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Replacing a ^{252}Cf Source with a Neutron Generator in a Shuffler- A Conceptual Design Performed with MCNPX

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Abstract

The ^{252}Cf shuffler has been widely used in nuclear safeguards and radioactive waste management to assay fissile isotopes, such as ^{235}U or ^{239}Pu, present in a variety of samples, ranging from small cans of uranium waste to metal samples weighing several kilograms. Like other non-destructive assay instruments, the shuffler uses an interrogating neutron source to induce fissions in the sample. Although shufflers with ^{252}Cf sources have been reliably used for several decades, replacing this isotopic source with a neutron generator presents some distinct advantages. Neutron generators can be run in a continuous or pulsed mode, and may be turned off, eliminating the need for shielding and a shuffling mechanism in the shuffler. There is also essentially no dose to personnel during installation, and no reliance on the availability of ^{252}Cf. Despite these advantages, the more energetic neutrons emitted from the neutron generator (14.1 MeV for D-T generators) present some challenges for certain material types. For example when the enrichment of a uranium sample is unknown, the fission of ^{238}U is generally undesirable. Since measuring uranium is one of the main uses of a shuffler, reducing the delayed neutron contribution from ^{238}U is desirable. Hence, the shuffler hardware must be modified to accommodate a moderator configuration near the source to tailor the interrogating spectrum in a manner which promotes sub-threshold fissions (below 1 MeV) but avoids the over-moderation of the interrogating neutrons so as to avoid self-shielding. In this study, where there are many material and geometry combinations, the Monte Carlo N-Particle eXtended (MCNPX)[1] transport code was used to model, design, and optimize the moderator configuration within the shuffler geometry. The code is then used to evaluate and compare the assay performances of both the modified shuffler and the current ^{252}Cf shuffler designs for different test samples. The matrix effect and the non-uniformity of the interrogating flux are investigated and quantified in each case. The modified geometry proposed by this study can serve as a guide in retrofitting shufflers that are already in use.

[1] *J.F. Pelowitz (Editor), "MCNPX^{TM} USER'S MANUAL Version 2.5.0," Los Alamos National Laboratory report LA-CP-05-0369 (2005).*

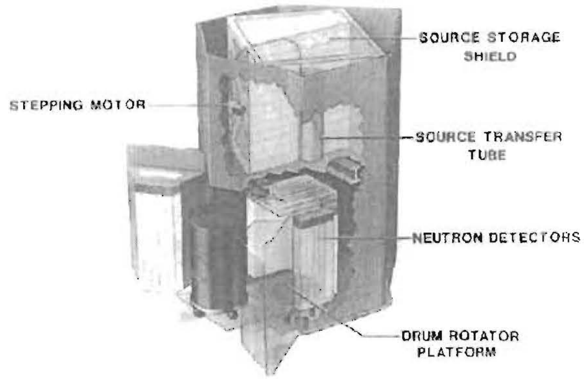

Figure 1: Cut-away geometry of the 55-gallon waste drum shuffler [1]

1 Introduction

A current ^{252}Cf shuffler design is shown in Figure 1. The top half of the diagnostic is a storage block for the isotopic source, with walls made of polyethylene for shielding. The bottom half is the assay chamber, which houses the sample to be assayed. During irradiation, the source is scanned vertically along the height of the drum and the turn table rotates to obtain an even response from fissile materials at different locations in the drum. After the source is shuffled back to the storage block, delayed neutrons produced by the fission fragments within the sample are detected by ^3He detectors, which surround the assay chamber. These detectors are embedded in polyethylene walls. The delayed neutron count rate is proportional to the fissile mass.

Californium-252 , the isotopic source of choice, exhibits a high spontaneous fission neutron yield, approximately 2.3 x$10^{12}\frac{n}{g\cdot sec}$ with a half-life of 2.65 years, and an average neutron energy of 2.3 MeV, a median energy of 3.27 Mev, and a most probable energy of 0.90 MeV. A standard 550 μg of ^{252}Cf used in the shuffler, emits 1.265x $10^9\frac{n}{sec}$. On the other hand, D-T neutron generators emit mono-energetic 14.1 MeV neutrons at comparable and even greater emission rates.

