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EPITHERMAL INTERROGATION OF FISSILE WASTE*

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ABSTRACT

Self-shielding of interrogating thermal neutrons in "lumps" of fissile material can be a major source of error in transuranic waste assay using the widely employed differential dieaway technique. We are developing a new instrument, the combined thermal/epithermal neutron (CTEN) interrogation instrument to detect the occurrence of self-shielding and mitigate its effects. Neutrons from a pulsed 14-Mev neutron generator are moderated in the graphite walls of the CTEN instrument to provide an interrogating flux of epithermal (60-800μs) and thermal (800-2800 μs) neutrons. The induced prompt fission neutrons are detected in ^{4}He proportional counters as a function of time after the generator pulse; these distributions of ^{4}He detector counts differ markedly for plutonium and uranium. We report the results of measurements made with the CTEN instrument, using minimal and highly self-shielding plutonium and uranium sources in 55-gal. drums containing a variety of mock waste matrices. Fissile isotopes and waste forms for which the method is most applicable, and limitations associated with the hydrogen content of the waste package/matrix are described..

INTRODUCTION

The assay of fissile waste in large containers, such as 55-gal drums, is subject to several potentially large sources of interference. In the widely-used DDT (differential dieaway technique) active assay method, the interrogating thermal neutrons can be highly attenuated in the outer layers of plutonium or uranium "lumps," resulting in severe underestimates of their mass. This effect is termed self shielding. Monte Carlo calculations [1] of self shielding in a sphere of U_3O_8, containing 100-g of ^{235}U (93% enrichment), indicate that the expected fission yield is only 11% of that which would be obtained if the uranium were widely dispersed in a non-self-shielding configuration. Smaller lumps are less affected, but the effect can be large even for gram-sized lumps of oxides and sub-gram lumps of metals.

The effect in plutonium is even larger than in uranium, but for gram quantities of plutonium a passive coincidence measurement can often provide a more accurate assay than the active interrogation measurement. A passive assay result that is significantly larger than the active assay value can serve as an indicator that self-shielding is occurring in the active measurement. For uranium measurements, however, there is no passive signal, and, therefore, no way of knowing that self-shielding is occurring in the active measurement.

A neutron-based assay instrument that can perform the standard DDT instrument assay, but which has several additional capabilities, has been designed and built at the Los Alamos National Laboratory. One of the new features, epithermal neutron interrogation [2], is intended to detect and mitigate the self-shielding effect. This CTEN (Combined Thermal/Epithermal Neutron) instrument interrogates with both thermal and epithermal

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neutrons. Because the fission cross sections for neutrons on uranium and plutonium generally decrease with increasing energy above the thermal region, epithermal neutrons of sufficiently high energy will be able to more uniformly penetrate lumps of plutonium and uranium, and, thus, should provide for more accurate assay results if such lumps are present in the waste. However, the composition of the waste matrix and other factors, such as reduced detection sensitivity, will limit the applicability of the epithermal interrogation feature. The studies described below are intended to elucidate the characteristics and limitations of the CTEN epithermal interrogation technique as applied to large fissile waste containers.

EPITHERMAL INTERROGATION

The interrogating neutrons are supplied by a small, pulsed, 14-Mev neutron generator**, located in one corner of the assay cavity (Figure 1). It operates at 100 pulses per second, with a nominal output of 10^6 neutrons per 20-μs-wide pulse. Many of these neutrons are quickly moderated in the 12-inch-thick graphite inner walls of the cavity and in the waste matrix of the 55-gal waste drum. (Some polyethylene shielding/detector components, which contain hydrogen, are located in the walls, and they also contribute to the moderating process.) The moderated neutrons reach epithermal energies of a few eV or less in about 50 microseconds and, if no significant amounts of moderators or absorbers are present in the waste matrix, they then continue to be reduced in energy and intensity with a dieaway half-time (as measured with a cadmium covered He-3 detector) of 34 μs. After approximately 400 μs, the interrogating flux is no longer detectable with a Cd-covered cavity flux monitor, but the flux takes several hundred more microseconds to reach thermal equilibrium. The thermal interrogating flux subsequently dies-away with a half-time of approximately 420 μs.

