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LA-UR--89-2124

DE89 014225

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TITLE NEUTRON DIEAWAY METHODS FOR CRITICALITY SAFETY MEASUREMENTS OF FISSILE WASTE

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SUBMITTED TO Nuclear Criticality Safety Division of the American Nuclear Society International Topical Meeting on Safety Margins in Criticality Safet San Francisco, California November 26-30, 1989

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ABSTRACT

The differential dieaway technique (DDT), which uses a pulsed neutron source to interrogate containers of fissile materials with thermal neutrons, is reviewed. This method is widely used for certifying transuranic nuclear wastes for eventual emplacement at the Waste Isolation Pilot Plant. For purposes of criticality safety, an upper limit of 200 g of fissile material is permitted in a 55-gal waste drum. Problems involving waste-matrix effects and self-shielding may severely limit the accuracy of the DDT measurement. A dieaway method that uses both thermal and epithermal neutron interrogation, which has the potential for reducing these problems, is being developed. Recent experimental and calculational results for this development are described.

I. INTRODUCTION

Before transuranic nuclear wastes can be stored at the Waste Isolation Pilot Plant (WIPP), the waste generator must certify that each container and its contents satisfy a number of specific requirements, which include a minimum transuranic radioactivity level and a maximum fissile material content. The transuranic radioactivity level (for long-lived alpha emitters) was originally set at 10 nCi/g of waste, averaged over the container contents, but this was later changed to its current value of 100 nCi/g. Waste with less transuranic radioactivity generally can be disposed of as low-level waste at greatly reduced cost. A maximum fissile content has been set at 200 g for the standard 55-gal (200-*l*) waste drums, while up to 600 g may be permitted for some larger waste containers. These limits were imposed to ensure that criticality excursions could not occur during handling or storage. Recently, the real possibility has been raised that even more restrictive fissile limits may be imposed as a requirement for transportation to the WIPP site. It is still uncertain when WIPP will open, and waste has not yet been shipped there, although a substantial quantity of waste has been certified in anticipation of WIPP opening soon.

The need to measure low levels of transuranic radioactivity in the tens of thousands of containers destined for emplacement at WIPP led to the development of the differential dieaway technique (DDT) assay device by W. Kunz and J. Caldwell about 10 years ago.[1] As detailed in the next section, this method uses a pulsed neutron source to induce fission and detects the prompt fission neutrons with ^{3}He proportional counters. Kunz and Caldwell reported detection limits as low as 1 nCi/g in 100 kg of waste containing low-burnup plutonium, which corresponds to ~1 mg of fissile material.

While this exceptionally good sensitivity is not needed to verify that waste containers meet the WIPP criticality limits, such DDT systems are routinely used for that purpose, since the same assay data can be used for both the transuranic and fissile determinations. In fact, the DDT method has become the standard for assaying

55-gal and larger containers destined for emplacement at WIPP. Built by the Los Alamos National Laboratory, one or more of these devices is in use at the major defense transuranic waste storage and generating sites at Hanford, Idaho Falls, Oak Ridge, Savannah River, Rocky Flats, and Los Alamos. In addition, a mobile system, containing two assay units, has been built for use at these and other facilities operated for the Department of Energy. Differential dieaway assay devices have also been built and tested in other countries[2,3] and are now being manufactured commercially in the U.S.A.[a] The most intensive use of any of the Los Alamos DDT systems has occurred at the Idaho National Engineering Laboratory, where over fourteen thousand 55-gal drums were assayed with the original neutron generator before it failed; near the end of its life, the neutron output had decreased to about one-third of its original rate because of ^{3}H depletion.[4]

All of these systems were built to assay the contact-handled (CH) transuranic wastes, which have low levels of gamma and neutron radiation, that make up the great majority of the transuranic waste inventory in the USA. Those wastes with gamma-ray exposure rates >200 mR/h at the surface of the container are categorized as remote-handled (RH); in addition to the radiation-safety problems they cause, the high gamma-ray fluxes may be falsely detected by the neutron proportional counters (because of pulse pileup), preventing an accurate assay. In an RH-DDT unit built at Los Alamos for assaying containers of RH waste,[5] this problem has been overcome by heavy gamma shielding of a greatly reduced detector volume. These changes increased the detection limit to ~50 mg of ^{235}U, which is sufficient to ensure compliance with the fissile limit, but is not low enough to screen wastes at the 100-nCi/g transuranic limit in the 1-gal inner waste-container volume. For the Los Alamos RH waste, this lack of sensitivity is not crucial, because most of the containers are thought to have much more than 100 nCi/g of transuranic radioactivity, and the total number of containers is small. However, for other RH waste streams, the capability to screen wastes at the 100-nCi/g level may be more important.

