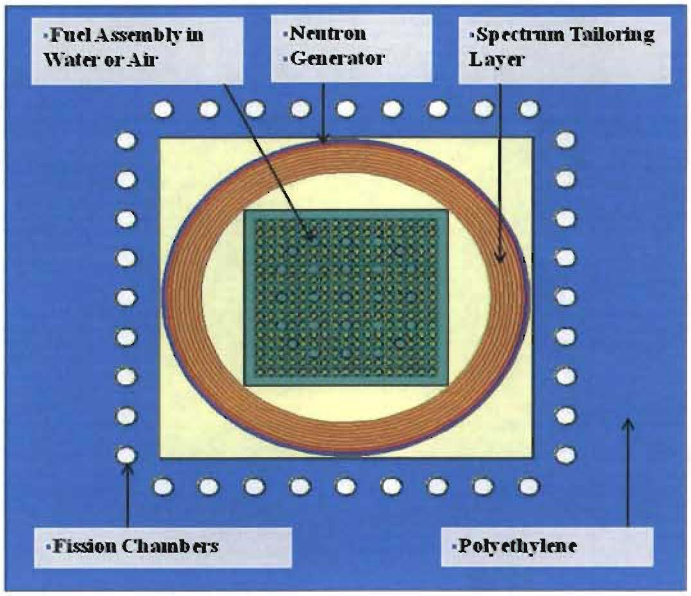

Figure 2: Cross sectional schematic of the delayed neutron generator. The Spectrum Tailoring ring is the orange ring surrounding the fuel assembly.

Choosing a base material

For primary analysis it was decided that a two material spectrum tailoring cylinder could be used and would utilize the advantages provided by different elements. To choose the base material that would be used in all the simulation runs, the (n,2n) cross sections were plotted as a function of incident energy and are shown in figure 3. Of the elements shown in figure 3, Tungsten (W) has the widest cross section for (n,2n) reactions in the desired energy region (0-14 MeV). As a result it was chosen to be the base material for the spectrum tailoring cylinder. To get a reference for how many neutrons are created per each source neutron as a function of tungsten thickness, a very simple MCNPX run was done in which a 14 MeV isotropic point source was placed at the center of a sphere of tungsten. The thickness of the tungsten was varied and the number of neutrons crossing the outer surface of the sphere was tallied. Figure 4 shows a plot of the number of neutrons per source neutron crossing the outer surface of the sphere as a function of material thickness. The number of neutrons exiting the tungsten peaks at around 9 cm. However, due to the cost of tungsten (from $75 to $300 per kilogram) and the size limitations of the spectrum tailoring cylinder, 2 cm of tungsten was chosen for this analysis. Two centimeters of tungsten produces 1.22 neutrons per source neutron and is thin enough to be both affordable and provide enough room for a second material.

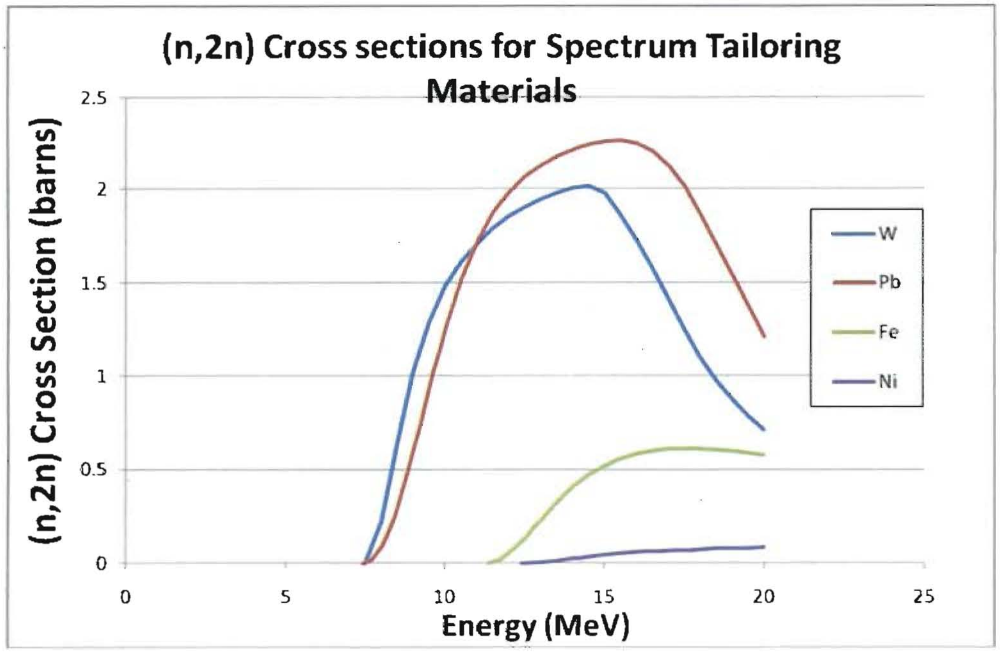

Figure 3: (n,2n) cross section as a function of incident energy of the elements used to optimize the spectrum tailoring ring in the delayed neutron technique. Tungsten has the widest spectrum in the desired energy region of 0-14 MeV.