However, if significant amounts of moderators or absorbers are in the waste matrix, these die-away characteristics can be greatly changed. Of obvious concern for epithermal interrogation would be the presence of significant quantities of hydrogen in the drum. Hydrogen-containing moderators (and some other low-Z materials) have the effect of more rapidly thermalizing the interrogating flux, reducing the time that epithermal neutrons are effectively present.

After the neutron pulse, He-4 proportional counters, located immediately inside the graphite walls, recover from the initial burst of high-energy interrogating neutrons in a time period of about 60 μs from the start of neutron production. These detectors are then able to detect prompt fission neutrons without interference from the low-energy interrogating neutrons that are still present in the cavity. The signals from the ^{4}He detectors are routed to five time-gated scalers, with time windows as shown in Table 1. In addition, the signals are routed to the PATRM [3], a list-mode neutron counter; these data are subsequently analyzed in software to form multichannel scaler spectra, thus providing a record of counts versus time throughout the interrogation cycle.

Table 1. Gated Scaler Time Regions. (Times measured from the start of neutron production.)

Scaler #.	Time (μs)	Interrogation Region
1	60-100	1st epithermal
2	100-240	2nd epithermal
3	240-800	3rd epithermal
4	800-2800	thermal
5	5800-9800	background

Several flux monitors are used to monitor the neutron generator output and the interrogating thermal and epi-cadmium neutron fluxes. In addition, a large number of Cd-covered ^{3}He detectors are used to record DDT-type data in windows 4 and 5, after their recover period of about 800 μs; these data do not contain prompt-fission epithermal interrogation information and are not discussed in this paper.

EXPERIMENTAL RESULTS

No Interfering Matrix

Figure 2a shows the raw counts obtained with the ^{4}He detectors as a function of time, from 40 to 800 μs after the start of the neutron generator pulse, for two enriched uranium (93% ^{235}U) sources. The sources were located in the center of an empty, 55-gal steel drum, centered in the CTEN assay chamber. Data from a background measurement,

** Model A210 TTA, manufactured by M F Physics Corp., Colorado Springs, CO.

obtained with no source present in the drum, is also shown.

Figure 2b shows the same data for the two sources, with background subtracted from both sets of data and the curves normalized to the same average value for the thermal interrogation region (from 800 to 2000 μs). Data obtained before 60 μs is not shown, as the large background from the initial interrogating neutron burst generally results in poor precision for the net counts. (See the earliest background data point in Fig 2a.) Figures 3-8 have all had background subtracted and have been normalized to the same average value for the thermal interrogation region as Figure 2b.

In interpreting these types of data plots, it should be borne in mind that the responses are functions of the fission cross section, self shielding, and neutron energy, as well as the interrogating flux intensity, which is decreasing rapidly with time.

The top curve in Figure 2b was obtained with four 10-g uranium metal spheres, a highly self-shielding configuration, while the bottom curve was obtained with the same amount of uranium, but contained in a 3-mil-thick metal foil. The latter configuration gives rise to a relatively small amount of self shielding. It can be seen that the largest difference in response (after normalization to the thermal region) occurs at the earliest times, when the epithermal neutrons have the highest average energy. As the energy decreases (increasing time), the difference in response decreases until about 700 μs when the curves appear to coalesce. Qualitatively, at least, this is the expected behavior, in view of the approximate 1/v dependence of the ^{235}U(n,f) cross section with neutron energies in the region below one eV, and the dispersed energy spectrum of the interrogating neutrons that is expected in the "slowing down" process in the large CTEN chamber.