II. STANDARD DDT ASSAY DEVICE

A. Description of Device and Method

A typical Los Alamos-built assay device has a central cavity, where the waste container is placed, that is enclosed by an inner layer of graphite and outer layers of polyethylene and borated polyethylene. A door allows access to the cavity, and the entire device is covered inside and out by an aluminum frame. A small, 14-MeV Zetatron neutron generator is located in one corner of the cavity; neutrons are produced by the $^{3}H(^{2}H,n)^{4}He$ reaction. A variety of ^{3}He-filled, neutron proportional counters are located in the cavity and in the walls behind the graphite layer. Some of the latter detectors are enclosed in a small amount of polyethylene moderator, which is shielded by layers of cadmium and borated rubber; these packages will only detect neutrons with energies above the cadmium cutoff at ~0.4 eV. All of the other ^{3}He counters are most efficient for thermal neutron detection. Figure 1 shows a recently built 55-gal-drum assay device.

Fig. 1. Photograph of a Los Alamos-built, 55-gal-drum DDT assay device.

a Pajarito Scientific Corp., Los Alamos, NM 87544.

During the "active" mode of operation, the neutron generator is usually pulsed at a rate of 50 s^{-1}, producing a nominal output of 10^6 14-MeV neutrons per pulse. Within a few hundred microseconds after a pulse, the low-Z materials in the walls (and in the waste container) moderate the neutrons to thermal energies (<0.4 eV), and the shielded detector packages no longer see the interrogating neutrons. Subsequently, the thermalized interrogating flux decreases in intensity (dies away) with time at a rate that depends on the details of the device construction and the contents of the waste container; neutron moderators and absorbers in the waste matrix can have large effects on both the interrogating flux intensity and dieaway time. The dieaway flux pattern can usually be well approximated by an exponential (or sum of two exponentials) curve over wide time regions, with half-lives typically in the 1/2- to 1-ms range. Thus, within ~10 ms or less, the thermal interrogating flux has decreased to negligible levels. During the dieaway period, the interrogating flux induces fission in the fissile isotopes in the waste. The prompt fission neutrons can then be readily detected in the shielded detector packages, with a total efficiency of about 3%. Figure 2 demonstrates the typical patterns observed in a multichannel scaler.

Recent Los Alamos-built, drum-size DDT systems are also able to perform "passive" singles and coincidence neutron measurements, with a total singles counting efficiency of ~12% for fission neutrons[6] Since ^{240}Pu (a spontaneous fission isotope) is always present with ^{239}Pu, the passive coincidence measurement provides an additional method of determining the ^{239}Pu mass value if (1) the ratio of the two isotopes is known, (2) enough ^{240}Pu is present to provide reasonable coincidence count rates, and (3) other neutron emitters do not overwhelm the detection system.

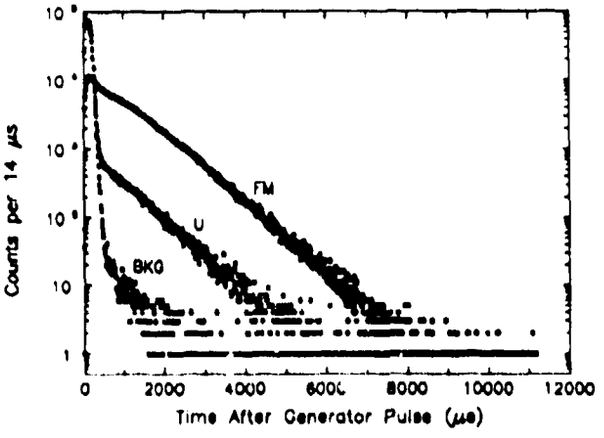

Fig. 2. Multichannel spectra obtained with a DDT device during 12000 neutron generator pulses. FM is a bare ^3He interrogating flux monitor. The other two spectra are for cadmium- and boron-shielded ^4He detectors: one with an empty cavity (BKG) and one with a 9-g uranium foil (93% ^{235}U) centered in the cavity.