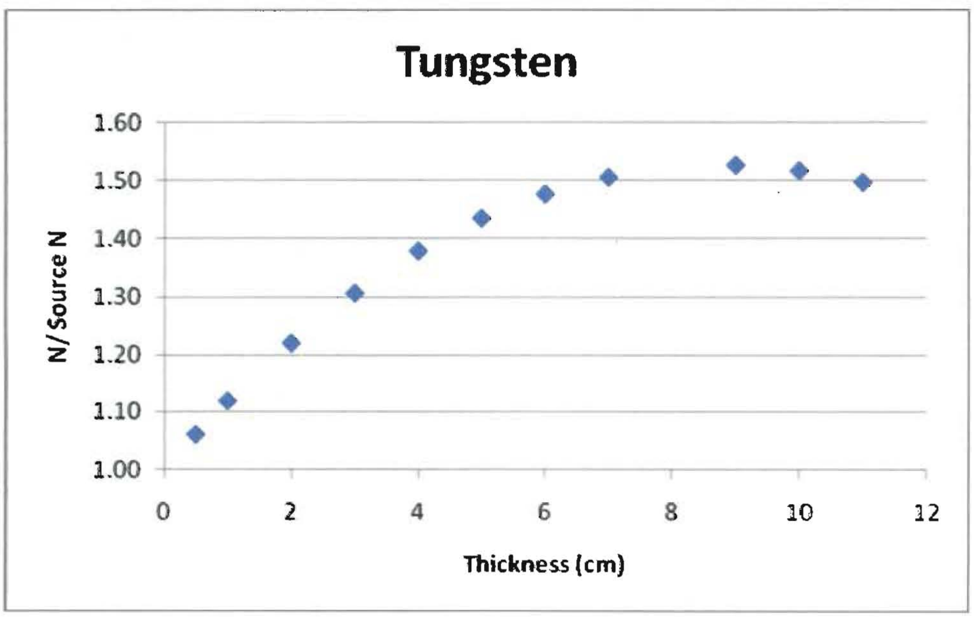

Figure 4: Number of neutrons per source neutron crossing the outer surface of the Tungsten sphere as a function of sphere radius. The neutrons per source neutron peaks at 9 cm.

Description of MCNPX model:

The simple geometry for the MCNPX model was an isotropic point source located at the origin emitting 14 MeV neutrons. The source was surrounded by a 2 centimeter sphere of tungsten at natural density. The tungsten was surrounded by 5 centimeters of various materials. The first run used copper; the second, nickel; the third, iron; the fourth tellurium; and the fifth lead. To simulate an increase in thickness the materials were tested at their natural density and two, four and six times their natural density. The concentric spheres were placed in a void with two other spheres at radii of 8 and 9 centimeters. Theses outer two spheres were used as tally surfaces and have nothing to do with the physics of spectrum tailoring. A surface current tally was placed on the 8 centimeter sphere to count and record the energy of each neutron that passes through it. The simple geometry is illustrated in Figure 5. The simplified model is sufficient because the goal of these tests is to determine how to lower the energy of fast moving neutrons while increasing the number of neutrons.

