Non-destructive assay of radioactive waste
Topical meeting

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MEASUREMENTS OF URANIUM WASTE USING A CALIFORNIA SHUFFLER

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Summary

We describe a passive/active neutron counter (PAN) based on a 252Cf shuffler for 208-L drums. It is a flexible instrument that can be used to measure the nuclear material content of large containers. This instrument is installed in the Portsmouth Gaseous Diffusion Plant in Piketon, Ohio. This paper describes the results of a calibration for iron matrix. For 0 to 100 g 235U, the PAN meets our accuracy goal of 10% and our precision goal of 1% for 100 g 235U. With its passive and active capability, this shuffler addresses future needs for materials control and accountability, and health, safety, and environment. The hardware portion of the counter is a good candidate for transfer to the commercial sector. We plan to focus our future waste assay efforts on developing more sophisticated analysis techniques for this generic hardware, rather than developing customized hardware for each application.

1. INTRODUCTION

Uranium contaminated waste is often packaged in large containers, such as 208-L drums, because process operators find drums more economical to handle than small containers. These drums often contain large quantities of matrix materials and small quantities of uranium. We measure the uranium content of these drums to satisfy nuclear material safeguards and accounting requirements and provide improved criticality safety. Waste does not require measurements of the same accuracy as those performed on the refined product, because waste contains nuclear material in a less desirable form. However, measurements of waste with uncertainties of 10-20% are more useful than measurements of waste with uncertainties of 50-100%, especially for accounting purposes.

Because the drums often contain 1 to 100 g of 235U embedded in 10 to 100 kg of waste, the assay method must be extremely sensitive to the uranium but not sensitive to the matrix components. The passive radiation from the 235U is not adequate for an assay because the neutron signal is weak and the gamma rays do not penetrate well. Therefore, we must use an active technique; preferably one that is less sensitive to the matrix. We give up some sensitivity to the 235U to have a method that is less sensitive to the matrix effects.

Deciphering the signal from mixtures of special nuclear material (SNM) has been a problem, in part, because some facilities have traditionally mixed their waste streams without properly segregating the nuclear material and matrices. Gamma-ray-based measurements can easily distinguish between isotopes, but do not provide accurate measurements of scrap and waste in large containers, unless the matrix and SNM are uniformly distributed, the matrix consists of low-density and low-atomic-number constituents, and the SNM loading is small compared with the matrix. In general, if these conditions are not met, the gamma-ray assay underestimates the uranium contained in the drums.

However, neutron measurements penetrate the contents of the drums, but do not measure the matrix. We must perform the two measurements simultaneously to determine the ratio of 235U to 238U in the sample. The shuffler is based on the principle that the sample chamber is filled with a cadmium source, and we use neutron transport properties to determine the ratio of 235U to 238U in the sample. The design of the shuffler is shown in Figure 2.

We used the shuffler to measure the uranium content of the sample chamber. The instrument is mounted on a 400-kW reactor, and the sample is taken from the reactor core. The shuffler is based on the principle that the sample chamber is filled with a cadmium source, and we use neutron transport properties to determine the ratio of 235U to 238U in the sample. The design of the shuffler is shown in Figure 2.

The Portsmouth shuffler is a good candidate for transfer to the commercial sector. One of our objectives was to develop a method that is less sensitive to the matrix effects. We used the shuffler to measure the uranium content of the sample chamber. The instrument is mounted on a 400-kW reactor, and the sample is taken from the reactor core. The shuffler is based on the principle that the sample chamber is filled with a cadmium source, and we use neutron transport properties to determine the ratio of 235U to 238U in the sample. The design of the shuffler is shown in Figure 2.