B. Data Analysis and Errors

In the Los Alamos DDT systems, the fissile assay values are derived from the net count rates recorded in a selected time region relative to the interrogating pulse; for the data in Fig. 2, the region used is 700 to 4200 μs. Corrections for variations in the interrogating neutron flux intensity and waste-matrix moderation and absorption are made to the net counts. Such correction factors are made with data obtained from two flux monitors during the interrogation and, for waste drums containing the appropriate radioactive isotopes, with passive counting of spontaneously emitted neutrons before or after the active measurement. Calibration factors, which depend on the fissile isotope present, are then applied to the corrected active count data. The factors and procedures used for deriving them for the Los Alamos drum-size DDT devices are described in detail in Ref. 6.

The various factors and corrections can have a large effect on the derived fissile values. For example, when interrogated with thermal neutrons, ^{239}Pu emits 1.5 times as many neutrons as ^{235}U per unit mass, and the matrix correction factors can easily differ by as much as a factor of 5, or more, between hydrogenous and non-hydrogenous waste. However, since the isotopic composition of the fissile material is well known for most waste streams, it is the uncertainty associated with the matrix correction factors that generally contributes most to the assigned measurement error, which is 10% or more.

In making this type of analysis, a number of simplifying assumptions are made regarding the assay measurements:

(1) the waste matrix is uniform throughout the drum volume;

(2) fissile material is uniformly distributed in the waste; and,

(3) there are no "lumps" of fissile material large enough to cause self-shielding problems when interrogated by the thermal neutrons.

While these assumptions may be reasonably valid for some waste streams, they are clearly not for others. Unfortunately, there is generally no way to demonstrate that all of them are valid for particular waste drums. However, because most waste drums are also radiographed, it would be possible in some cases to know that the first assumption was not valid, but then still not know what correction to apply to the results to compensate for the non-uniformity. In "benign" waste matrices, non-uniformity in either the matrix or the fissile-material distribution might have little effect on the assay results, while for highly moderating or absorbing matrices, the effect could be quite large, easily a factor of 2 or more.

Possibly the largest potential assay errors, however, are associated with the third assumption: no significant self-shielding. If some of the fissile material is present as lumps or pieces, then self-shielding will occur, and unless corrected, the active assay result will tend to understate the fissile mass present. This happens because the mean-free-path length of thermal neutrons in fissile material is quite small: ~0.03 cm in either ^{235}U or ^{239}Pu metal. Thus, for lumps with dimensions of this magnitude or larger, only a surface layer of the material is thoroughly interrogated by the thermal flux, and consequently, the fission yield is more closely related to the surface area than the volume or mass. Just how large the self-shielding effect can be is illustrated for metal spheres of enriched uranium in Table I.

The self-shielding in low-burnup plutonium metal (93% ^{239}Pu) is even somewhat greater.[2,9] However, it should be emphasized that these calculations are for the worst-case scenario of highly enriched metal spheres. Lower enrichment or density, or any non-spherical shape will result in less self-shielding. Nonetheless, it seems possible that in some cases, perhaps rare, self-shielding could result in active assay values an order-of-magnitude under the actual fissile content.

For plutonium wastes, the presence of plutonium lumps would have no significant adverse effect on the observed passive coincidence counts; hence, if self-shielding were appreciable, the passive value should be considerably larger than the active assay results. This discrepancy would provide an indication that the problem existed, and the passive value would generally be more accurate. Experimental data on an actual plutonium waste stream have been evaluated to estimate the average amount of self-shielding present as a function of the calculated mass.[6] These data have been fitted to an exponential function, and subsequently, that function has been used in some Los Alamos-built DDT devices to correct for self-shielding for other plutonium waste streams. The magnitude of this correction is relatively small; for example, when the uncorrected active measurement result is 50 g, the reported mass obtained with this self-shielding factor is 68 g. (Additional data are being analyzed, and the self-shielding factor may be changed.[10]) Clearly this average self-shielding factor would not be adequate for wastes containing a large fraction of lumpy, high-density, fissile material.

For plutonium wastes containing large quantities of other spontaneous fission isotopes, or where singles neutron rates are quite large because of (α,n) reactions, the passive coincidence data may not be a reliable indicator of self-shielding. This may also be true for waste containing primarily uranium, or uranium and plutonium in unknown ratios.

