Measurement of Uranium and Plutonium in Solid Waste by Passive Photon or Neutron Counting and Isotopic Neutron Source Interrogation
This report was not edited by the Technical Information staff.

This work was supported by the US Department of Energy, Office of Transuranic Waste Management.
Measurement of Uranium and Plutonium in Solid Waste by Passive Photon or Neutron Counting and Isotopic Neutron Source Interrogation

T. W. Crane
CONTENTS

ABSTRACT .............................................................................................................. 1

I. INTRODUCTION .................................................................................................. 1
   A. Scope .............................................................................................................. 1
      1. Waste Category ....................................................................................... 1
      2. Measurement Principles ....................................................................... 2
   B. Passive Neutron Assay ............................................................................. 2
   C. Active Assay ............................................................................................... 3
   D. Passive Photon Assay ............................................................................... 4

II. MEASUREMENT TECHNIQUES ....................................................................... 5
   A. Neutron Detection ....................................................................................... 5
      1. Detection Principles .............................................................................. 5
      2. Detector Applications .......................................................................... 5
   B. Isotopic Neutron Sources ......................................................................... 8
      1. Alpha-n Sources .................................................................................... 8
      2. Photoneutron Sources .......................................................................... 8
      3. Californium Neutron Sources ............................................................... 9
   C. Useful Combinations for Active Neutron Assay ..................................... 10
   D. Photon Detection ....................................................................................... 10
      1. Scintillation Detectors .......................................................................... 11
      2. Semiconductor Detectors .................................................................... 12
      3. Proportional Gas Detectors .................................................................. 12

III. PASSIVE NEUTRON ASSAY TECHNIQUES .................................................. 13
   A. Thermal Neutron Detector Wells ............................................................... 13
      1. Measurement Principle ........................................................................ 13
      2. Coincidence Circuits ........................................................................... 14
      3. Lead Shielding ...................................................................................... 17
      4. Instrument Examples ........................................................................... 19
      5. Expected Performance ........................................................................ 20
   B. Fast Neutron Scintillation Detector Wells .............................................. 25
      1. Measurement Principle ........................................................................ 25
      2. Instrument Examples ........................................................................... 26
      3. Expected Performance ........................................................................ 27

IV. ACTIVE ASSAY TECHNIQUES .......................................................................... 27
   A. Alpha-n Interrogation and Thermal Neutron Detection .......................... 27
      1. Measurement Principle ........................................................................ 27
      2. Instrument Example ............................................................................. 28
      3. Expected Performance ....................................................................... 28
   B. Alpha-n Interrogation and Fast Neutron Detection .................................. 31
      1. Measurement Principle ........................................................................ 31
      2. Instrument Example ............................................................................. 31
      3. Expected Performance ....................................................................... 32
   C. Alpha-n Interrogation and Neutron Detection with Scintillators .......... 34
      1. Measurement Principle ........................................................................ 34
      2. Instrument Example ............................................................................. 35
      3. Expected Performance ........................................................................ 35
FIGURES

1. One-shot coincidence circuit ................................................. 14
2. Multiplicity coincidence circuit ........................................... 14
3. Variable dead-time coincidence circuit .................................. 15
4. One-shot delayed one-shot coincidence circuit .......................... 15
5. Shift register coincidence circuit ......................................... 15
6. Variance to mean coincidence circuit ..................................... 15
7. Fission product energy spectrum .......................................... 18
8. Gamma-ray attenuation for lead shield ..................................... 18
9. Transportable passive coincidence counter for 55-gal barrels ...... 22
10. Coincidence well counters at the LASL plutonium facility. Foreground left is the 2-liter sample size well counter; background is the 30-gal barrel counter with the 60-cm-thick water-filled shield ...................... 22
11. Schematic diagram of the Active Well Coincidence Counter (AWCC) .... 28
12. Neutron collar for LWR fuel assemblies ................................... 32
13. Jumbo Random Driver .......................................................... 35
14. Assay system constructed within a 55-gal barrel for measurement of irradiated pebble bed reactor fuel .................................. 36
15. Assay system employing $^{124}$Sb-Be neutron interrogation and detection of penetrating neutrons from induced fissions ...................... 37
16. Proposed $^{124}$Sb-Be assay system for safeguards measurement of 55-gal barrels of waste ................................................... 37
17. Photoneutron assay system for fuel pellets or rods inserted into the center opening. Two $^{124}$Sb sources are placed in the beryllium core. Thirty $^4$He fast neutron detectors are placed in the outer nickel cylinder. The assembly is surrounded by a boron-loaded polyethylene shield (not shown) ........................................ 40
18. Assay system for the measurement of irradiated Rover fuel. Top: Data acquisition and analyses electronics located outside the hot cell. Bottom: Fuel tube positioned in front of the entrance to the $^{226}$Ra-Be neutron interrogation region. The $^4$He-filled fast neutron detectors are located on the sides of the interrogation assembly. The hot cell environment protects the operator from the dose from the $^{226}$Ra-Be source and the irradiated fuel. ................................. 41
19. Uranium borehole logging sonde .................................................. 41
20. Proposed barrel-size $^{252}$Cf Shuffler assay unit. Two interrogation positions are indicated. .................................................. 44
21. Conceptual drawing for a $^{252}$Cf Shuffler assay unit. The californium source is scanned axially past the barrel during interrogation ....... 45
22. Savannah River Plant (SRP) $^{252}$Cf Shuffler for the assay of scrap and waste in the uranium fuel fabrication facility ....................... 47
23. Neutron interrogator designed for the Idaho Chemical Processing Plant 47
24. Prototype Shuffler ................................................................. 49
25. Barrel-size $^{252}$Cf Shuffler test bed schematic diagram .................. 49
26. Barrel-size $^{252}$Cf Shuffler test bed ........................................... 50
27. Schematic diagram showing a cross section of the cylindrical LWR fuel rod assay system .................................................. 55
28. Multienergy californium assay system ....................................... 58
29. Thermal neutron $^{252}$Cf fuel-rod assay system with modifications for pellet-to-pellet scan. Indicated dimensions are in inches .......... 58
30. SGS for barrel-size samples ....................................................... 62
31. Multiple NaI detector barrel counter ......................................... 62
32. Multi-Energy Gamma-ray Assay System (MEGAS) .......................... 64
33. Elephant gun detector schematic drawing ................................. 67
34. Elephant gun, electronic unit, and waste box ............................... 67
### TABLES