2 Spectrum Tailoring Materials

Spectrum tailoring materials surrounding the interrogating source can be appropriately chosen for either fast or thermal neutron interrogation. Our goal is to minimize the fissions of ^{238}U caused by super-threshold energy neutrons ($E > 1$ MeV) originating from the 14-MeV source. MCNPX simulations are performed to obtain the neutron leakage spectrum from the surface of a single-material sphere of 10 cm radius, made either of tungsten, lead, iron, copper, nickel, polyethylene, graphite, or beryllium. Each sphere contained a 14-MeV isotropic point source at its center. The most suitable material should minimize the number of leaked neutrons with

Table 1: Comparison of Spectral Characteristics of Candidate Materials for a Fast-Neutron Moderator Configuration

Mat.-Radius [cm]	Neutron source	X= W_e+W_c	W_e	W_c	L	M	H	FOM	L[%]	M[%]	H [%]
W-10	14 MeV	1.71	1.52	0.19	0.16	0.90	0.45	1.33	10.9	59.5	29.7
Pb-10	14 MeV	1.50	1.50	0.00	0.03	0.50	0.97	0.25	2.0	33.2	64.9
Fe-10	14 MeV	1.20	1.11	0.10	0.04	0.40	0.67	0.23	3.3	36.2	60.5
Cu-10	14 MeV	1.29	1.23	0.06	0.06	0.62	0.55	0.65	4.6	50.9	44.5
Ni-10	14 MeV	1.07	0.75	0.32	0.01	0.20	0.54	0.07	1.6	26.8	71.6
CH_2-10	14 MeV	1.00	0.89	0.11	0.11	0.04	0.74	0.00	12.3	4.6	83.1
C-10	14 MeV	1.00	0.92	0.08	0.04	0.08	0.81	0.01	4.6	8.2	87.2
Be-10	14 MeV	1.66	1.60	0.06	0.33	0.30	0.97	0.07	20.6	18.9	60.5
None	^{252}Cf	1.00	1.00	0.00	0.01	0.28	0.71	0.11	1.3	28.0	70.7

$E > 1$ MeV, but also minimize those with $E < 0.1$ MeV, an arbitrarily chosen energy that ensures a penetrating spectrum. Overall, the material should maximize the number of neutrons leaving the surface of the sphere by minimizing neutron absorption and maximizing neutron multiplication within the sphere. Table 1 lists the total neutron leakage from the surface of the sphere, W_e, in Monte Carlo weight, as well as the total neutron capture, W_c, within the sphere. The sum of the two results is equal to the net multiplication factor, X, of the material. Spectral information is obtained from L, the weight of neutrons with $E < 0.1$ MeV; M, the weight of neutrons with $0.1 < E < 1$ MeV; H, the weight of neutrons with $E > 1$ MeV. The figure-of-merit, FOM, combines these various results to assess the overall performance of each material, as shown in Equation 1. The first value in the FOM product, M, accounts for absolute weight of desired neutrons emitted, while the remaining $\frac{M}{L+H}$ quantifies the desired spectral characteristics. Although a material may emit neutrons with the right energy distribution, it may emit only a small number of these neutrons.

$$FOM = \frac{M^2}{L+H} \tag{1}$$

Tungsten exhibits the highest neutron multiplication, and the largest figure-of-merit, with desired spectral properties. Another simulation is performed with a bare ^{252}Cf source for the sake of comparison. Note than the approximately 71% of ^{252}Cf neutrons are above the ^{238}U fission threshold energy compared to only 30% of the tungsten-tailored D-T neutrons. Although tungsten exhibits the desired spectrum tailoring properties, a moderator configuration consisting solely of tungsten would be prohibitively expensive and heavy, thus a combination of materials, which includes tungsten will be used in the modeled shuffler geometry. In this simulation, the isotropic point source is replaced by the physical geometry of a neutron generator. The accelerator head is shown in Figure 2. The neutron target consists of a titanium hydride layer and a copper substrate. The rear cathode is modeled as copper, and all the other components are modeled as iron. The 14-MeV neutron source is modeled as a isotropic point source originating in the metal hydride target. Figure 3 incorporates the neutron generator into

Table 2: Spectral characteristics of Tungsten spheres compared to ^{252}Cf source