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underestimates the uranium content. A low bias in the uranium result is often not acceptable for diversion or criticality safety decisions. If the waste is of high density or contains any metal, active-neutron nondestructive assay techniques must be used. Epithermal or fast neutrons penetrate the contents of the drums better than low-energy $^{235}$U passive gamma rays. However, neutron measurements are not without difficulties. Neutron poisons, ($\alpha$,n) contaminants, multiplication, the inability to distinguish between isotopes, and moderation can significantly bias the measurements. Neutron measurements usually overestimate the amount of uranium, unless one corrects for these effects. Segregating the waste according to its neutron transport properties allows the operator to apply unique calibration standards to each waste category. When these standards have been applied, the $^{252}$Cf shuffler has performed well and has the best potential to assay uranium in large, dense containers. This potential has been confirmed by Monte Carlo calculations.

2. DESCRIPTION OF THE PASSIVE/ACTIVE NEUTRON COUNTER

This paper describes a passive/active neutron (PAN) counter based on a californium shuffler that has been delivered to the Portsmouth Gaseous Diffusion Plant in Piketon, Ohio. A californium shuffler has four major components: the storage module for the californium source, the sample chamber and detectors, the control electronics (including the drive system for the californium source), and the software. Figure 1 is a conceptual drawing of the instrument. The shuffler is based on the principle that about 1% of the neutrons released following fission of $^{252}$Cf come from the fission products with half-lives ranging from a fraction of a second to a few minutes (delayed neutrons). We interrogate the sample for several cycles; in each, the californium is moved close to the sample for 10 s, it is withdrawn, and the delayed neutrons are counted for 7 s. The source storage module shields both the instrument operator and the sample chamber from the californium source neutrons.

We designed this shuffler to have a detection efficiency for fission neutrons of about 20%, large enough to make the counter suitable for passive coincidence counting. We also included a shift-register circuit for passive neutron coincidence counting. To better correct for matrix effects, we use neutron-flux monitors. To correct for isotopic variations, we can use two modes of interrogation. Either thermal or epithermal neutrons can be used in the active interrogation. The epithermal mode is achieved with a steel reflector behind the $^{252}$Cf source and a cadmium liner completely covering the inner surface of the counting chamber. In the thermal mode we add a 2.5-cm-thick polyethylene liner between the drum and the cadmium liner. Dual-energy interrogation allows us to correct for the $^{238}$U effects in samples of unknown uranium isotopic distribution.

The Portsmouth shuffler is improved over previous 208-L drum shufflers in several ways. One of our objectives has been to make the new shuffler so versatile that private companies would find it an attractive instrument to market. We added a dual interrogation mode to determine the ratio of $^{235}$U to $^{238}$U when the uranium enrichment is unknown. The inside of the sample cavity has been completely lined with cadmium to enhance the differences between the two interrogation modes. A better reflector design and higher detection efficiency each contributed 20 to 30% to reducing the source size without decreasing the sensitivity of the measurement. The instrument has more $^3$He detector tubes than earlier designs and faster AMPTEK-based electronics. The polyethylene shielding has been simplified. The source shield is now an integral unit that can be lifted into place with a forklift. The innovative use of borated materials in the source shield eliminated the need for a 2.5-cm-thick lead outer cover on the shield. Figure 2 is a cross section of the (4 ft on a side) source storage cube. The tungsten core absorbs 10% of the neutron flux. More neutrons are captured near the center; consequently this design has more boron there to reduce the intensity of the hydrogen capture gamma rays.
Improvements in the electronics and software make the user interface more pleasant for the operator. The modular software package uses the more versatile and portable C language. This software package will have built-in hardware diagnostic capabilities and automatically archived parameters, results, and data; and will perform more sophisticated data analyses. The computer now performs more of the functions previously left for the operator.