TABLE I. Calculated Self-Shielding in Enriched (93% ^{235}U) Uranium Metal Spheres

Mass (g)	Fraction of Maximum Possible Fission Rate[a]
0.001	0.54
0.01	0.35
0.1	0.19
1.0	0.092
10.0	0.044
100.0	0.021
200.0	0.017

[a]Calculated using a 585-group Maxwell-Boltzman thermal-neutron distribution[7] and the ENDF/B-V cross sections.[8] The metal spheres (ρ = 18.7 g cm^{-3}) were placed in a void, with the source located on the surface of a sphere with a larger radius.

The need to assay the latter type of waste at Los Alamos recently resulted in the development of a technique that provides additional information about self-shielding and may mitigate the effect for some waste forms.[11] While providing thermal neutron interrogation as in the DDT method, the new technique also interrogates the container with epithermal neutrons, which are subject to much less self-shielding in fissile materials. This combination of interrogating fluxes has the potential for greatly reducing some of the assay uncertainties inherent in the standard DDT method.

III. COMBINED THERMAL/EPITHERMAL NEUTRON INTERROGATION

A. RH-CTEN Device

Built specifically to assay small containers (30 cm high by 21 cm in diameter) of RH waste, the RH-CTEN (remote-handled, combined thermal/epithermal neutron) device is constructed almost entirely of lead bricks (see Fig. 3). The lead is used to shield the neutron detectors from the high gamma fields associated with the RH waste, and it serves as the moderator of the interrogating neutrons. Lead moderates the interrogating flux more slowly than the low-Z materials used in the standard DDT system, thus providing an epithermal component for several hundred microseconds after the neutron generator pulse. After ~500 μs the flux is entirely thermalized.

Proportional counters filled with ^{4}He, which only detect neutrons with energies of several hundred kiloelectron volts or greater, are used to detect fission neutrons in the presence of both the epithermal and thermal interrogating fluxes. Thus, after these detectors recover from the initial burst of interrogating neutrons (40-50 μs), they detect the fissions induced by the moderated flux as it changes from predominantly epithermal at early times to entirely thermal at later times.

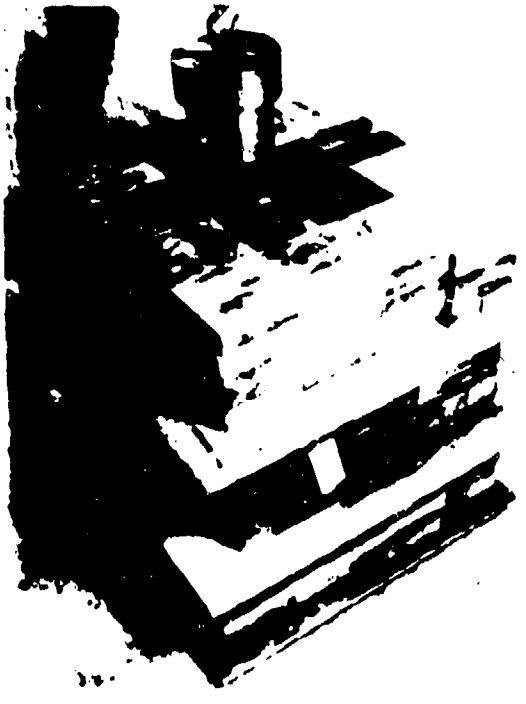

Fig. 3. Photograph of the RH-CTEN assay device.

Because of the increase in self-shielding as the average energy decreases with time, the observed time response patterns of uranium spheres are markedly different from those of thin foils. Therefore, the method provides the possibility of detecting self-shielding and correcting for it. (Reference 11 contains a detailed description of the method.)

Recently, a significant improvement in the sensitivity of the RH-CTEN device was achieved at Los Alamos by covering the ^{4}He proportional counters with borated rubber. This covering greatly reduces the background counts observed in the multichannel scalers after 50-100 μs. Apparently, most of those background counts were the result of thermal and epithermal neutrons interacting with the ~1 ppm of ^{3}He present in the helium fill gas. The large interrogating flux and the very large ^{3}He(n,p) cross sections resulted in a sizeable number of (n,p) events in the detectors, which were indistinguishable from the fast-neutron-induced ^{4}He recoils. The borated rubber reduced the observed background by more than an order of magnitude, because the ^{10}B(n,α) reaction prevented most low-energy neutrons from entering the detector. This background reduction decreased the lower limit of detection, defined as 4 std dev of the background, from ~100 mg to 20 mg of ^{235}U for a 3-min measurement. Figure 4 shows multichannel scaler spectra obtained with the boron-shielded ^{4}He detectors.