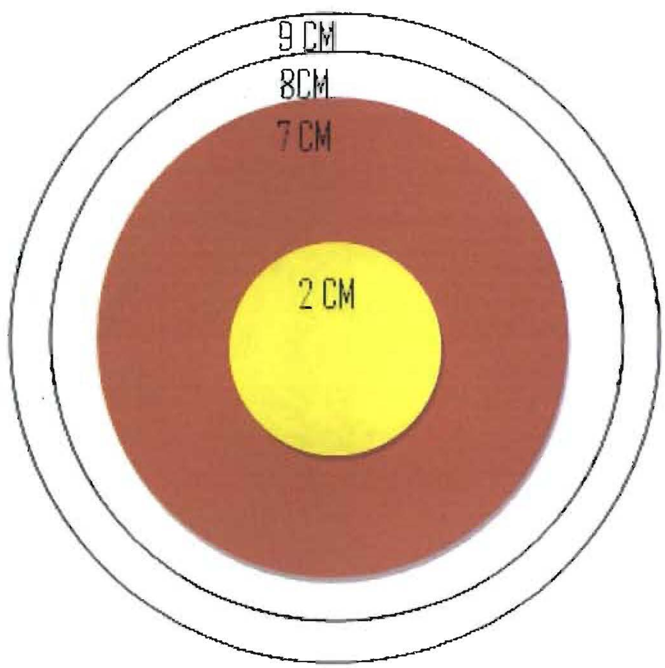

Figure 5: Sliced view of the four spheres that make up basic geometry of the MCNPX simulations. The inner 2 cm sphere is tungsten at natural density while the 7 cm sphere is changed from Cu to Ni to Fe to Te.

Results of the MCNPX runs

The tables below show the percentages of the 14 MeV generated neutrons that are above and below 1 MeV of energy after passing through various materials. One MeV was chosen as the divider because the goal of delayed neutron counting is to calculate the amount of fissile material in an assembly. As illustrated in figure 1, the cross section of ^{238}U (non-fissile) drops substantially at 1 MeV while the cross sections for ^{235}U, ^{239}Pu, and ^{241}Pu remain high[3]. The tables also show the total amount of neutrons (non energy specific) that pass though the 8 centimeter sphere. This value is normalized per one source neutron.

W Cu				
	x1	x2	x4	x6
% Under 1 MeV	47.30	68.00	88.70	98.20
% Over 1 MeV	52.70	32.00	11.30	1.80
Total N/Source N	1.35	1.35	1.12	0.39

Table 1 Neutron energy for a Tungsten-Copper cylinder.

W Ni				
	x1	x2	x4	x6
% Under 1 MeV	35.70	53.20	79.50	96.10
% Over 1 MeV	64.30	46.80	20.50	3.80
Total N/Source N	1.09	0.99	0.80	0.35

Table 2 Neutron energy for a Tungsten-Nickel cylinder.

W Fe				
	x1	x2	x4	x6
% Under 1 MeV	44.40	63.70	85.60	97.40
% Over 1 MeV	55.60	36.30	14.40	2.62
Total N/Source N	1.32	1.35	1.20	0.54

Table 3 Neutron energy for a Tungsten-Iron cylinder.

W Te				
	x1	x2	x4	x6
% Under 1 MeV	40.40	55.10	74.20	88.90
% Over 1 MeV	59.60	44.90	25.80	11.10
Total N/Source N	1.39	1.52	1.63	1.23

Table 4 Neutron energy for a Tungsten-Tellurium cylinder.

W Pb				
	x1	x2	x4	x6
% Under 1 MeV	35.30	46.70	65.20	87.60
% Over 1 MeV	64.70	53.30	34.80	12.40
Total N/Source N	1.45	1.60	1.76	1.78

Table 5 Neutron energy for a Tungsten-Lead cylinder.

Tables 1-5: Percentage of tallied neutrons that were greater than and less than 1 MeV of energy at densities of 1, 2, 4, and 6 times the natural density. It also shows the total tally of neutrons per source.

Conclusions

The purpose of the spectrum tailoring ring is to lower the contribution from fission fragments produced when ^{238}U fissions relative to the fission of fissile isotopes. As a result, the spectrum tailoring ring needed to decrease the energy of about 80% of the initial neutrons to energies of less than 1MeV. The data from above shows promise in lead at six times natural density, copper at four times natural density, and iron at four times natural density. Tellurium also shows relevant numbers but due to the cost ($100 per pound in 2006) it was not considered in the analysis.

The next consideration was the amount of neutrons each method produced per source neutron. The more neutrons that penetrate the spectrum tailoring ring, the higher the chance of fission and the more accurate the technique can be. Of the three cases listed above, lead at six times natural density produces the most neutrons per source at 1.78. Figure 6 shows the percentage of neutrons at different energy levels that contribute to the total neutron signal after the signal is passes through 2 cm of tungsten and 5 cm of lead at 6 times the natural density.