I. Neutron Detector Characteristics ........................................... 7  
II. Americium-Lithium Neutron Source Characteristics .......................... 8  
III. Photoneutron Sources .......................................................... 9  
IV. Californium Neutron Source Characteristics ................................. 10  
V. Neutron Interrogation and Detection Schemes .................................. 11  
VI. Plutonium Neutron Emission Rates .......................................... 21  
VII. Detectability Limits for a Passive Thermal Neutron Well Counter  
     Operated in the Singles Counting Mode ..................................... 24  
VIII. Detectability Limits for a Passive Thermal Neutron Counter  
      Operated in the Coincidence Counting Mode ............................... 25  
IX. Active Well Coincidence Counter (AWCC) Specifications .................... 29  
X. Savannah River Plant $^{252}$Cf Shuffler Performance Data ................... 47  
XI. Prototype $^{252}$Cf Shuffler Characteristics with  
    a 0.5 mg $^{252}$Cf Source .................................................. 49  
XII. U-235 Content of Twelve 30-gal Barrels Containing Furnace Parts  
     and Reduction Residues Measured by the SGS and the Barrel-size  
     Shuffler Test Bed ............................................................ 51  
XIII. Uranium, Plutonium, and Americium Gamma-ray Emission Rates .......... 61  
XIV. MEGAS Detectability Limits ................................................ 65  
XV. Detectability Limits Summary ............................................... 69
MEASUREMENT OF URANIUM AND PLUTONIUM IN SOLID WASTE
BY PASSIVE PHOTON OR NEUTRON COUNTING AND ISOTOPIC
NEUTRON SOURCE INTERROGATION

by

T. W. Crane

ABSTRACT

A summary of the status and applicability of nondestructive assay (NDA) techniques for the measurement of uranium and plutonium in 55-gal barrels of solid waste is reported. The NDA techniques reviewed include passive gamma-ray and x-ray counting with scintillator, solid state, and proportional gas photon detectors, passive neutron counting, and active neutron interrogation with neutron and gamma-ray counting. The active neutron interrogation methods are limited to those employing isotopic neutron sources. Three generic neutron sources (alpha-n, photoneutron, and $^{252}$Cf) are considered. The neutron detectors reviewed for both prompt and delayed fission neutron detection with the above sources include thermal ($^3$He, $^{10}$BF$_3$) and recoil ($^4$He, CH$_4$) proportional gas detectors and liquid and plastic scintillator detectors. The instrument found to be best suited for low-level measurements (<10 nCi/g) is the $^{252}$Cf Shuffler. The measurement technique consists of passive neutron counting followed by cyclic activation using a $^{252}$Cf source and delayed neutron counting with the source withdrawn. It is recommended that a waste assay station composed of a $^{252}$Cf Shuffler, a gamma-ray scanner, and a screening station be tested and evaluated at a nuclear waste site.

I. INTRODUCTION

A. Scope

1. Waste Category. The waste category considered in this report is solid waste with uranium or plutonium contamination. The packaging containers are assumed to be large, such as 55-gal (208 liter) barrels. The nondestructive assay (NDA) instruments discussed for this waste category will be used
primarily for sorting. The more sensitive instruments will sort plutonium contaminated waste at the 10-nCi/g fiducial (nonretrievable storage).\textsuperscript{1}

The solid waste may or may not contain significant levels of beta-gamma activity. Of the instruments described some will measure only cold waste. The remainder will handle containers with up to 1 R/h gamma-ray activity with no modification; and with lead shielding the acceptable gamma-ray activity can be orders of magnitude higher.

2. Measurement Principles. This report summarizes the status and applicability of several techniques for the detection and measurement of uranium and plutonium in solid wastes. The NDA techniques described are limited to those that detect neutrons or gamma rays emitted either naturally or induced by interrogation with an isotopic neutron source. Assay systems that detect naturally emitted x rays are also included.

The available neutron and gamma-ray detection and neutron interrogation techniques will be listed. The combinations of techniques appropriate for waste measurement will then be identified and the capabilities and limitations of each instrument will be considered. Emphasis will be placed on the type of material measured, the detectability limit, and the accuracy.

Comprehensive and detailed guides to the fundamental principles for NDA instrumentation have been prepared for "monitoring of plutonium contaminated solid waste streams" through the Commission of the European Communities, Joint Nuclear Research Centre, Ispra Establishment, Italy.\textsuperscript{2-6} The general scope of those reports is about the same as this report except a discussion of neutron generators is included in their chapter on active neutron systems.\textsuperscript{6} However, the emphases of the European series and this report are different but complementary. The emphasis in the European reports is on the basic physics of an instrument category, while this report emphasizes the expected performance of a technique for assay of 55-gal barrel-size containers.