Data obtained in a similar fashion for a series of increasingly thick uranium sources is shown in Figure 3. The bottom three curves were obtained with the same set of foils arranged in different configurations to vary the self-shielding effect; the top curve is for the four 10-g spheres described above. It can be seen that as self shielding increases, the response in the early time regions increases relative to the response in the thermal interrogation region, where all curves have been normalized.

Figure 4 shows the time dependent data obtained for "thick" and "thin" (highly and minimally self-shielding, respectively) plutonium sources, in the center of the CTEN assay chamber. The minimally self-shielding source is 7-g low-burnup Pu, as finely dispersed plutonium oxide mixed with diatomaceous earth in a can of 10-cm-diameter x 28-cm-height. The highly self-shielding Pu consists of four metal chunks, with a total low-burnup Pu mass of 79 g, in an identical can with diatomaceous earth filling.

It can be seen that the shape of the thin Pu curve is markedly different from those for the thin U curve described above in Fig 2b. At the earliest times after the generator pulse, the thin Pu response is well below that for the thick Pu, but then quickly rises somewhat above the thick curve at about 150 μs. It remains above until approx. 250 μs when it falls below the thick curve and stays significantly below until the thermal region is reached. This behavior can be explained qualitatively as due to the large resonance in the ^{239}Pu(n,f) cross section [4] at about 0.3 eV, which is superimposed on the typical 1/v cross section shape. Thus, at epithermal energies somewhat above thermal equilibrium, the fission cross section is actually higher than at thermal energies, and self shielding is worse! Only at energies above this resonance does the cross section drop enough to observe a large decrease in self shielding.

The difference between the thin uranium and thin plutonium responses can most readily be seen it Figure 5, with responses obtained with 7-g of each material. Of sources available for use on this project, these had the least self-shielding to thermal neutrons, but there is some. Roughly, the Pu source gives a response that is about 90% of that expected from a source of the same mass with no self shielding, and the corresponding value for the uranium foil is estimated to be about 80%.

Effects of Moderators

The curves shown in Figure 6 were obtained with uranium sources inside two steel 55-gallon drums, one with a moderate-density, 90-mil polyethylene liner and the other filled with polyethylene shavings. The steel drums themselves have negligible effects on the response curves, but it can be seen that the liner reduces the difference in response between thick and thin sources, and the polyethylene shavings have a larger effect. But, even in the latter case, the difference between thin and thick U sources is readily apparent.

Figure 7 shows the effect of the same polyethylene liner on plutonium measurements. Compared to the curves in Figure 4, it is apparent that the liner has greatly reduced the differences between thin and thick sources during the earliest time regions. In fact, the only significant difference at early times is in the first time bin (60-80 μs). But, there is still a significant difference between the thin and thick curves in the region from about 240-800 μs, corresponding to scaler 3. In fact, the difference in the relative total counts in this region is just as large (approx. 10%) as for the no-liner case.

However, when the thin versus thick comparison is made for the drum containing the polyethylene shavings (data not shown here) the curves are indistinguishable across the entire range of times; self-shielding in plutonium cannot be detected in plutonium at such hydrogen loadings.

Effects of Absorbers

Generally, it appears that absorbers are less detrimental to the epithermal interrogation technique than moderators. Figure 8 shows data for uranium buried in a drum of iron scrap. Though the iron significantly absorbs the interrogating flux (as evidenced by flux monitor data not shown here), the curves indicate that self-shielding in this case is still readily observable. Additional measurements were made in the iron scrap drum at different positions, to determine if the degree of self shielding was dependent on source location. These results for the time-gated scalers described in Table 1 are summarized below in Table 2.