Moderator Config. mat-radius[cm]	Neutron Source	Total Leakage W_c (MC weight)	Fractions of neutrons with E_n>0.1 MeV	Fractions of neutrons with 0.1 MeV < E_n< 1 MeV	Fraction of neutrons with E_n>1 MeV
W-5	14 MeV	1.43	4.54%	40.51%	54.95%
W-3	14 MeV	1.31	2.63%	27.52%	69.85%
W-2	14 MeV	1.22	1.80%	19.56%	78.65%
No mat.	^{252}Cf	1.00	1.30%	27.95%	70.73%

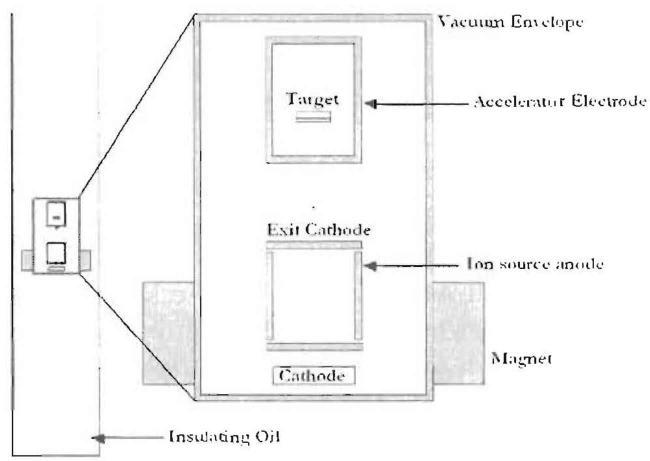

Figure 2: MCNPX model of the neutron generator accelerator head, with a magnified view of the sealed neutron tube

the modeled shuffler geometry, where regions 1, 2, and 3 are highlighted. Region 1 is the 10-cm thick slab of material directly between the generator and the sample, and region 2, consists of 3 slabs of reflector material surrounding the generator on the remaining three sides. The lateral slabs in region 2 are both 3 cm thick, while the back reflecting slab is 5 cm thick. All slabs in the moderator configuration are 116 cm high. Different materials will be simulated for regions 1 and 2, in order to find the optimum moderator composition. Region 3 is a tungsten crown, 3-cm thick and 20-cm high, and is centered on the neutron generator. A thickness of 3 cm for the crown was chosen, since a tungsten sphere, with a radius of 3 cm, provides approximately a factor of 1.3 in neutron multiplication, and approximately the same fraction of super-threshold neutrons as a bare ^{252}Cf source, as seen in Table 2. This is only a rough approximation since the crown is not a sphere, but a cylindrical shell, and will not intercept all the source neutrons, so the crown will be coupled with additional materials for adequate spectrum tailoring. The height of the crown was arbitrarily chosen to provide adequate interception of the 14-MeV source neutrons.

Table 3 lists the different material combinations for regions 1 and 2, and compares the

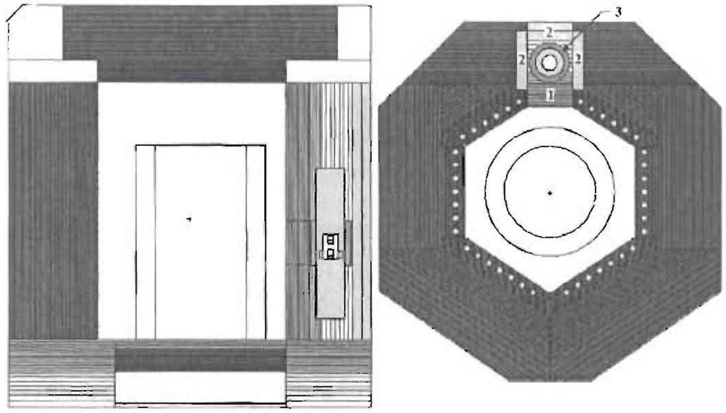

Figure 3: MCNPX modeled moderator configuration for generator-based Shuffler design *(YZ and XY planes respectively)*

neutron spectra incident on the 55-gallon drum located within the irradiation chamber. Note that the fraction of low energy neutrons ($E < 0.1$ MeV) have increased due to moderation in the polyethylene walls. Although the spectra gives valuable information on the tailoring effects of the proposed material, it is also important to investigate the overall effect that the configuration has on the fission rate of the sample within the drum. In one simulation, the drum is filled entirely with low-density ^{235}U. The fission rate of ^{235}U is tallied within the drum, for each of the configurations listed in Table 3. A simulation is also performed on the same sample using the original ^{252}Cf shuffler geometry. The resulting ^{235}U fission rate is almost three-fold compared to the tailored geometry, since three times more neutrons are incident on the drum when there is no tailoring. Simulations were also performed with ^{238}U in the drum, and the ratio of the fission rates of ^{238}U to ^{235}U was tallied for each case. The ratio of the fertile-to-fissile fission rate should be minimized.