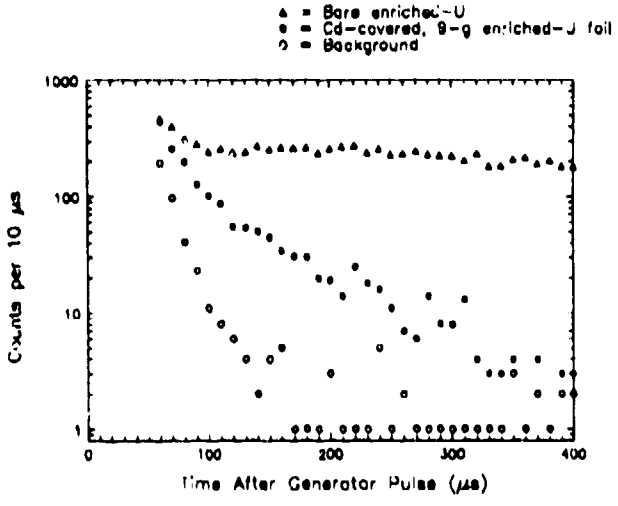

Fig. 4. Multichannel scaler spectra obtained with the RH-CTEN device for 9000 neutron generator pulses. The cadmium cover used was 0.8 mm thick.

B. Extension to Larger Containers

The feasibility of extending the CTEN method to 55-gal drum-size containers of CH waste has recently been studied at Los Alamos using Monte Carlo calculations.[12] It was concluded that this method might be valuable in resolving self-shielding problems in wastes with moderate- and high-Z matrices. Unfortunately, the presence of even modest amounts of hydrogen would attenuate the epithermal component too rapidly for it to be used directly to assay waste; however, the thermal component could still be used and is actually enhanced. It was suggested that even for such wastes, measuring the pattern of epithermal attenuation might be used indirectly to provide additional matrix correction factors to increase the accuracy of the assay for finely divided fissile material.

Assay devices constructed entirely of lead or graphite, or a combination of the two materials were studied. (The long transit time of slow neutrons across the large cavity accounts for a long-lived epithermal component even when graphite is the primary moderator.) Based on the calculations and experience with the RH-CTEN device, it was concluded that a drum-size graphite/Pb device with the capability of detecting ~100 mg of ^{235}U could be readily built. The study also raised the possibility that the current generation of DDT devices, which have graphite liners, might have significant epithermal fluxes. Such fluxes could be used to provide some degree of epithermal interrogation, if ^{4}He proportional counters were added to the devices to detect the fission neutrons. Both experiments and Monte Carlo calculations, reported in the next section, have been undertaken to examine this possibility.

IV. EPITHERMAL FLUXES IN DDT DEVICES

A. Experimental Results

The 55-gal-drum DDT device shown in Fig. 1 was used for the epithermal and thermal flux measurements. Six ^{4}He proportional counters were attached to the inside of one wall; each detector was 5 cm in diam by 25 cm long, with a fill pressure of 220 psia (15 atm). Measurements were made with and without a borated rubber shield (3 mm thick, 25% boron by weight) around the detectors. A multichannel scaler was used to record counts in the detectors as a function of time after the generator pulse. Figure 5 shows 2 multichannel scaler spectra, each collected using 36000 generator pulses. The lower curve was

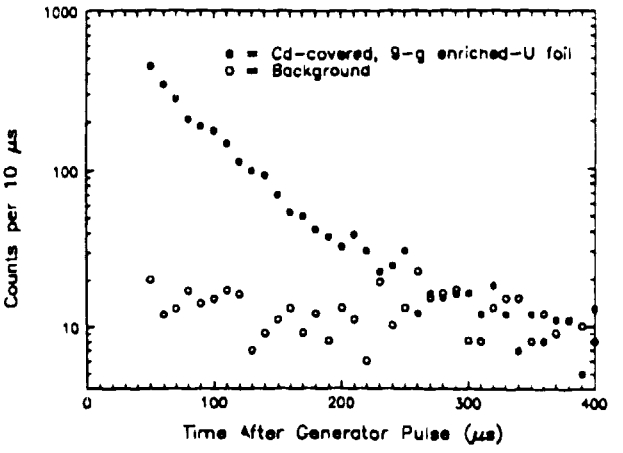

Fig. 5. Multichannel scaler spectra obtained with bare ^{4}He detectors for 36000 neutron generator pulses; the DDT device pictured in Fig. 1 was used for the measurements.

