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TITLE A PASSIVE-ACTIVE NEUTRON DEVICE FOR ASSAYING REMOTE-HANDLED TRANSURANIC WASTE

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A PASSIVE-ACTIVE NEUTRON DEVICE FOR ASSAYING  
REMOTE-HANDLED TRANSURANIC WASTE

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Summary

A combined passive-active neutron assay device was constructed for assaying remote-handled transuranic waste. A study of matrix and source position effects in active assays showed that a knowledge of the source position alone is not sufficient to correct for position-related errors in highly moderating or absorbing matrices. An alternate function for the active assay of solid fuel pellets was derived, although the efficacy of this approach remains to be established.

1. INTRODUCTION

We have developed and constructed a passive-active neutron (PAN) assay device for certifying remote-handled transuranic (RH-TRU) waste for eventual emplacement at the Waste Isolation Pilot Plant (WIPP) near Carlsbad, New Mexico. This device is essentially a smaller, detector-shielded version of earlier PAN assay devices built at Los Alamos National Laboratory [1], combining the active differential die-away technique (DDT) with passive neutron coincidence counting to yield two separate but complimentary fissile mass measurements. Because of design differences and the difficult nature of the RH-TRU wastes to be assayed, the so-called "second-generation" matrix effects correction algorithm [2] used in the earlier Los Alamos PAN devices was not used with this device.

According to WIPP criteria, RH-TRU waste is defined as transuranic waste with an exposure rate of more than 0.2 R/h at the surface of the container. As the name implies, such waste must be handled remotely to ensure that workers are not exposed to excessive levels of radiation. The RH-TRU waste generated at Los Alamos consists mainly of irradiated breeder-reactor fuel pellets containing mixtures of uranium and plutonium of varying isotopic composition and degree of burn-up. These have been reduced in metallurgical studies to sections of fuel pellets and to grindings and cuttings from the pellets. The composition of this waste, therefore, varies from absorbed solutions to gram-size lumps of fissile material, in various matrices. Before assaying, the 3.8-L cans of waste are sealed inside 21-cm-diam by 30-cm-high steel cans. The surface gamma-ray exposure rates of these cans can be as high as 1000 R/h. Because of these high exposure rates, the assay device was designed to be operated inside a hot cell.

The principal design modification required for assaying RH-TRU waste was the addition of 15 cm of lead shielding in front of the detectors to attenuate the intense gamma-ray fields from the waste. The  $^3\text{He}$  proportional counters used in PAN assay devices detect low-energy neutrons via the  $^3\text{He}(n,p)^3\text{H}$  reaction, which produces a charge pulse significantly larger than those produced by gamma rays. When assaying

contact-handled TRU waste, this allows nearly complete discrimination against gamma radiation. Unfortunately, the gamma-ray rates from the RH-TRU waste are so high that without heavy shielding of the detectors, pulse pileup can defeat the discrimination.

Because it is necessary to place the sample as close as possible to the "on-can" flux monitor (see Sec. 2) to get a useful matrix effects correction, we were concerned that the cadmium shielding around that detector would locally depress the thermal flux and that a uniform interrogation could only be obtained by raising the sample up some distance from the on-can detector. Therefore, in addition to developing a (uniform) matrix effects correction for the DDT assays, we have tested whether taking DDT measurements with the waste container in two different positions can significantly improve the assay accuracy.

## 2. DEVICE DESIGN

Figures 1a and 1b show the schematic design of our RH-PAN device. Its dimensions are 142 by 142 cm by 86 cm high, and it is constructed mostly of lead, polyethylene, and graphite, with steel and aluminum supports (not shown). The neutron source for DDT assays is a sealed 14-MeV neutron generator (zetatron). The upper and lower parts of the moderating cavity are packed with additional graphite not shown in the figures, but indicated by the dashed lines in Fig. 1a.