The penetrability of the interrogating spectrum is also important to preserve. In each case, the volume of the drum consists of an inner cylinder and an outer cylinder of the same volume as that of the inner cylinder, as shown in Figure 3. These inner and outer cylinders are used to find the fission rates in the inner and outer volumes of the drum. This result estimates the penetrability of the interrogating spectrum, a method used in [2]. For a fully penetrating spectrum, the ratio of inner to outer volume is unity. As the penetrating ability of the incident neutrons decreases, the ratio falls below one. From Table 3, it is evident that configurations having region 1 as copper or iron yield a higher fraction of neutrons in the desired energy range compared to the existing ^{252}Cf shuffler design, and show little variation amongst them. In the current design, however, at least three times more neutrons are incident on the sample than achieved in the moderated geometry. Configurations where beryllium is present for region 1 show the largest fraction of neutrons in the low-energy range, and the least in the desired range. Due to the decrease in interrogation flux for the moderated geometry, the ^{235}U fission rates are less than the ^{235}U fission rate in the current design. All moderator assemblies, however, exhibit a smaller ratio of ^{238}U to ^{235}U fission than the current design, and most have more penetrable spectra. Beryllium is the least effective at moderating 14-MeV neutrons, and also slows down

Table 3: Fast-neutron moderator configurations

Reg. 1	Reg. 2	Weight W inc (MC weight	Neutrons E_n<0.1 MeV	Neutron 0.1 MeV < E_n<1 MeV	Neutrons E_n>1 MeV	^{235}U [fissions / source neutron]	Ratio of 238/235 [%]	inner / outer fissions
Fe	Fe	0.20	35.2%	38.9%	25.9%	1.16e-05	0.047%	0.83
Fe	Cu	0.22	38.6%	38.5%	22.9%	1.34e-05	0.041%	0.84
Fe	Ni	0.20	36.9%	38.0%	25.0%	1.23e-05	0.043%	0.85
Fe	Pb	0.21	35.9%	38.2%	25.9%	1.28e-05	0.047%	0.83
Cu	Fe	0.17	36.4%	39.9%	23.7%	1.03e-05	0.042%	0.83
Cu	Cu	0.18	39.1%	39.4%	21.5%	1.11e-05	0.039%	0.84
Cu	Ni	0.17	38.1%	39.1%	22.8%	1.03e-05	0.042%	0.85
Cu	Pb	0.18	36.9%	39.2%	23.9%	1.06e-05	0.042%	0.83
Be	Fe	0.19	45.6%	18.5%	35.9%	1.56e-05	0.054%	0.82
Be	Cu	0.20	47.7%	18.1%	34.2%	1.68e-05	0.048%	0.83
Be	Ni	0.20	46.6%	18.2%	35.2%	1.58e-05	0.052%	0.84
Be	Pb	0.20	45.9%	18.6%	35.6%	1.64e-05	0.054%	0.83
^{252}Cf		0.66	24.3%	32.5%	43.2%	3.17e-05	0.066%	0.82

neutrons to lower energies, resulting in an increase of both fissile and fertile fission.

Since an all-iron configuration is relatively cheap compared to copper and nickel, exhibits comparable performance, and is not difficult to machine, in the case of beryllium, or heavy, like lead, this configuration was chosen as the optimum fast-neutron moderator configuration. Figure 4 shows the incident spectra on the 55-gallon drum during the first irradiation interval. The tailored generator source interrogates the sample with the desired spectrum, which peaks right below the 1- MeV threshold energy. There are still 14-MeV neutrons incident on the sample, since only an excessive amount of tailoring would make them negligible. The data in Table 3 is used to calculate the ratio of neutrons above the 1-Mev threshold, to those below the 1-MeV threshold for both designs. The ^{252}Cf shuffler has a ratio of 0.76 while the generator-based design has a ratio of 0.35. There is a larger fraction of super-threshold neutrons in the ^{252}Cf shuffler interrogating spectrum.