obtained with no fissile material in the chamber, while the upper curve is for a Cd-covered, enriched-uranium foil, centered on and in contact with the bare detector package. This geometry maximized the count rate and clearly demonstrates the existence and shape of the epithermal (i.e., epicadmium) flux.

Figure 6 shows data obtained in the same geometry, which demonstrate the difference in the spectra for bare 10.7-g spheres and a 9-g foil (both 93% ^{235}U); 18000 generator pulses were used for each spectrum. At very early times, the four spheres gave a higher response than the foil, while at later times, as the epithermal component of the interrogating flux became negligible, the converse

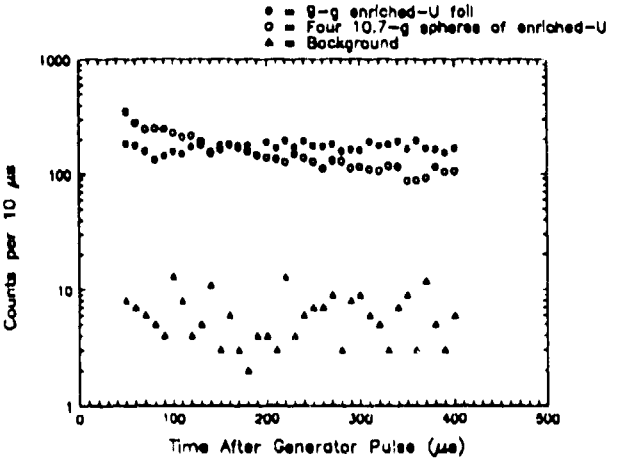

Fig. 6. Multichannel scaler spectra obtained with bare ^{4}He detectors in a DDT device using 18000 neutron generator pulses for each. The four 10.7-g spheres were well separated from each other.

was true. The relative response of the foil and spheres changed by a factor of 4 over the time domain shown, which is due to the difference in self-shielding between them and the changing epithermal-to-thermal flux ratio. Such differences in response to the combined thermal/epithermal neutron interrogation might be exploited to detect self-shielding in waste containers and improve assay accuracy where it is detected. Of course, different waste matrices also would affect the shape of the multichannel spectrum and would complicate observation and quantification of self-shielding. Unfortunately, the self-shielding effect might be completely obscured in some matrices, especially hydrogenous ones.[12] More extensive investigations are required to better understand the utility and limitations of the method.

To determine the fissile sensitivity that could be obtained using the CTEN method in existing DDT systems, the bare uranium foil was placed in the center of the cavity, ~25 cm from the ^{4}He detectors. When the detectors were not shielded with boron, the measured detection limits were 0.5 g for bare ^{235}U and 3 g for Cd-covered ^{235}U for 9000 generator pulses. When the detectors were covered with borated rubber, the measured limit of detection was 0.1 g of ^{235}U. No detection-limit measurements were made on Cd-covered ^{235}U when the detectors were shielded with boron, but based on calculation the value would be ~1 g.

All of these measurements were made without a "waste" drum in the cavity. A limited number of additional measurements were made with 55-gal drums containing various matrix materials. These measurements showed that the background counts increased above the empty-cavity background, with the amount depending on the matrix. Thus, detection limits for ^{235}U in actual waste drums could be somewhat higher than those indicated above.

B. Monte Carlo Calculations

The Los Alamos neutron transport code, MCNP,[8] was used to model a 55-gal-drum DDT device for calculating the expected thermal and epithermal fluxes. (The same procedure described in Ref. 12 was used, and more details can be found there.) An inner core of 10-cm-thick graphite enclosed a 100-cm-high by 66-cm by 66-cm cavity. Layers of polyethylene and borated polyethylene, with a total thickness of 25 cm, surrounded the graphite. The amount of boron was adjusted to obtain a thermal die half-life of about 500 μs in the 1- to 3-ms regi. match the experimental values. This procedure was used to simulate, in a gross fashion, the effect on the flux of the cadmium and boron shields used in various locations in the DDT device. The neutron generator was located in one corner of the cavity; it was assigned a composition of hydrogen, carbon, oxygen, and fluorine to simulate the moderating effects of the actual plastic housing and insulating fluid in it.