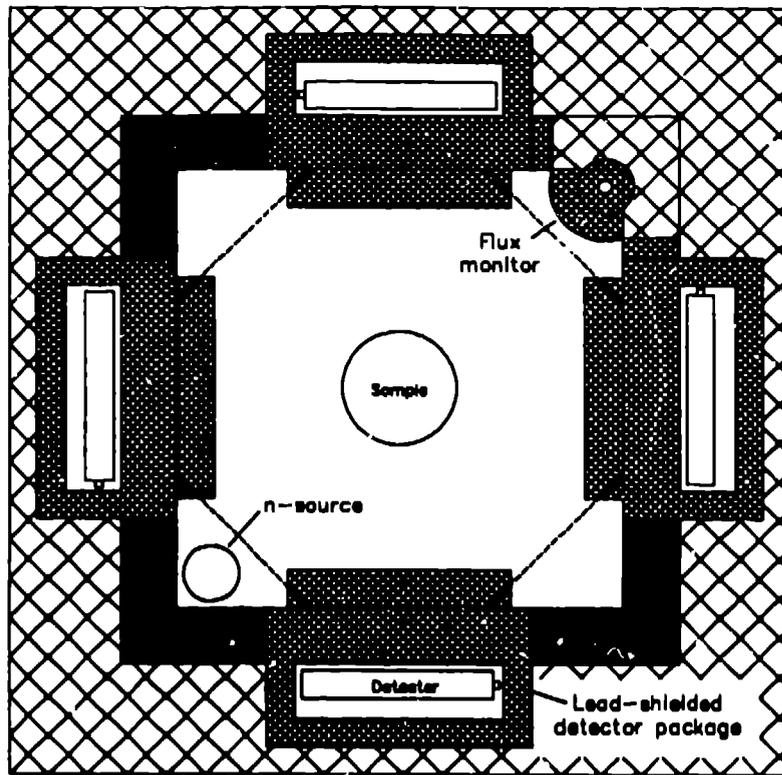
Imbedded in each of the four vertical walls is a pair of  $^3\text{He}$  detectors: one bare and one shielded (against thermal neutrons). The shielded detectors are surrounded inside-to-outside with 1.3 cm of polyethylene, 0.16 cm of cadmium, and 0.32 cm of borated rubber. The shielded detectors provide the basic signal for the DDT assay. The bare and shielded detector counts are summed for passive neutron coincidence counting. The total system efficiency (bare plus shielded) for counting neutrons from  $^{240}\text{Pu}$  is approximately 1.1%.

A  $^3\text{He}$  detector mounted in the corner of the cavity opposite the neutron generator monitors the flux for DDT assays. To first order, this detector gives a count rate proportional to the thermal neutron flux inside the cavity, and is used for normalization. Imbedded in the floor of the device is a partially cadmium-shielded, boron-lined proportional counter, referred to as the "on-can" flux monitor. The cadmium shielding, which was carefully designed to minimize the amount of cadmium used, blocks thermalized neutrons entering from the sides but is open to neutrons entering from above. To first order, this detector measures the thermal neutron flux inside the waste container and is used to correct for matrix effects.

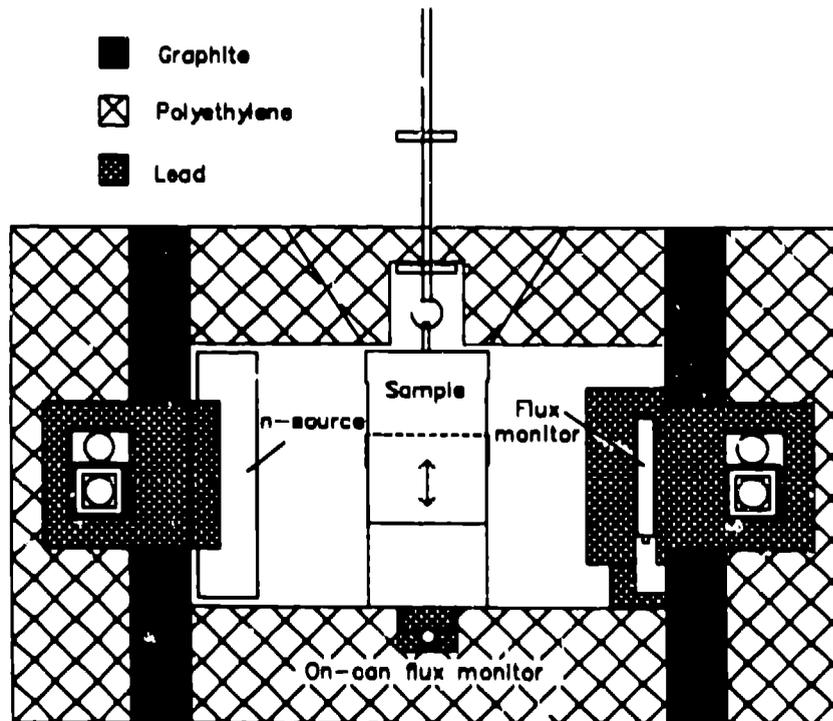
The polyethylene lid to the chamber is suspended from above (not shown) by the chain of a remotely-operated crane motor that is mounted on a sliding track. To allow for reproducible dual-positioning of the sample can, the lid is attached to a sliding rod with adjustable stops. Figure 1b shows the two measurement positions for the sample, referred to here as "up" (solid lines) and "down" (dashed lines). The sample can attaches to the end of the sliding rod by a small hook.

## 3. EXPERIMENTAL METHOD

We made a series of measurements using real and mockup waste cans to characterize the active and passive systems. To derive a matrix effect correction for the active assay and to study the use of two measurement positions, over forty (4000-pulse) DDT assays were performed in both the



(a)



(b)

FIGURE 1. (a) Top cut-away view of the RH assay device. The dashed lines indicate the location of additional graphite at the top and bottom of the unit. (b) Side cut-away view of the device indicating the up and down measurement positions for the sample by solid and dashed lines.

up and down positions on an 8.67-g, 0.005-cm-thick enriched  $^{235}\text{U}$  foil (93%  $^{235}\text{U}$ ). This was a fairly large foil, having a surface area of  $\sim 90\text{ cm}^2$ . The foil was placed at various positions and orientations within the container, in matrices ranging from no-matrix to closely-packed high density polyethylene blocks (highly moderating matrix) to Borax mixed with vermiculite (highly absorbing matrix). Some of the measurements were made with the  $^{235}\text{U}$  foil flat against the bottom or top of the can. Because such position extremes are unlikely with real waste, we have excluded those data points when deriving a function to estimate correction factors and in our evaluation of the dual-position method. They are useful, however, in making qualitative comparisons.