The iron slabs in the tailoring assembly can be composed of smaller, more manageable slabs that can be easily removed from the diagnostic in segments when spectrum tailoring of the source is not desired or when the diagnostic needs to be transported. When a direct 14-MeV interrogating spectrum is required, the iron slab in between the source and the sample can be removed and placed behind the generator for additional reflection of neutrons toward the sample.

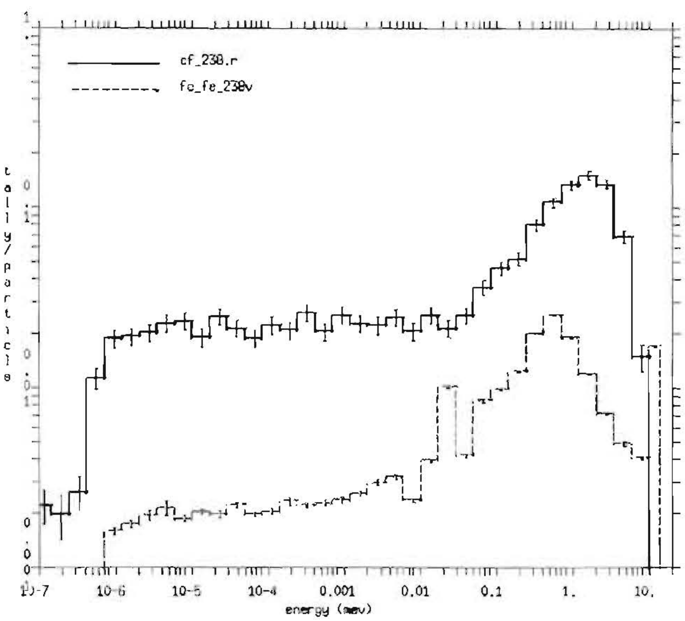

Figure 4: The incident neutron spectra on the 55-gallon during irradiation for the current ^{252}Cf design and the tailored, neutron generator shuffler design

3 PERFORMANCE

3.1 The position effect

The measured delayed-neutron count rate obtained from shuffler is converted into a fissile mass using calibration curves. The measured count rate should be independent of physical characteristics of the sample, such as its size, shape, density, mass, isotopics, hydrogen content, matrices, and material content. Realistically, this is not the case. As previously discussed, the fissionable isotopes, such as ^{238}U in the sample, can contribute to the count rate. The measured count rate may also depend on the position of localized fissile material within the drum due to the spatial variation of the irradiating flux and the presence of shielding matrix material within the drum. Even when the fissile mass is homogeneously distributed within the drum, different types of matrix material cause count rate variations. For example, a given matrix may be a prolific neutron absorber, or an excellent moderator. The non-uniformity of the interrogating flux was investigated for both designs.

The Cf-based design attempts to even out the non-uniformity by scanning the ^{252}Cf source vertically, and the drum also sits on a rotating platform. It would be difficult to scan the neutron generator in this manner, so we must quantify the spatial count rate variation due to stationary generator. For the neutron generator centered vertically on the drum, the neutron flux peaks at the drum's mid height, and then decreases away from that mid-point, for an empty drum. A first concern is to quantify the variation in measured count rate due to this effect, excluding any possible variations due to the presence of matrices. Simulations were performed with localized fissile material at varying heights, 10 cm apart from 1cm to 81cm, along the empty drum's

Table 4: Measured count rate dependence on the height of fissile mass in an empty drum

	Existing Cf-252 design		N. gen. design	
Height z [cm]	Tally (n p) reactions per src neutron	Count rate [cnt/sec]	Tally (n p) reactions per src neutron	Count rate [cnt/sec]
1 37	1 81E-07	73	5 73E-08	23
11 37	1 77E-07	71	6 09E-08	25
21 37	1 78E-07	72	6 98E-08	28
31 37	1 78E-07	72	7 07E-08	29
41 37	1 74E-07	70	7 20E-08	29
51 37	1 82E-07	73	7 08E-08	29
61 37	1 83E-07	74	6 39E-08	26
71 37	1 72E-07	70	5 79E-08	24
81 37	1 69E-07	68	5 15E-08	21
average count rate		71		26
std dev		1 88		2 96
max to min ratio		1 09		1 38

center axis. The localized fissile material was 263 g of pure ^{235}U, at reduced density of 2.0 g/cm^3 , in the shape of a small cylinder, of radius 3 cm and height 4.64 cm. The simulation tallied the delayed neutron detection probability in the ^{3}He detection banks during the first count interval. The results are summarized in Table 4.