Some of the calculational results are shown in Fig. 7. The calculated epithermal (>0.4 eV) flux dieaway half-life was 36 μs, in good agreement with the experimental measurement of 40 μs. Both the calculated thermal and epithermal flux magnitudes were approximately a factor of 5 to 8 less than those previously calculated[12] for the graphite/Pb CTEN device. The calculated epithermal/thermal flux ratio was somewhat lower in the DDT system, showing that a device built of graphite and Pb should provide better CTEN assays than a current-generation DDT device with ^{4}He detectors added. However, the latter device might be very useful for some "problem" drums, and it only involves the addition of a small number of ^{4}He detectors and a modest amount of electronics to systems already built.

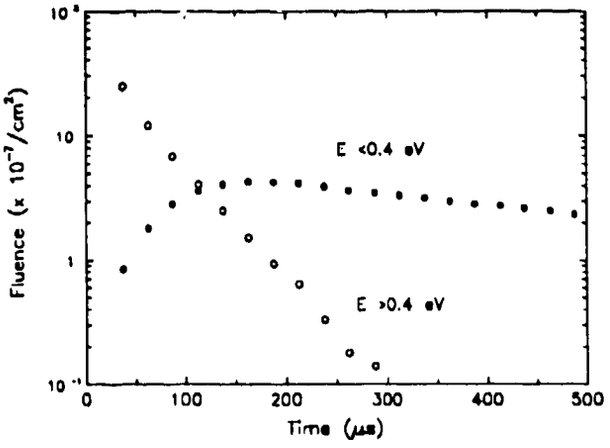

Fig. 7. Monte Carlo calculations for a 55-gal-drum DDT device. Fluence values per starting neutron are shown on the vertical axis; points are plotted horizontally with time at the center of 25-μs-wide time bins referenced to the neutron starting time.

V. CONCLUSIONS

It is apparent that using the passive-active DDT system can result in large fissile measurement errors for some waste forms. What fraction of waste containers might give rise to significantly large errors is unknown; however, because of the economic value of fissile material and the control

measures used in generating waste, it seems reasonable to assume that few, if any, will actually exceed the criticality limits.

Rather, the major problem is being able to certify, on the basis of the assay measurement, that a particular container does not exceed the limit when the measured active values are tens of grams or more. For low-burnup plutonium waste streams, the passive coincidence counts can often be used to ensure that the fissile value reported is not grossly in error. However, for some waste streams, the passive count data may not be as helpful. Additional techniques, such as the CTEN method, could be very useful in some of these situations.

Adding several ^{4}He detectors to existing DDT systems would permit some degree of epithermal interrogation. For suspect drums, longer-than-normal interrogations using the ^{4}He detectors might provide data of sufficient statistical precision to resolve questions about self-shielding, at least in some waste matrices. This would certainly be a less costly approach than building a dedicated graphite/Pb CTEN device. Regardless of which type of unit is used, a considerable experimental and calculational effort would be required to determine how to interpret the results for the various waste forms that might be encountered. Experimental evaluation of the RH-CTEN unit on waste mockups is currently in progress at Los Alamos, and experiments with real RH wastes are planned for late 1989.

Another approach to deducing possible self-shielding and non-uniform fissile distribution in waste, using the basic DDT method, has recently been reported.[13] The response of individual neutron detectors is recorded to provide positional information about the fissile distribution in a 55-gal drum. The possibility of using such data to obtain "images" of the drum content was also examined A separate analysis of the potential resolution possible with this method has been made using computer simulation techniques.[14] The latter study also provides a similar analysis of another imaging technique that uses bremsstrahlung beams from a small linear accelerator to interrogate the waste container.

ACKNOWLEDGMENTS

The various assay devices used in the recent experiments desribed above were built by Krag Allander, Clarence Herrera, and Joe Lujan. Ray Hastings assembled and maintained the neutron generators, and Robert Estep converted the data acquisition electronics used with the RH assay devices to IBM-based systems. Many other members of the Advanced Nuclear Technology Group also provided valuable assistance.

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