B. Passive Neutron Assay

The instruments that exclusively measure naturally occurring radiation are referred to as passive assay techniques. The natural neutron radiation arises either from spontaneous fission or from alpha particle interactions. Spontaneous fission has been observed for thorium and heavier elements;\textsuperscript{7} however,
for waste measurement only the even isotopes of plutonium and curium make a
significant spontaneous fission neutron contribution. An average of two or
more neutrons are emitted in the spontaneous fission process, and instruments
that detect spontaneous fissions rely on the coincidence (time correlation) of
the neutrons.

The longer lived heavy radioactive elements predominately decay by emission
of an alpha particle. The energetic particle can interact with light elements
(oxygen, fluorine, etc.) to yield a single neutron. Depending on the neutron
emission rate, the electronics used to distinguish between the single alpha-
induced neutrons and the coincident spontaneous fission neutrons can become
quite sophisticated. Unless the chemical and isotopic composition of the waste
has been well characterized, passive measurements of the total neutron emission
rate can only place an upper bound on the quantity of uranium or plutonium in
an item.

C. Active Assay

The instruments that induce fissions in the material being assayed are
called active assay techniques. This report is limited to isotopic neutron
sources for interrogation of solid waste materials with detection of neutrons
and in a few cases gamma rays from the fission process. The techniques will
primarily measure the fissile isotopes ($^{233}$U, $^{235}$U, $^{239}$Pu, and $^{241}$Pu);
however, in some circumstances other fissionable isotopes are also detected.

The active assay techniques employ one of three characteristics of fission
neutrons to distinguish the induced neutrons from the interrogating neutrons.
Those characteristics are neutron multiplicity, high-energy neutrons, and
delayed neutrons. Techniques that rely on neutron multiplicity use an inter-
rogation source that emits single neutrons and detect coincident neutrons from
the fission process. Techniques that rely on neutron energy use a low-energy
neutron interrogation source and detect the energetic neutrons from fission.
Techniques that measure delayed neutrons use an intense interrogation source;
then the source is withdrawn and delayed neutrons are counted.

The interrogating neutron source must generally be strong enough so that
the signal from the induced fissions is detectable above the passive neutrons
and other backgrounds. Some of the active instruments also measure the
passive neutron signal so that a better characterization of the waste can be
made by combining the active and passive results. For similar neutron
detection schemes a passive system is less costly, and the health and safety
problems associated with the neutron source used in active systems are avoided.
However, for fissile isotope measurement an active system is generally the only
practical method for low-level waste assay.

D. Passive Photon Assay

The photon emissions from uranium, plutonium, and their daughters can be
separated into two categories, gamma rays and x rays. Gamma rays arise from
the nuclear decay process and are emitted immediately after either alpha or
beta decay when the daughter nucleus is left in an excited state. The energies
and intensities of the gamma-ray radiation are unique to the particular radio-
active isotope undergoing the decay process. The gamma rays have energies as
low as about 20 keV and as high as about 3 MeV. Passive gamma-ray counting
has the advantage that a particular isotope can be identified and, by analysis
of the data, the quantity of the isotope can also be determined. On the other
hand, some isotopes emit gamma rays for only a small fraction of the decays so
that detection of small quantities becomes difficult. Also, low-energy gamma
rays can be absorbed before they escape from the container, making detection
impossible.

X rays result from the rearrangement of the atomic electrons in an atom
following a decay process in the nucleus. In certain nuclear decays an
atomic electron usually in the K or L shell is either absorbed by the nucleus
(electron capture) or ejected from the atom (internal conversion). The vacancy
in the atomic structure is then filled by electrons from higher energy states.
The electron transitions to the more tightly bound states yield the character-
istic x-ray energies of the daughter nucleus. The alpha decay of heavy
elements results in x-ray production primarily by the internal conversion
process with a small fraction of the x rays resulting from internal photoelec-
tric absorption, photoelectric absorption of photons emitted by other nuclei,
and electron excitation by alpha and beta particles. The K x rays associated
with uranium or plutonium have energies in the range of 90-115 keV and the L x
rays have energies ranging from 10-20 keV. Because x rays have relatively low
penetrabilities, the NDA instruments that use x rays must be restricted to
small containers of low-density waste. An instrument that measures L x rays
has been employed to sort combustible waste at the 10-nCi/g fiducial.
II. MEASUREMENT TECHNIQUES

A. Neutron Detection

1. Detection Principles. Three different neutron detection methods are used in the NDA instruments discussed in this report. Each has its advantages for certain applications. The methods include thermal or low-energy neutron detection with gas proportional counters, fast neutron detection with gas proportional counters, and fast neutron detection with scintillation counters.