To determine an alternative mass formula valid for fuel pellets, DDT measurements were made on a set of 13 stainless steel cylinders (each 0.9 cm in diameter by 5.1 cm long) containing unirradiated fuel pellets with diameters from 0.67 cm to 0.82 cm and lengths from 0.64 cm to 0.88 cm. The composition of the fissile material in the cylinders was  $\sim 20\%$  plutonium and  $75\%$  uranium of varying isotopic ratios. The cylinder contents fell into three categories:

- 1) high burn-up plutonium (12%  $^{240}\text{Pu}$ ) with depleted uranium;
- 2) low burn-up plutonium (6%  $^{240}\text{Pu}$ ) with moderately enriched uranium (40%  $^{235}\text{U}$ ); and
- 3) low burn-up plutonium (6%  $^{240}\text{Pu}$ ) with highly enriched uranium (93%  $^{235}\text{U}$ ).

(The isotopic percentages are relative to the element.) Active measurements were made on single cylinders and on combinations of cylinders, with fissile masses ranging from 2 to 180 g. (Unless otherwise stated, fissile masses are expressed as the equivalent mass of low burn-up plutonium, i.e., as 94%  $^{239}\text{Pu}$  and 6%  $^{240}\text{Pu}$ . For this reason, what we are calling the "true" masses of the mockup samples will generally be different for passive and active assays.) The measurements were made in matrices varying from no-matrix, to high-density polyethylene blocks (highly moderating), to steel blocks (highly absorbing).

Following the DDT assays, the non-irradiated fuel pellet mockup samples were assayed using 900-s and 30000-s (overnight) passive coincidence counts. These data were used to characterize the passive system.

All data were collected in an IBM AT-based system using four Ortec ACE multichannel scaler cards under the control of computer software developed at Los Alamos National Laboratory [4].

#### 4. RESULTS AND DISCUSSION

##### 4.1 Active results

##### 4.1.1 Correction for matrix and position effects

Figure 2 shows the measured shielded-detector response in the down position vs the on-can response in the down position for DDT assays of the  $^{235}\text{U}$  foil. The foil positions are indicated in the figure by different symbols. The background-corrected shielded and on-can responses were normalized to the corresponding flux monitor counts, and the shielded response was further normalized so that a value of 1.0 was obtained for the case in which the foil was centered in the can with no matrix present. Because the same mass was measured in each case, with this normalization the inverse of the shielded response can be taken as

the active correction factor. Figure 3 shows the shielded detector response in the up position vs the on-can response in the down position for the same mockup cans measured in Fig. 2.

Figure 2 shows that the correction factor required for the down measurement with a given matrix is smallest when the fissile material is near the top of the can, and largest when the material is near the bottom or at the center. By contrast, Fig. 3 shows that the correction factor required for the up measurement is smallest when the material is either near the top or the bottom of the can, and largest when the