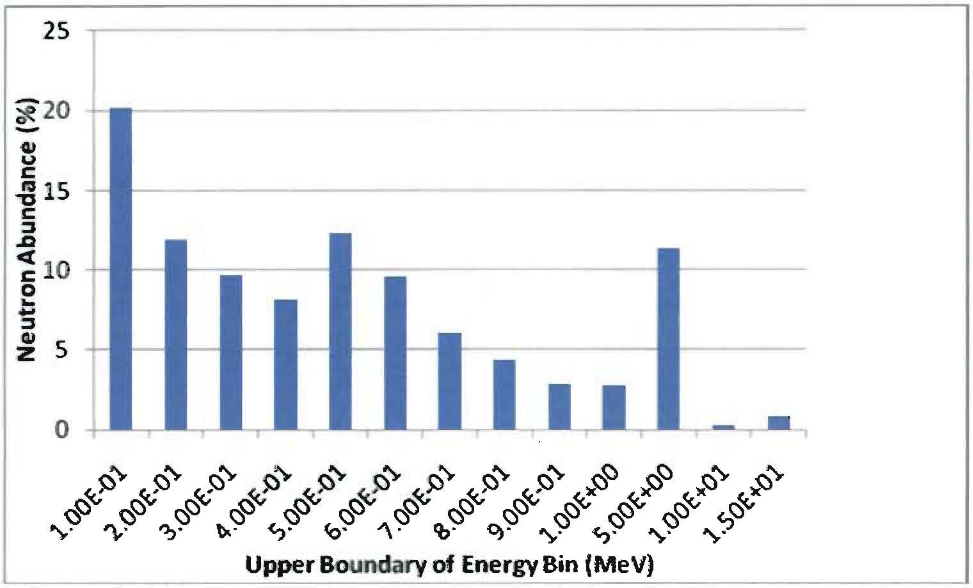

Figure 6: The abundance of neutrons at specific energy levels after passing through 2 cm of tungsten and 5 cm of lead at 6 times the natural density.

Only about 1% of the total neutron flux remains at 14 MeV after passing through the two materials. A majority of the neutrons that pass through with greater than 1 MeV of energy have less than 5 MeV of energy. Having 2 cm of tungsten and 30 cm of lead (equivalent to 5 cm of lead at 6 times the natural density) placed between the neutron generator and the fuel assembly would be a good way to both raise the number of neutrons entering the fuel and lower the energy of the ones that do. Ten percent of the neutrons would still have between 1 and 5 MeV of energy to penetrate the outer regions of the assembly and cause fissions in the center pins.

A spectrum tailoring ring with 2 cm of tungsten and 30 centimeters of lead would increase the size of the instrument considerably, so even though the analysis shows it to be a viable option, it is not realistic. Of the isotopes studied, none give the desired result while still using less than 10 cm of material, which it the current thickness of the spectrum tailoring ring. One possible solution would be to use a material that has a large number of low Z atoms. Polyethylene (CH_2) contains a large number of hydrogen atoms which can lower the energy of a neutron from 2 MeV to 0.025eV in 27 collisions[4]. To have the same energy reduction in higher Z elements like uranium, it takes over 2000 collisions.[4] Using CH_2 would lower the energy of the neutrons using less material. However, when the polyethylene was tested using the same MCNPX geometry described above, all the neutrons per source neutrons were less than 1.

Future work

Of the materials studied in this analysis, no combination of two elements give the desired results of lowering at least 80% of the neutrons to below 1 MeV of energy, maintaining a non-diminishing neutron flux, and using less than ten centimeters of material. Future work will include investigating a spectrum tailoring cylinder with three materials. A possible design would be 2 cm of tungsten surrounding 5 cm of lead to get a high neutron flux with energies around 7 MeV, and filing the rest of the space between the fuel assembly and the spectrum tailoring ring with polyethylene to moderate the neutrons to energies below 1 MeV.

References

1. Nathan P. Sandoval, Stephen J. Tobin, Howard O. Menlove, Martyn T. Swinhoe, Will Koehler, Mike L. Fensin, Vladimir Mozin, Melissa Schear, Josh Richard, and Jianwei Hu, "Determining the Pu Mass in LEU Spent Fuel Assemblies – Focus on Delayed Neutron Detection," Los Alamos National Laboratory Report: LA-UR-09-05317, Los Alamos, NM (2009).

2. M.B. Chadwick, P. Oblozinsky, M. Herman at al.,"ENDF/B-VII.0: Next Generation Evaluated Nuclear Data Library for Nuclear Science and Technology", Nuclear Data Sheets, vol. 107, pp. 2931-3060, 2006.

3. Melissa A Schear "Monte Carlo Study of Replacing Californium in a Shuffler with a Neutron Generator," University of Illinois: 2007

4. D. Reilly, N. Ennsslin, H. Smith, Jr., "Passive Nondestructive Assay of Nuclear Materials," March 1991.