Gas-filled proportional counters detect thermal- or low-energy neutrons by sensing the electric charge that is caused by the neutron reacting with the detector gas. Neutrons emanating from the sample being assayed are thermalized in the vicinity of the detector tube by a highly moderating material such as polyethylene. The detector tubes are filled with a gas with a high neutron absorption cross section, either $^3$He or $^{10}$BF$_3$. (In the boron trifluoride case, the $^{10}$B atom reacts with the neutron.) The kinetic energy of the charged particles released in the exothermic nuclear reaction is absorbed by the detector gas partly through the creation of ion pairs. The multiplication and collection of the negative ions, electrons, produces the electrical charge sensed by the detector electronics. Proportional gas counters detect fast neutrons by having the detector filled with a gas that has a large scattering cross section for energetic neutrons. Either helium ($^4$He), hydrogen, or methane is used. Hydrogen or helium nuclei recoiling from the scattering interaction ionize the gas initiating the detection process. Plastic or liquid scintillation counters detect fast neutrons when recoiling hydrogen nuclei trigger the scintillation process. The scintillation light is detected by photomultiplier tubes (PM) and then the signal from the PMs is processed by the neutron pulse sorting logic.

2. Detector Applications. The three neutron detector types used in NDA instruments have their own advantages for certain measurement applications. The gas-filled proportional counters for thermal neutron detection are used in passive well counters and systems employing delayed neutron counting. The detection wells have efficiencies ranging from 10 to 65% with the most typical value being about 25%. The thermal neutron detectors have the advantage that each nuclear reaction deposits a fixed amount of energy into the detector.
Even with edge and end effects in the detectors it is possible to set a detection threshold that eliminates noise and gamma-ray pulses while accepting all the neutron pulses. With this mode of operation a stability of better than 0.1% is possible. The fill gas may be either $^3\text{He}$ or $^{10}\text{BF}_3$. However, $^3\text{He}$ is the preferred choice. The $^3\text{He}$ detectors are more efficient per detector and per unit cost, the $^3\text{He}$ detectors operate at a lower bias voltage reducing the risk of high-voltage breakdown, helium is not a health hazard in case of leakage while boron trifluoride is dangerous, $^3\text{He}$ detectors can be used in active assay systems with intense neutron sources while $^{10}\text{BF}_3$ tubes cannot because of the buildup of electronegative gases such as fluorine, and if the $^3\text{He}$ detectors are purchased without the argon buffer gas then the gamma-ray insensitivity for the two fill gases is about equal. The thermal gas-filled detectors can be operated in a gamma-ray radiation level of about 1 R/h with no loss in neutron detection efficiency and up to about 10 R/h with a slight loss in efficiency. Lead shielding can be used to reduce large radiation fields to acceptable levels for operating the detectors.

The gas-filled detectors that detect energetic neutrons by recoil interactions have been used in NDA instruments that use low-energy interrogation neutrons while detecting fast neutrons from the induced fissions. Photoneutron sources can supply low-energy interrogating neutrons. Because of the high gamma-ray fields accompanying photoneutron sources, the gas-filled fast neutron detectors are the only practical detector for this application. The efficiency of gas-filled fast neutron detection systems is low, about 1% of the efficiency of either a thermal neutron gas proportional counter system or a plastic or liquid scintillation counter system. The recoil reaction that triggers the charge collection in the detector can deposit any energy up to the maximum energy allowed by the kinematics. Thus, no convenient value is available for setting the threshold. In actual operation the number of fast neutron-induced pulses increases with lower pulse heights so that the count rate is sensitive to the threshold setting or gain drifts in the electronics. The fast neutron gas-filled detectors can be operated at about the same gamma-ray radiation levels as the thermal neutron gas-filled detectors. However, the fast neutron detectors always lose neutron detection efficiency as the gamma-ray levels become higher, because the threshold level must be increased.
The scintillation counters are used in systems that detect coincident neutrons from fission\textsuperscript{21}. The scintillation counters have about the same efficiency as the thermal neutron detectors for a similar system; however, the scintillation counters have a much faster response time.\textsuperscript{22} The resolving time for coincident neutrons is on the order of 100 ns for large scintillation counters. The systems employing the scintillation counters can count coincident neutrons when a large singles background is present. These systems can measure the coincident neutrons from plutonium fluoride in either an active or passive mode. The scintillation counters are sensitive to gain and threshold drifts like the fast neutron gas-filled detectors. Because the scintillation counters are efficient gamma-ray detectors,\textsuperscript{23} they are only suitable for assaying "cold" materials.

The characteristics of the three neutron detector types used in NDA instruments are summarized in Table I.

<table>
<thead>
<tr>
<th>Detector Type</th>
<th>Detector Assembly</th>
<th>Neutron Energy Range for the Assembly</th>
<th>Detection Efficiency for Assembly</th>
<th>Gamma-ray Sensitivity Limit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal Neutron Proportional Gas</td>
<td>$^5\text{He} + 5% \text{CO}_2$,\textsuperscript{a} or $^{10}\text{BF}_3$ (Moderator)</td>
<td>All</td>
<td>$\sim 25%$</td>
<td>$\geq 1 \text{ R/h}$</td>
</tr>
<tr>
<td>Fast Neutron Proportional Gas</td>
<td>$^4\text{He} + 5% \text{CO}_2$,\textsuperscript{a} CH$_4$ or H$_2$</td>
<td>$E_n \geq 400 \text{ keV}$\textsuperscript{c}</td>
<td>$\sim 0.5%$</td>
<td>$\geq 1 \text{ R/h}$</td>
</tr>
<tr>
<td>Scintillator</td>
<td>Plastic or Liquid\textsuperscript{b}</td>
<td>$E_n \geq 250 \text{ keV}$\textsuperscript{c}</td>
<td>$\sim 25%$</td>
<td>&lt;10 mR/h</td>
</tr>
</tbody>
</table>

\textsuperscript{a}Polyatomic gas required for proper operation
\textsuperscript{b}Manufactured by Nuclear Enterprises, Inc.
\textsuperscript{c}Noise and gamma-ray interference may result in a higher minimum neutron energy.
B. Isotopic Neutron Sources

1. Alpha-n Sources. Alpha-n neutron sources use the reaction between an energetic alpha particle and a light element for neutron production. The neutrons produced by the alpha-n interaction are emitted singly. NDA instruments can employ alpha-n sources to interrogate a sample and detect coincident neutrons as the signature for fissile material.