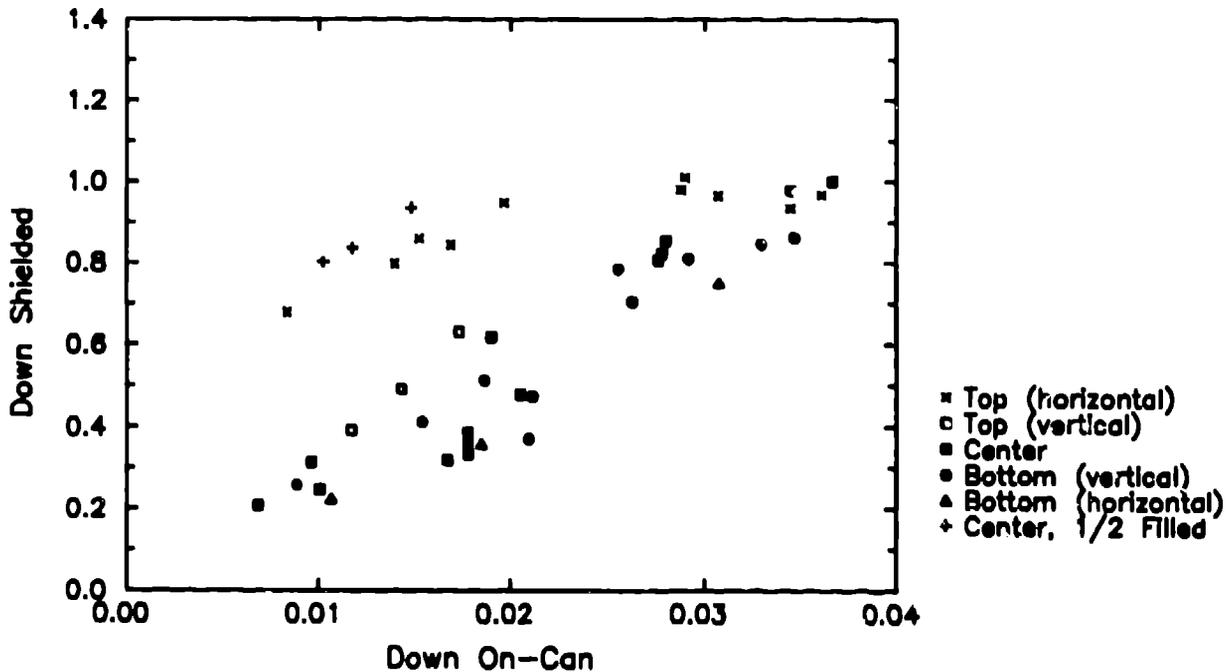


FIGURE 2. The down shielded detector response vs the down on-can response for active assays of the  $^{235}\text{U}$  foil. The down shielded response is normalized to equal 1.0 for the indicated reference point, which represents the no-matrix case with the foil centered in the can.

material is in the center. This demonstrates that the sample receives a more uniform interrogation when the can is in the up position, and confirms our hypothesis that the on-can flux monitor causes a significant local depression of the thermal flux.

Because the up and down measurements respond differently as a function of the foil position, one might suppose that position of the foil could be (approximately) determined by taking the ratio of the two measurements. This is verified in Fig. 4, which shows the ratio of the down-to-up shielded responses vs the approximate height of the foil within the canister. Although there is a great deal of scatter in the data, the general trend is that the down-shielded to up-shielded ratio increases more-or-less linearly with the height of the foil.

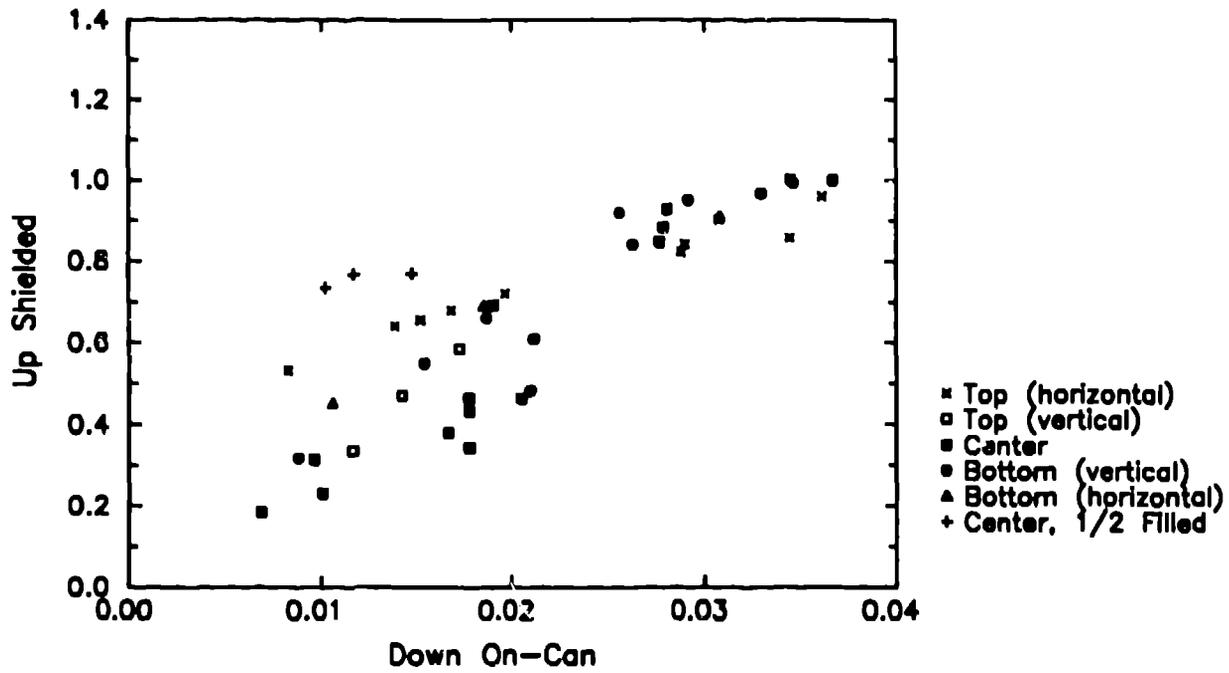


FIGURE 3. The up shielded detector response vs the down on-can response for active assays of the  $^{235}\text{U}$  foil.