3. RESULTS AND PERFORMANCE

First we measured the passive spatial response of the PAN to a fission source, californium, and a lower-energy neutron source, AmLi. Then we mapped the active response of the shuffler as we moved a small quantity of uranium in the sample chamber. Because of the large sample size, we found it necessary to move the californium source alongside the drum during the 10-s irradiation to get a similar response from a small amount of $^{235}$U placed at the

Fig. 1. The PAN controls diagram. Three major components of the PAN are shown; the sample chamber, the californium source storage module, and the electronics/controls (including those for the californium source transfer). The fourth component is the software.
The surface more pleasant for and portable C language, and automatically sophisticated data analyses. The operator.

to a fission source, calibrates the active response of the chamber. Because of the source alongside the drum, the unit of 235U placed at the top, center, or bottom of the drum. The scan is 33 in. long, with a pause at the top and bottom. In 10 s, three up-and-down scans are performed with a total pause of 4 s. The response to all samples was normalized to 1.000 at the center. Table I lists the detection efficiencies and summarizes the response for the two passive surveys and the active survey. The standard deviation of the response indicates that the chamber's efficiency is relatively flat over the sample volume. The minimum response is typically at the top of the drum.

<table>
<thead>
<tr>
<th>Neutron Source</th>
<th>Detection Efficiency (%)</th>
<th>Center</th>
<th>Average</th>
<th>Std. Dev.</th>
<th>Maximum</th>
<th>Minimum</th>
</tr>
</thead>
<tbody>
<tr>
<td>Californium</td>
<td>17.4</td>
<td>1.00</td>
<td>0.98</td>
<td>0.06</td>
<td>1.08</td>
<td>0.90</td>
</tr>
<tr>
<td>AmLi</td>
<td>19.5</td>
<td>1.00</td>
<td>0.92</td>
<td>0.07</td>
<td>1.04</td>
<td>0.81</td>
</tr>
<tr>
<td>Uranium</td>
<td>--</td>
<td>1.00</td>
<td>1.01</td>
<td>0.04</td>
<td>1.06</td>
<td>0.95</td>
</tr>
</tbody>
</table>
For an iron matrix (130 kg) in a 208-L barrel, the response was flat to about 20%. Using a 300-μg californium source, the active response for this shuffler for 235U is 1.46 counts/(s·g), for 238U it is 0.062 counts/(s·g) and for 239Pu it is 0.73 counts/(s·g). The die-away time is 70 μs and the deadtime is 0.44 + 0.14 T x 10^-6 μs, where T is the total count rate. We have developed matrix drums with three small (3-cm) tubes that run the vertical length of the drum. The tubes are located on the center axis, at one half the radius, and at the outer radius of the drum. The drums can be filled with matrix materials such as iron, graphite, polyethylene, Raschig rings, or alumina, and small vials of dilute or pure uranium compounds can be placed in the tubes. Our experience has indicated that these matrix materials exhibit a range of neutron response effects as large as those found in nuclear facilities. The small vials contain 40 g of alumina in 2.5-cm-diam by 10-cm-high aluminum cylinders. Each vial contains 0.1, 1.0, 5.0, or 10 g of uranium enriched to 10 or 94%. Even 5 g of 94% uranium is sufficiently dilute to not exhibit self-shielding. These vials are placed at various positions in the drum, with up to 21 vials (containing 105 g of uranium) in the chamber at one time.

Figure 3 shows the shuffler’s response as a function of 235U mass for a matrix drum containing 130 kg of iron scrap. A least squares fit using the Deming approach gives the following fit:

\[ \text{Response} = -2.2567 \times 10^{-4} m_{235}^2 + 1.4586 m_{235} + 6.2472 \times 10^{-2} m_{238} \]  

(1)

Note that the nonlinear term in Eq. (1) is 4 orders of magnitude smaller than the linear term. This is demonstrated more clearly in Fig. 4, where the response per gram is plotted as a function of 235U mass. The scatter in the data for Fig. 4 is consistent with the counting statistics.
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diskey is the total count
shakes that run the vertical
half the radius, and at the
materials such as iron,
furred or pure uranium
that these matrix materi-
and in nuclear facilities.
igh aluminum cylinders.
94%. Even 5 g of 94%
ials are placed at various
m) in the chamber at one
t mass for a matrix drum
ning approach\(^3\) gives the

\[ 0.2 m_{238} \quad (1) \]
smaller than the linear term.
gram is plotted as a func-
the counting statistics.