Although a neutron source could be fabricated from any alpha-emitting element, practical considerations favor certain isotopes. Factors important in selecting the alpha-emitting isotope include a reasonable half-life, freedom from spontaneous or induced fission neutrons, low gamma-ray activity, and availability. The selection of the light element affects the neutron production rate per alpha particle and the energy of the emitted neutron. The alpha-emitting isotope and light element must be chemically compatible so that the two elements can be intimately mixed on a near-atomic scale. The combination that has been most frequently employed for NDA instruments is $^{241}$Am-lithium. The properties of americium-lithium neutron sources are summarized in Table II.

2. Photoneutron Sources. Photoneutron sources use the reaction between energetic gamma rays and either beryllium ($^9$Be) or deuterium ($^2$H, usually in the form of heavy water) to produce neutrons. The gamma rays must be above the neutron binding energy, 1.665 MeV for beryllium and 2.225 MeV for deuterium, to yield a neutron. Because the remaining stable isotopes have neutron binding energies above 6 MeV, an energy larger than that available from gamma-ray sources, no other materials are possible. The sources that emit gamma rays with energies sufficiently high to produce photoneutrons tend to have short half-lives. A few sources have half-lives longer than a month making them possible candidates for NDA instruments provided changing a hot gamma-ray source can be incorporated into the operating procedures of the facility.

<table>
<thead>
<tr>
<th>Alpha-emitting Isotope</th>
<th>$^{241}$Am</th>
</tr>
</thead>
<tbody>
<tr>
<td>Alpha Energy</td>
<td>5.5 MeV</td>
</tr>
<tr>
<td>Half-life</td>
<td>434.1 yr</td>
</tr>
<tr>
<td>Activity</td>
<td>3.42 Ci/g</td>
</tr>
<tr>
<td>Chemical Form</td>
<td>$^{16}$O</td>
</tr>
<tr>
<td>Target Material</td>
<td>Lithium</td>
</tr>
<tr>
<td>Weight per cent in source</td>
<td>89%</td>
</tr>
<tr>
<td>Mean Neutron Energy</td>
<td>0.5 MeV</td>
</tr>
<tr>
<td>Maximum Neutron Energy</td>
<td>1.6 MeV</td>
</tr>
<tr>
<td>Neutron Production Rate</td>
<td>$6 \times 10^4$ n/s/Cl $^{241}$Am</td>
</tr>
</tbody>
</table>
TABLE III
PHOTONEUTRON SOURCES

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life (days)</th>
<th>Neutrons $^{a}$ $10^6$/s/Ci</th>
<th>Beryllium Target $^{a}$ Mass for $^{c}$ $10^8$ n/s (g)</th>
<th>Mean Energy $^{a}$ Mean Energy $^{b}$ (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{126}$Sb</td>
<td>60.2</td>
<td>3.2</td>
<td>0.0018</td>
<td>26.94% &amp; 360.6%</td>
</tr>
<tr>
<td>$^{88}$Y</td>
<td>107</td>
<td>3.4</td>
<td>0.0021</td>
<td>150</td>
</tr>
<tr>
<td>$^{144}$Ce</td>
<td>284</td>
<td>&lt;0.1</td>
<td>&gt;0.3</td>
<td>510</td>
</tr>
<tr>
<td>$^{228}$Th</td>
<td>700</td>
<td>1.6</td>
<td>0.076</td>
<td>830</td>
</tr>
<tr>
<td>$^{226}$Ra</td>
<td>1600 yr</td>
<td>1.0</td>
<td>100</td>
<td>&lt;670</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>0.08</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Deuterium Target $^{a}$</th>
<th>Neutrons $^{b}$ $10^6$/s/Ci</th>
<th>Mass for $^{c}$  $10^8$ n/s (g)</th>
<th>Mean Energy $^{b}$ (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>


$^{c}$Mass of gamma-ray emitting isotope

NDA instruments, which use low-energy neutrons from photoneutron sources to interrogate a sample, detect energetic or penetrating neutrons as a signature for fissile material. The energies of the photoneutrons must be low enough for the detector to separate the interrogating neutrons from the fission neutrons. Table III lists the characteristics of several possible photoneutron sources. Instruments have been built using $^{126}$Sb-Be and $^{226}$Ra-Be sources.

3. Californium Neutron Sources. Californium-252 provides the most intense neutron source for NDA applications. A summary of the $^{252}$Cf neutron source characteristics is given in Table IV.

NDA instruments employing delayed neutron detection are typically designed for sources containing about 1 mg of $^{252}$Cf. The californium is doubly encapsulated as a safety precaution with the outer capsule having approximately a 1-cm diameter and 2.5-cm length. The source is attached to a cable so that it can be used for sample interrogation and the source can be withdrawn to an
isolated storage container for delayed neutron counting. The small encapsulation size of the \(^{252}\)Cf source is important for rapid, reliable transfer of the source.