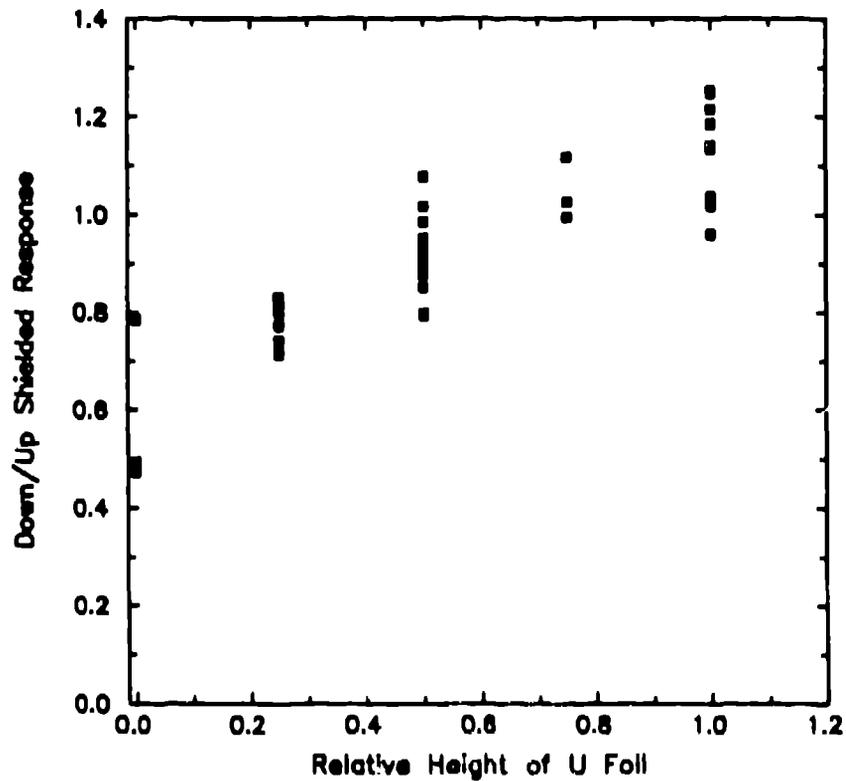


FIGURE 4. The ratio of the down shielded response to the up shielded response for active assays of the  $^{235}\text{U}$  foil vs the approximate height of the foil in the waste can.

For the down measurement alone, the active mass is given by

$$MA = a_1 \cdot SHD \cdot CF \quad , \quad (1)$$

where MA is the active fissile mass, SHD is the normalized down-shielded response,  $a_1$  is a constant calibration factor, and CF is the matrix effect correction. In the simplest idealization, the correction factor, CF, should be proportional to the inverse of the down on-can response. Caldwell, et al., [2] used a particular exponential function of the on-can (barrel) flux monitor response for an absorber correction in their "second-generation" approach. Because we are using the same variable to correct for both absorber and moderator effects, we have not restricted ourselves to their function. Instead, we have tested several functions. The one that seems to give the best results on the  $^{235}\text{U}$  foil data in both Figs. 3 and 4 (excluding the extreme position cases) is

$$CF = \text{constant} \cdot \text{ONCAN}^{a_2} \quad , \quad (2)$$

where ONCAN is the down on-can response, and  $a_2$  is a constant to be determined.

To test whether an additional measurement in the up position can improve the results, we have compared results from Eq. 1 with those obtained using the more general function,

$$MA = (a_3 \cdot SHD + a_4 \cdot SHU) \cdot CF \quad , \quad (3)$$

where SHU is the shielded response in the up position, and  $a_3$  and  $a_4$  are constants to be determined. Note that the weighted average in Eq. 3 represents the only way to combine SHU and SHD that is additive for multiple sources. That is, to the extent that Eq. 3 works as a position correction for point sources, it is also guaranteed to work on distributed sources.

Figure 5 shows the ratios of the calculated mass to the true mass as a function of the down on-can response, computed for both the single- and double-measurement approaches. The coefficients  $a_1$  were determined with an unweighted, iterative least-squares fitting procedure. Thus, the mass formula for the single-measurement approach is

$$MA = 2.65 \cdot SHD \cdot \text{ONCAN}^{-.93} \quad ; \text{and} \quad (4)$$

the mass formula found for the double-measurement approach is

$$MA = 1.82 \cdot (0.124 \cdot SHD + 0.876 \cdot SHU) \cdot \text{ONCAN}^{-1.0} \quad . \quad (5)$$

To evaluate the goodness-of-fit of Eqs. 4 and 5, we can compare the standard deviations in the final, calculated masses. With the single measurement, the standard deviation in the mass is 16.4%. This is much larger than the 1-3% error that would be expected based on counting statistics alone. With the double measurement, the standard deviation in the mass is 15.2%, only slightly better than we obtained for the single-measurement approach. Based on this, we have decided against routinely using the double-measurement approach with this device, because the benefit is too slight to justify the extra assay time.