For the generator-based design, the average count rate is 26 counts/sec, approximately three times less than the average of 71 counts/sec measured for the existing design, which is consistent with the fact that roughly three times fewer neutrons are incident on the sample for the tailored assembly, compared to the existing design, as stated in Table 3. The count rate fluctuations are slightly larger for the generator-based design, with a 1.38 maximum-to-minimum count rate ratio, and a standard deviation of approximately three counts per second, or 12% of the average count value. The standard deviation of the current design is 3% of the average value. There is a 3% statistical error on all tally results. These axial count rate variations may be mitigated by the use of iron reflectors in the generator-design.

3.2 The Matrix Effect

The matrix is considered the non-fissile material in the assay chamber. For simulation purposes, the matrix was homogeneously distributed throughout the drum, surrounding a localized mass of ^{235}U, located in three center-axial positions (z- 1, 41, 81 cm). We investigated how different matrices affected the delayed neutron count rate for a given fissile mass in these axial positions. There are three matrices used in the simulations: low-density polyethylene (0.056 $\frac{g}{cm^3}$), high-density polyethylene (0.56 $\frac{g}{cm^3}$), and reduced-density iron (2.62 $\frac{g}{cm^3}$). The reduced-density iron matrix is not considered to have moderating properties, since it scatters neutrons without much energy loss, but there is minimal absorption of low energy neutrons, which may decrease the delayed neutron count rate. The polyethylene matrices have significant neutron moderating properties, and can significantly affect the measured count rate. The low-density polyethylene matrix is used to represent polyethylene shavings, while the high-density polyethylene matrix represents polyethylene beads.

Figure 5 shows the count rate variation for these axial positions, in the different matrices, for the generator-based design. The highest count rate is obtained for the low-density polyethylene matrix, at the center of the drum, where there is the highest population of thermalized neutrons,

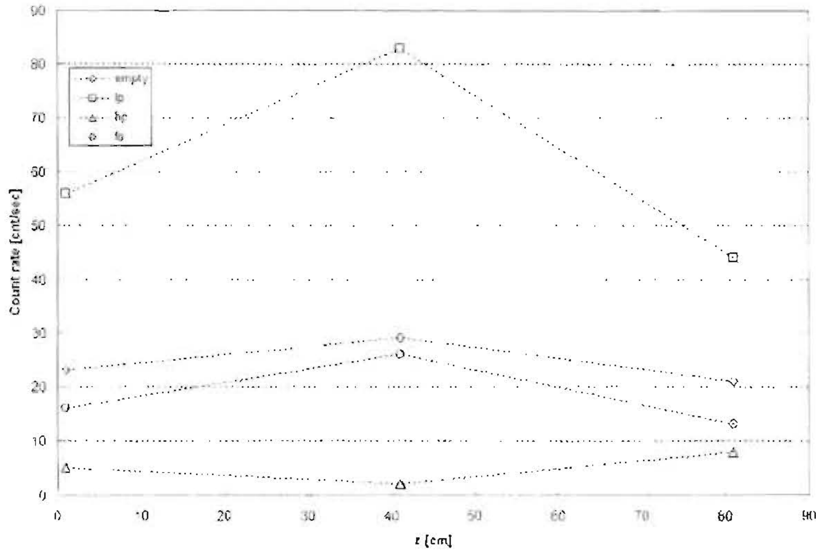

Figure 5: Count rate variation as a function of height for inner points for generator-based design for different matrices