The personnel shielding for systems using a 1-mg \(^{252}\)Cf source is massive and in fact dominates the weight of the NDA instrument. A polyethylene shield approximately 120 cm across is required to reduce the neutron dose from the source to 0.5 mRem/h at the surface. With additional gamma-ray shielding the total mass can exceed 2500 kg.

C. Usable Combinations for Active Neutron Assay

Three generic types of isotopic neutron sources (alpha-n, gamma-n, and \(^{252}\)Cf) are available for sample interrogation and three neutron detection methods (thermal and fast gas-filled detectors and scintillation detectors) are available. If the combinations of schemes are further divided into prompt fission neutron or delayed neutron detection, then a total of 18 assay instruments are possible. The 18 combinations are displayed and commented on in Table V. Ten of the combinations can be eliminated because of noncompatibility between the interrogation and detection scheme as noted in Table V. Of the eight acceptable combinations, NDA instruments have been built using all but one of the possible schemes. Examples of these active assay instruments and their estimated detectability limits are discussed in Sec. IV.

D. Photon Detection

For passive photon counting the most important characteristics are efficiency and resolution of the detector.\(^8\) For both x-ray and gamma-ray counting the two detectors that have been most frequently employed are germanium semiconductor detectors and sodium (or cesium) iodide scintillation detectors. The sodium iodide crystals can be fabricated into a larger, more
absorptive detector thus making the system more efficient. Germanium detectors have better resolution than the scintillation detectors by a factor of 20 or more. Depending on the application either the higher efficiency or resolution can determine which detector type is better. For x-ray counting at energies below 30 keV, two additional detector types are usable, silicon semiconductor and proportional gas detectors. A brief description of each type of photon detector follows.

1. Scintillation Detectors. A scintillator detector converts the energy of a gamma ray or x ray to an electrical pulse proportional to the incident photon energy by the following multistep process. First, the incident photon is absorbed by the NaI(Tl) crystal. X rays usually deposit their entire energy in the crystal by photoelectric absorption. More energetic gamma rays experience a number of Compton collisions followed by photoelectric absorption. Second, the absorbed energy is converted to visible and ultraviolet light. The electrons produced by the Compton and photoelectric interactions excite the atoms in the NaI(Tl) crystal. The de-excitation processes result in the

a) Insufficient source strength
b) Insufficient detection efficiency
c) Gamma-ray interference
d) Source overrides signal
light production. Third, the light is absorbed by the photocathode of the PM. The inside walls of the crystal container are painted with a highly reflective coating to maximize the light reflected onto the photocathode. Fourth, electrons are emitted from the photocathode by the photoelectric effect. And fifth, the electrons emitted by the photocathode are collected and multiplied by the dynodes in the PM. The pulse from the output of the PM is then available for pulse sorting electronics such as multichannel analyzers or scalers.

2. Semiconductor Detectors. Solid-state semiconductor detectors are fabricated from either germanium or silicon. The conversion of the incident gamma ray or x ray to an electrical pulse requires fewer steps than for the scintillation counter.\(^8\) First, the incident photon energy is absorbed by Compton scattering and photoelectric absorption. Because the density and atomic number of silicon are both low, silicon detectors can only efficiently absorb x rays that have an energy less than about 30 keV. Second, the electrons produced by the Compton and photoelectric effect create electron-hole pairs in the semiconductor. Third, a bias voltage applied across the semiconductor prevents the electrons and holes from recombining, and the electrons and holes drift toward the opposite polarity terminals on the semiconductor. A preamplifier attached directly to the semiconductor amplifies the pulse and impedance matches it to the pulse sorting electronics. A multichannel analyzer is used with semiconductor detectors to record the energy spectrum of the incident gamma rays and x rays. Often a computer with sophisticated algorithms is employed to extract the desired information from the spectrum. Semiconductor detectors are more expensive than scintillation detectors for the initial capital investment. In addition germanium and silicon semiconductor detectors must be operated at liquid nitrogen temperatures, adding an upkeep cost to operating the detectors. (Hyperpure germanium and some silicon detectors may be stored but not operated at room temperature.) The high quality electronics required to exploit the resolution of the semiconductor detectors also increases the cost of semiconductor detector systems compared to scintillation detector systems.\(^{32}\)

3. Proportional Gas Detectors. Proportional gas photon detectors operate on principles similar to the neutron proportional gas detectors except a photon rather than a neutron initiates the detection process (see Sec. II.A.1).
fill gas is 2 atm xenon for detection of x rays having energies up to about 20 keV. For detection of L x rays from uranium and plutonium, gas proportional detectors represent a compromise between scintillation detectors and semiconductor detectors. The resolution of the gas-filled detectors is worse than that of semiconductor detectors and better than that of scintillation detectors. On the other hand, the active area is larger than that available for semiconductor detectors and smaller than that available for scintillation detectors. Gas counters are less expensive than either semiconductor or scintillation detectors and they require about the same level of support electronics as scintillation detectors.

III. PASSIVE NEUTRON ASSAY TECHNIQUES

A. Thermal Neutron Detector Wells

1. Measurement Principle. Thermal neutron detector wells measure the naturally occurring neutrons emitted by a radioactive isotope. The item being measured is surrounded by neutron detectors to improve the detection efficiency. Assemblies that completely surround the item (top, bottom, and sides) are referred to as $4\pi$ detector wells. The neutron detectors are gas-filled proportional counters with the fill gas being either $^3$He or $^{10}$BF$_3$ (see Sec. II.A.2).