That the double-measurement approach did not give significantly better results than the single-measurement approach suggests that the position-related errors are less important than originally supposed. This may be due to the small size of the our RH cans relative to the source-detector distances (compared with the larger, contact-handled PAN assay units) and to the relatively large size of the uranium foil. If so, then it would be unwise to generalize our results to the larger

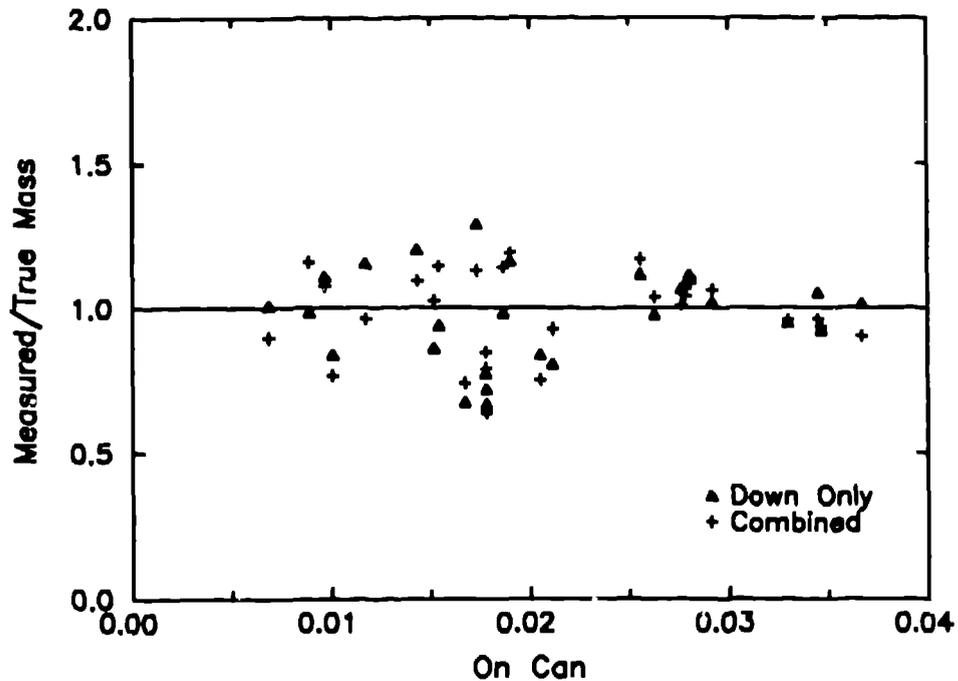


FIGURE 5. The ratio of the measured to true fissile mass for active assays of the  $^{235}\text{U}$  foil vs the down on-can response.

units. Another explanation is that it is not the absolute position of the source that is significant, but the position of the source relative to whatever absorbing or moderating materials are present. This view is supported by DDT measurements made with the can filled only halfway with matrix material, and the  $^{235}\text{U}$  foil placed flat on top of the matrix. Three such measurements are shown in Figs. 2 and 3. The assay values are approximately the same as when the can is completely filled with matrix and the foil is laid flat on the top. (These data points were not used in fitting Eqs. 4 and 5.)

It has been observed in other DDT systems that the DDT assay error generally increases with the correction factor [2]. In Fig. 5, our errors seem to be largest for intermediate values of the correction factor. This problem can be attributed to the higher uniformity of those matrices that required the largest correction factors (mixtures of vermiculite and Borax). The group of points with the highest scatter (between ONCAN = 0.012 and 0.02) had matrices of stacked blocks (2.5- to 5-cm edges) of polyethylene and iron. While these were stacked as tightly as possible, there were necessarily some gaps.

We estimate that the lower limit of detection (4 times sigma of background) for DDT assays with this device under ideal conditions is ~50 mg of enriched uranium for a 4000-pulse (80-s) irradiation.

#### 4.1.2 Alternate mass function for fuel pellets

Some of the Los Alamos RH-TRU waste will contain intact or large segments of fuel pellets. Such waste is difficult to assay using the DDT method because the fissile material is in the form of lumps, and only the outer skin can be penetrated by the interrogating thermal neutron flux. We have derived an alternate function for estimating the fissile mass when it is known that only fuel pellets are present, based on the assumption that the geometry of the pellets is regular enough that approximately the same amount of self-shielding occurs with each pellet. To the extent that this approach works, it can be applied to wastes of unknown composition to estimate the maximum fissile content.

To determine the alternate mass function, DDT measurements were made on the mockup samples of non-irradiated fuel pellets in various matrices (see Sec. 3). Figure 6 shows the measured mass of the various fuel samples (calculated from Eq. 4) vs the true fissile mass. The formula used to fit this data (the curve in Fig. 6) was

$$MA = 0.47 \cdot M^{2/3} ,$$

which gives the alternate function,

$$MPELLET = 3.1 \cdot MA^{1.5} , \tag{6}$$

where MA is the mass calculated for the normal case, M is the true mass, and MPELLET is the estimated fuel pellet mass. The standard deviation in the ratio of the measured-to-true mass is 48%. Because of the large amount of scatter in Fig. 6, it is difficult to judge how well the function actually fits our fuel pellet data.