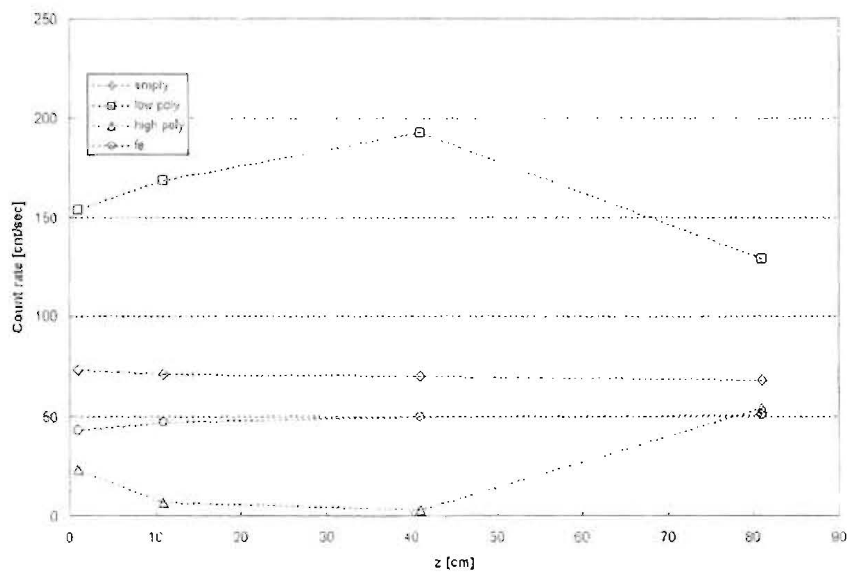

Figure 6: Count rate variation as a function of height for inner points for ^{252}Cf shuffler for different matrices

thus the highest fission and delayed neutron count rate. Neutrons here are least likely to escape the drum before causing fissions. The empty matrix produces the next highest count rate. Although there is no neutron absorption to decrease the delayed neutron count rate, there is no moderator to increase the fission probability. Similar to the low-density polyethylene matrix, the delayed-neutron count rate peaks at the center of the drum, where the stationary generator flux is highest. Although the high-density polyethylene matrix is a very good moderator, the higher hydrogen concentration is detrimental to the both the interrogation of the fissile material and the detection of the delayed neutrons, due to over-moderation of the neutrons. This effect is readily observed since the lowest count rate is in the center of the drum. Even the iron matrix, with no moderating properties and absorbing properties, exhibits higher count rates. A similar matrix effect can be seen in Figure 6 in the ^{252}Cf-based design. Overall the count rates in the original design are higher than the generator-based design due to the increase in interrogating neutrons, as discussed earlier. For the ^{252}Cf-based design, there is little count rate variation for the empty and iron matrix. Since the source is scanned vertically, the axial response is uniform. For the generator ^{252}Cf-based design, there is more of a fluctuation for the iron and empty matrix due to the stationary source. For the low-density polyethylene matrix, the count rate at the center of the drum increased by 1.5 times for the^{252}Cf shuffler. There was a two-fold increase in count rate for the generator-based design. Note that the vertical scale in Figures 5 and 6 are different. For the high-density polyethylene matrix, the max-to-min count rate ratio for the ^{252}Cf shuffler was 24.5, but was lower, 7.2 for the generator-based design. Although the ^{252}Cf shuffler interrogates with a larger fraction of neutrons above 1 MeV, they are not energetic enough to penetrate the high-density polyethylene matrix. For the generator-based design, although there is a smaller fraction of neutrons above 1 MeV, the neutrons above this threshold are highly energetic, close to 14 MeV.

4 CONCLUSIONS

An optimum configuration, consisting iron and tungsten, produced an interrogating spectrum with a smaller fraction of super-threshold neutrons (>1 MeV), compared to the original design. The fission of ^{238}U is more likely in the^{252}Cf shuffler, but there was and increased spatial variation in the tailored interrogating flux, and the resulting count rate, due to the stationary nature of the neutron generator. The scanned ^{252}Cf source produced a more axially uniform response in all positions and matrices except for the high-density polyethylene matrix. Even though a larger fraction of the ^{252}Cf neutrons were above 1 MeV, they were not as penetrating as the 14-MeV generator neutrons that lost little energy in the tailoring configuration.

References

[1] The 252Californium Shuffler. Los Alamos National Laboratory Application Note, March 1990.

[2] H. O . Menlove and T. W. Crane. A 252 Cf Based Non-destruction Assay System for Fissile Material. In *Nuclear Instruments and Methods*, number 152, pages 549–557, 1978.