After emission the neutrons are moderated to near thermal energies, before detection. The moderation process results in a statistical fluctuation of the time interval between the detection of the neutrons from the same fission event. The probability of detecting a second neutron after the first neutron is detected decreases exponentially as the time interval separating the two detection times increases. The characteristic time of the exponential decrease is called the die-away time. Thermal neutron detector wells have been built with detector die-away times ranging from 10 to 1000 $\mu$s. Factors that influence the die-away time include the size of the detector well, the neutron moderating and absorbing material in the item being assayed, the amount of moderating material surrounding the detectors, and the use of additional neutron absorbing materials in the well counter assembly.

The naturally occurring neutrons are emitted singly in the case of alpha-n reactions or with an average of two or more in the case of spontaneous fission. The die-away time of the well counters necessitates that a time interval about
as long as the die-away be used to allow for detection of "coincident" neutrons from spontaneous fission. Unfortunately the time interval for determining true coincidences is long enough to permit a high probability of accidental coincidences. These accidental coincidences include the detection of two alpha-n neutrons or one alpha-n and one spontaneous fission neutron. In order to separate the alpha-n (single) and spontaneous fission (coincident) neutron signatures, an electronic package is provided with the well counter. Advances in the electronic package have been made primarily in the direction of being able to process data in high count rate applications (≤100 g Pu metal, or ≤1 g PuF₃). For a given detector well the electronic package with the highest count rate processing capability will provide the largest assay dynamic range.

2. Coincidence Circuits. The electronic packages developed for use with thermal neutron detector wells contain coincidence circuits to separate the alpha-n (single) from the spontaneous fission (multiple) neutron signatures. The coincidences not arising from a spontaneous fission event are called accidental coincidences. All of the coincidence circuits are forced to accept the accidental along with the real coincidences because of the neutronic characteristics of the detector wells. The advantage of one circuit over another lies in the ease with which the real coincidence rate can be extracted without systematic errors caused by high count rates or changing backgrounds.

Figures 1-6 show coincidence circuits that have been used with

![Diagram of One-shot Coincidence Circuit](image1.png)

**Fig. 1.** One-shot coincidence circuit (EG&G Neg. #10261).

![Diagram of Multiplicity Coincidence Circuit](image2.png)

**Fig. 2.** Multiplicity coincidence circuit (EG&G Neg. #10254).
Fig. 3. Variable dead-time coincidence circuit (LASL Neg. #77-18271).

Fig. 4. One-shot delayed one-shot coincidence circuit (LASL Neg. #77-18267).

Fig. 5. Shift register coincidence circuit (LASL Neg. #77-18272)

Fig. 6. Variance to mean coincidence circuit (EG&G Neg. #10262).
thermal neutron detector wells. The circuit shown in Fig. 1 is called a one shot (OS). The accidental rate is estimated from the total count rate and no provision is made for handling a variable accidental rate such as might arise when radioactive samples are moved in the vicinity of the detector well. Figure 2 is a variation on the OS circuit that permits measurement of coincidences of different orders.\textsuperscript{35,36} Another variation of the OS circuit, shown in Fig. 3, is called the variable deadtime counter (VDC).\textsuperscript{37} The data obtained with the VDC is analyzed by an approach based on interval distributions. In calculating the quantity of material from the data, it is necessary to solve coupled equations by an iterative procedure.\textsuperscript{38}

Figure 4 shows a circuit that has a decoupling long delay to measure accidental coincidences during the assay. This circuit is referred to as the one-shot delayed-one-shot (OSDOS). The two one shots are of equal duration and the long delay is about 1 ms. A shift register is used for the long delay so that no counts are lost and there is an equal number of gates from the two one shots during the assay.\textsuperscript{38} The circuit shown in Fig. 5 is conceptually similar to the OSDOS shown in Fig. 4 except that a shift register replaces the two one shots. The shift register is coupled to an up/down counter that tallies the number of neutrons counted during the gate time (8-128 μs).\textsuperscript{39} When a neutron enters the circuit the number of neutrons in the shift register is added to the "reals + accidental" scaler and the total neutron count is incremented. After the neutron has been delayed 1 ms in a second shift register the current contents of the up/down counter at the later time is added to the "accidentals" scaler. The real coincidence rate is obtained by subtracting the "accidentals" from the "reals + accidentals" scaler. The shift register coincidence circuit shown in Fig. 5 is commercially available in a package that also contains a high-voltage supply for the neutron detectors, amplifiers and discriminators for the neutron pulses, a computer interface port, and a HP-97 calculator interfaced to the unit.\textsuperscript{40,41} Routine data reduction and a paper tape printout of the results is accomplished with the HP-97 calculator.

An approach different from that used in the first five circuits is employed in the circuit shown in Fig. 6. The measurement technique is based on variance measurements of the number of neutron counts in relatively short time intervals (0.5 to 4.0 ms). During an assay, typically about $10^6$ time intervals are observed. The variance of the singly emitted alpha-n neutrons is the square root of the number of neutrons counted; however, for the multiply
emitted spontaneous fission neutrons the variance is the square root of the number of fissions that result in detected neutrons and not the number of detected neutrons. The circuit shown in Fig. 6 compares the mean number of counts in each time interval to the variance about the mean to calculate the coincidence rate.42

Several intercomparisons between subgroups of the coincidence circuits shown in Figs. 1-6 have been made.43-47 The results of these comparisons show that only the OSDOS and shift register circuits are free from systematic bias when the background rate is changed during the assay. The shift register has the smallest deadtime correction to the coincidence count rate as a function of the total count rate. The shift register circuit is commercially available as a complete electronic package. Thus, for general thermal neutron detector well applications the shift register circuit is the preferred choice.