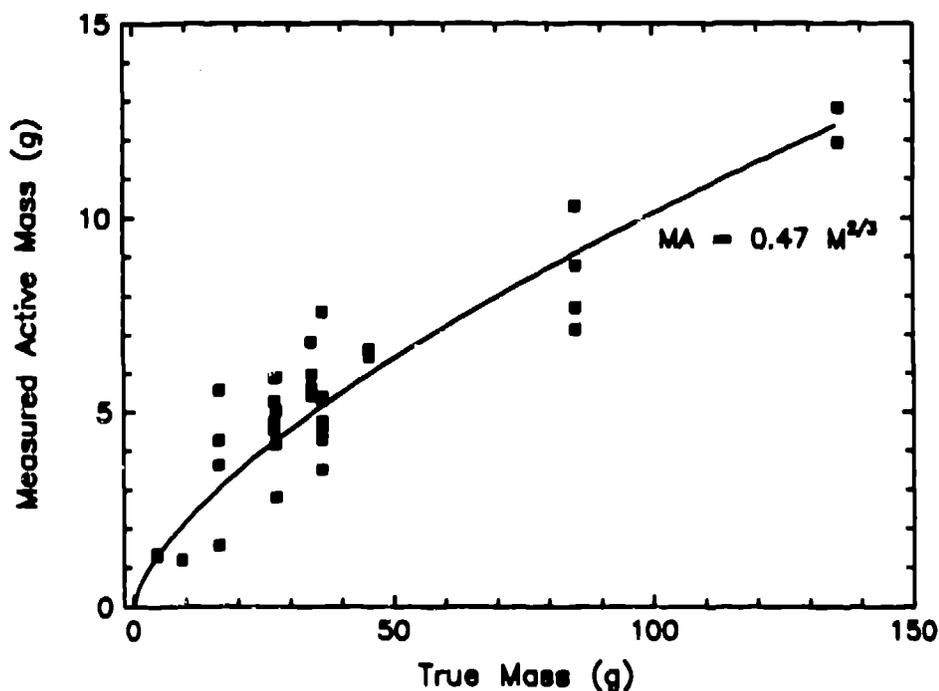


FIGURE 6. The measured active fissile mass of the mockup fuel pellet samples vs the true fissile mass.

#### 4.2 Passive results

The same mockup fuel pellet samples used above to obtain the alternate DDT calibration were also used to characterize the passive neutron coincidence system. Figure 7 shows the measured passive mass vs the true mass of the samples. The measured masses are calculated from the coincidence rate using the formula

$$MP = 408 \cdot \text{RATE} \quad (7)$$

where RATE is the net true coincidence rate (counts/second) and MP is the passive mass. Because of the low efficiency of the system, the statistical (counting) errors on the calculated masses are relatively large. For the data in Fig. 7, the counting error averages 26% for the 900-s counts and 7% for the 30000-s counts. The scatter in the data in Fig. 7 can be mostly accounted for by this error. The sensitivity of the passive assay system is ~10 g for the 900-s counts and falls to approximately 2 g for the 30000-s counts (overnight counts).

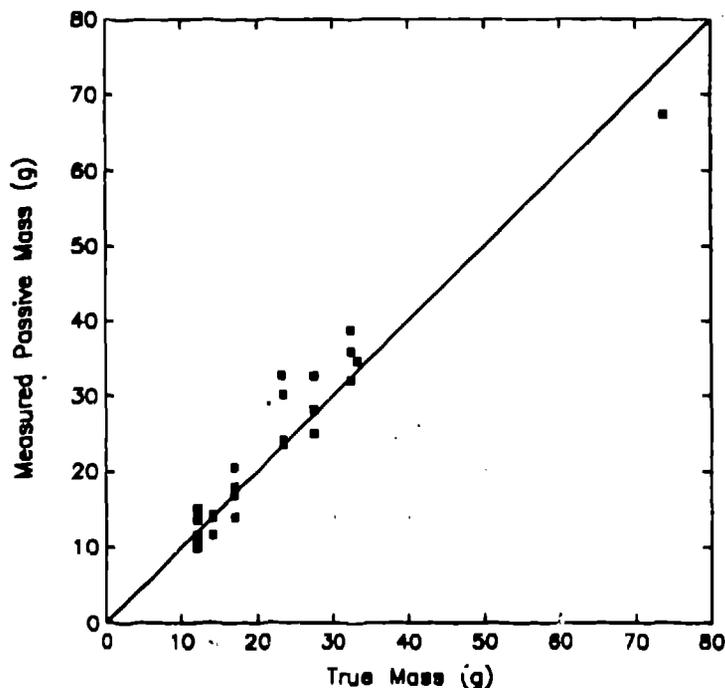


FIGURE 7. The measured passive fissile mass of the mockup fuel pellet samples vs the true fissile mass.