Figure 4. The shuffer response per gram as a function of $^{235}$U mass. Small
vials of 10- and 94%-enriched uranium were placed in a 208-L drum filled
with 130 kg of iron scrap. The multiple data points at 6, 12, 32, and 65 g
$^{235}$U illustrate the effects of measuring the same quantity of uranium at dif-
ferent positions in the drum. The scatter in the points is consistent with
counting statistics.

Figure 5 compares the shuffer response as a function of mass for three sample types. The small sample response curve is independent of enrichment (0.2-93%) and the matrix (graphite, low-enriched oxide, alumina). The small sample response is similar to the lead matrix response discussed above. The alumina matrix has a much higher response per gram of $^{235}$U. We plan to use the flux monitor response to correct for this effect. This barrel contains 400 lb of Al\(_2\)O\(_3\) with a nominal 3 wt% water. The scatter in the alumina data is caused by spatial response variations. The response at the bottom and edge of the drum is much lower than the response over the interior of the drum.

Table II summarizes some preliminary results that show the effects of various matrices on the two flux monitors. A 5-g 94% vial was placed in different locations in the matrix drum to measure the active response. Bare Flux Monitor and Cd Flux Monitor indicate the responses from the bare and cadmium-covered flux monitors, respectively. The number of points indicates how many different positions in the drum were sampled. At a minimum, the center and the upper and lower outside edges were always sampled. The average response column lists the average and standard deviation of the responses from different positions. The variation in the average response standard deviation indicates that it is not easy to achieve uniform detection capability over the entire drum volume. Altering the californium source scan parameters had no substantive effect on the response characteristics for the alumina matrix. It was encouraging to see the flux monitor signals track the average response.

4. CONCLUSIONS

Our initial investigations clearly showed the major source of error when measuring large waste containers is the influence of the samples rather than the sample chamber on the neutron
Fig. 5. The shuffler response as a function of $^{235}$U mass for three sample types. The response from assorted small samples of many enrichments and matrices is consistent with the response from the iron matrix also shown in Figs. 3 and 4. The alumina (Al$_2$O$_3$) matrix demonstrated a higher response per unit mass and much greater spatial sensitivity.

<table>
<thead>
<tr>
<th>Matrix Material</th>
<th>Average Response</th>
<th>Number of Points</th>
<th>Bare Flux Monitor</th>
<th>Cd Flux Monitor</th>
</tr>
</thead>
<tbody>
<tr>
<td>None</td>
<td>1.01 ± 0.04</td>
<td>6</td>
<td>1.00</td>
<td>1.00</td>
</tr>
<tr>
<td>Iron</td>
<td>1.12 ± 0.17</td>
<td>3</td>
<td>1.16</td>
<td>1.12</td>
</tr>
<tr>
<td>Alumina</td>
<td>6.70 ± 2.5</td>
<td>21</td>
<td>1.67</td>
<td>1.41</td>
</tr>
<tr>
<td>Graphite</td>
<td>3.68 ± 1.44</td>
<td>6</td>
<td>1.44</td>
<td>1.40</td>
</tr>
<tr>
<td>12-lb poly</td>
<td>2.26 ± 0.65</td>
<td>6</td>
<td>1.24</td>
<td>1.14</td>
</tr>
<tr>
<td>Poly chunks</td>
<td>4.06 ± 1.59</td>
<td>6</td>
<td>2.02</td>
<td>1.08</td>
</tr>
</tbody>
</table>

We are transferring the shuffler technology to the commercial sector. We have developed active analysis procedures for two sample types that yield measurements accuracies of 10%. We plan to focus our future waste assay efforts (on large containers) on developing more sophisticated analysis techniques based on this generic hardware, instead.
developing customized hardware for each application. With its passive or active capability, this instrument can measure SNM in a variety of matrices and containers.

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REFERENCES