Table 2. Response Ratio of Thick to Thin U in Iron Scrap After Normalization to Scaler 4

Position (cm) h=height r=radius	Scaler 1	Scaler 2	Scaler 3
r=25, h=8	4.3	2.6	1.3
r=20, h=56	5.5	2.8	1.5
r=12, h=40	5.5	2.9	1.4

The scaler 1 result for the first row is statistically significantly different from the other two scaler 1 values, while all scaler 2 and 3 values are internally consistent within the calculated statistical precision. This shows that for this waste form, at least, positional effects would be relatively unimportant in determining the degree of self shielding, compared with all the many other uncertainties associated with waste measurements in 55-gallon drums. Scaler 1 has the poorest precision since it has the fewest counts and has a relatively large background, due to the initial neutron burst. Generally, scaler 3 has the best precision of all the scalers, as it has the highest counts and a low background. In fact, both scaler 2 and 3 usually have higher counts than scaler 4, which is the scaler used for detecting thermally induced fission neutrons with He-4 detectors as described here and with Cd-shielded He-3 detectors in the standard DDT/PAN method.

DISCUSSION AND CONCLUSIONS

These data and analyses clearly demonstrate that the CTEN instrument is capable of detecting the occurrence of self-shielding in 55-gallon drums of waste, if certain criteria are met. First, the waste can only have a limited amount of hydrogen and other moderators. As demonstrated, self shielding can still be observed in drums with polyethylene liners, but it is more difficult to do so, especially for Pu. Assay of waste in drums without liners is more amenable to detection of self shielding. However, many waste forms have significant amounts of hydrogen in the waste matrix, which would interfere with the epithermal interrogation, especially for plutonium measurements.

This method of detecting self-shielding will only be useful when sufficient amounts of plutonium or uranium are present, at least several grams and, more practically, tens of grams. Certainly, the epithermal interrogation method is not applicable to drums with fissile loadings near the "TRU decision level" of 100 nCi/g, which corresponds to a few hundred mg or less of low-burnup plutonium in typical waste drums.. However, this method could be most useful for drums containing uranium near the criticality limit of 200g/drum for shipment to the TRU waste depository (Waste Isolation Pilot Plant). For this high-loading situation, self-shielding is more likely to occur, and cannot be detected in uranium by other currently available means.

Another problem that will arise when making measurements on real waste is the need to know enough about the waste form to predict (through Monte Carlo calculations or measurements on mock waste drums) the relative response of thin and thick Pu or U in the various scaler windows. This implies that "matrix-specific" calibrations are required for the epithermal measurements. It may be possible, however, to use the various flux monitors in the CTEN instrument to predict the expected epithermal behavior; this is an area of current research.

It is apparent that the "best" epithermal information is obtained at the earliest times, when the average energy of the interrogating flux is highest. Two factors currently prevent obtaining reliable data at times earlier than 60 μs, and also result in a significant background being present until 80 - 100 microseconds after the start of neutron production. The first relates to the apparent interrogation pulse shape from the neutron generator. Though nominally only some 20-μs wide, at appears that a very small second pulse is also present some 40 to 50 microseconds after the first pulse. This pulse, thus, directly interferes with early time measurements and appears to contribute to the undesirably high background observed in Scaler 1. This second pulse has been observed from several neutron generator systems of the type typically used on DDT/PAN instruments; the generator in use now was found to give the least interference of the generators available for testing. The second factor is the undesirably long time the neutron detector and associated electronics takes to recover from the large initial burst of neutrons from the generator. An electronics package that may mitigate the second problem is currently being built, but a fix for the first problem appears to be outside the scope of the current project.

While this CTEN instrument was designed and tested for large containers of waste it should be noted that the general technique could be applied to several "safeguards" related assay problems. The CTEN instrument is currently being used to investigate the detection of "contraband" SNM in packages and waste containers. Smaller versions of the CTEN instrument could be used to assay spent fuel or more compact fissile objects, where interference from hydrogen and other effects from waste matrices would not be significant. Smaller cavities and other design modifications could reduce the energy spread in the interrogating flux, while sharper, shorter duration neutron pulses, from sources such as electron linear accelerators, could increase the epithermal interrogation information available at early times following interrogation.

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