#### 4.3 Results with actual wastes

To date, only 10 cans of actual waste have been made available for assay. Table I summarizes the results from those measurements, expressed in equivalent grams of low burn-up plutonium. All that we know about the waste cans is that they contain grindings from irradiated fuel pellets (no intact pellets) and hot cell debris, and that the surface exposure rates are ~10 to 100 R/h. The fissile material present is presumed to be a mixture of uranium and plutonium of unknown isotopic composition. For purposes of accountability, a fissile mass of 0.5 g had been previously assigned to each of the cans. This was no more than a guess by the operator(s) who generated the waste and should not be taken as the true

mass. In addition to the "normal" active assay result, Table I shows the fissile mass computed using the special fuel pellet function (Eq. 6).

For each waste can, the passive assay showed a fissile mass of <10 g, which is below our sensitivity limit for 900-s passive assays. Because we know that grindings (and not intact fuel pellets) are involved, the normal active assay should be reliable. As shown in Table I, the normal DDT assay gives a fissile mass range for the 10 cans of 0.15 to 1.64 g, with an average value of 0.91 g. This demonstrates that the DDT assay system is not impaired at this exposure rate and that, for these 10 cans, the nominal value assigned by the operators was not greatly in error.

TABLE I. Measured fissile mass for actual waste cans.			
Can Number	Active Mass		Passive Mass (g)
	Normal (g)	Pellet (g)	
88	0.25 ± 0.04	0.38 ± 0.19	<10
84	1.32 ± 0.21	4.7 ± 2.3	<10
85	0.22 ± 0.04	0.33 ± 0.16	<10
80	1.32 ± 0.21	4.7 ± 2.3	<10
83	1.05 ± 0.17	3.3 ± 1.7	<10
78	0.16 ± 0.02	0.19 ± 0.09	<10
87	0.50 ± 0.08	1.1 ± 0.55	<10
86	1.62 ± 0.26	6.4 ± 3.2	<10
81	1.63 ± 0.26	6.5 ± 3.2	<10
82	1.05 ± 0.17	3.4 ± 1.7	<10

## 5. CONCLUSIONS

Our observation that making DDT measurements in two positions results in only a slight improvement in the assay accuracy implies that non-uniformity in the interrogating flux is not a primary source of error with this device. Position-related errors are largest when the matrix is highly moderating or absorbing and are probably due more to the amount of absorber or moderator immediately surrounding the source than to the absolute position of the source. Indications are that non-uniformity of the matrix is our chief source of assay error, and that the effects of source position and matrix distribution are tied together in such a way that merely knowing the position of the source is not sufficient to improve the assay significantly.

The alternate equation for DDT assays of fuel pellets seems to work moderately well up to 140 g, and it may be that this approach will prove useful for certifying wastes of unknown form as being below the 200-g limit set by WIPP. However, we feel that further testing of this approach, perhaps with real wastes, is required. We also note that when the error estimate is added to the assay result, as little as 12 g of finely divided fissile material may appear to exceed the 200-g limit with this approach. We intend to further evaluate this method by comparing it with the combined thermal-epithermal neutron [3] (CTEN) method.

Because of the difficult nature of the wastes to be assayed and because of the large assay errors that can be expected, we have tentatively recommended that the operators classify the wastes into three categories:

1. waste composed of finely divided fissile material;
2. waste containing fuel pellets; and
3. waste of unknown form.

With the first category, the combination of the (normal) DDT and passive assays should give the best estimate of the fissile mass in the cans. With waste known to contain fuel pellets, we recommend that the alternate fuel pellet function approach be used, but only to confirm the estimates of the mass made by the operators who generated the waste. For the third category, we can obtain upper and lower fissile limits using both approaches, but we're hopeful that measurements with the CTEN device will provide more definitive results.

#### ACKNOWLEDGEMENTS

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#### REFERENCES

1. KUNZ, W.E.; CALDWELL, J.T.; ATENCIO, J.D.; BERNARD, W.; FRANCE, S.W.; HERRERA, G.C.; HSU, H.H.; KUCKERTZ, T.K., and PRATT, J.C., "Current Status of the Multi-Isotopic Waste Assay System," Proc. Am. Nucl. Soc. Topical Meeting on Treatment and Handling of Radioactive Wastes, Richland, Washington, USA, April 19-22, 1982 (Battele Press, April 1982).
2. CALDWELL, J.T.; HASTINGS, R.D.; HERRERA, G.C.; KUNZ, W.E., and SHUNK, E.R., "The Los Alamos Second-Generation System for Passive and Active Neutron Assays of Drum-Size Containers," Los Alamos National Laboratory report LA-10774-MS (September 1986).
3. COOP, K.L.; CALDWELL, J.T., and GOULDING, C.A., "Assay of Fissile Materials Using a Combined Thermal/Epithermal Neutron Interrogation Technique," Proceedings of the Third International Conference on Facility Operations - Safeguards Interface, pp. 333-338, November 29 - December 4, 1987, San Diego, California, USA.
4. ESTEP, R.J., "GSHELL: A Data Acquisition Software Package for IBM-PC Compatible Computers," Los Alamos National Laboratory report, to be published.