In certain limited applications the features available in other circuits can be valuable. For counting in a lead shielded detector well, it would be desirable to separate the high multiplicity background neutron coincidences caused by cosmic-ray interactions in the lead from the spontaneous fission events. The circuit shown in Fig. 2 is being investigated for this application.35,36 The reduced variance circuit shown in Fig. 6 could be used to detect the fissions induced by a pulsed accelerator based active assay technique. In this application, which is outside the scope of this report, one counting interval would be used for each accelerator pulse.

3. Lead Shielding. The gas-filled neutron detectors used for either thermal or fast neutron detection can be operated in gamma-ray radiation levels up to about 1 R/h. To use the tubes to measure items with gamma-ray radiation levels appreciably above 1 R/h it is necessary to provide gamma-ray shielding for the tubes. Of the possible gamma-ray shield materials lead is the most practical choice in terms of effective gamma-ray absorption, low interaction cross section with neutrons, availability, and ease of fabrication.

The gamma-ray attenuation properties of a lead shield can be calculated from the gamma-ray energy spectrum and the lead absorption coefficients with dose build-up factors.48 The approximate gamma-ray energy spectrum for mixed fission products shown in Fig. 7 was used to calculate the gamma-ray attenuation for a lead shield surrounding a 55-gal barrel.49 Figure 8 shows the gamma-ray dose attenuation as a function of the lead thickness.50
details of the attenuation curve are sensitive to the fraction of low- and high-energy gamma rays in the initial gamma-ray energy spectrum. The high-energy gamma rays have a dose attenuation of a factor of 10 per 5 cm. This attenuation factor can be used to conservatively estimate the required lead shielding thickness for waste materials containing fission products.

In addition to the lead dose attenuation, a geometric dose reduction results from moving detectors farther from the sample to accommodate the shield as noted in Fig. 8. The dose reduction is measured relative to the surface of the container. In systems where only a portion of the available surface is used for the neutron detector banks, moving the detectors farther from the shield decreases the neutron detection efficiency because the solid angle subtended by the detectors is reduced and because the lead can scatter neutrons.
away from the detectors. On the other hand, systems with $4\pi$ detector geometry can retain the same efficiency with the lead shield; however, more or larger detector tubes are required to enclose the larger volume.

The gamma-ray shielding is quite massive for large containers. For example a $4\pi$ lead shield for 55-gal barrels designed to reduce a 100 R/h radiation level to 1 R/h has a mass of about 4000 kg. With that quantity of lead, cosmic-ray interactions can result in an appreciable neutron production rate.\textsuperscript{51-53} In a passive counter the 4000 kg of lead can yield a background coincidence rate equivalent to about 9 g of $^{240}\text{Pu}$ at the Los Alamos altitude of 2200 m.\textsuperscript{54} At sea level the cosmic rays are attenuated by a factor of 4, and an overhead cover of concrete 1 m thick reduces the background another factor of 20. Thus, for a typical facility at sea level a background coincidence rate equivalent to 0.1 g $^{240}\text{Pu}$ can be expected from the lead shielding in a passive coincidence detection system.\textsuperscript{55} In a few cases it may be possible to operate a passive system deep underground such as in a tunnel; in this situation the background rate from cosmic rays becomes negligible.\textsuperscript{56}

4. Instrument Examples. The use of thermal neutron detector wells is common practice at facilities that handle plutonium. The smallest detector wells are portable and are used primarily by the International Atomic Energy Agency (IAEA) inspectorate. Portability is achieved by making the well as light as possible and designing the instrument for verifying cans of small volume (several liters) and high plutonium content. Weight reduction in the instrument has been accomplished by using water for the moderator surrounding the detector tubes (the water is added to the detector at the inspection site)\textsuperscript{57} or by trimming the polyethylene moderating material to a bare minimum.\textsuperscript{40} Because these wells are used with essentially no exterior shielding, the electronics should yield coincidence count results that are independent of background rate variations such as the OSDOS circuit (Fig. 4) or the shift register circuit (Fig. 5).

The well counters used by the in-plant personnel for accountancy measurements or process control can be designed with more exterior shielding and a larger sample capacity than the units employed by the IAEA.\textsuperscript{58,59} The instruments can be wheelable or custom built into assay stations along production lines.\textsuperscript{60} These in-plant instruments like the inspectors' well counters generally measure plutonium having a well-characterized chemical composition.
The electronic circuit should be able to measure materials with high count rates especially if PuF$_4$ is to be assayed. Of the electronic circuits discussed, the shift register (Fig. 5) requires the smallest rate corrections.

A compromise between the portable instruments used by the IAEA and the in-plant instruments is a well counter that can be separated into sections, which can then be assembled around the item to be assayed. Because the assay geometry varies according to the item being assayed, an elaborate calibration scheme is necessary for interpreting the results. This type of instrument is useful in a collaboration between a facility and outside experts for a campaign of assaying a number of similar items on a one-time basis.

Large well counters are typically used at facilities for measuring waste materials with small quantities of plutonium usually in a low-purity or unknown chemical composition. At a production facility the large well counters can be used to sort waste in terms of recoverable or nonrecoverable quantities of plutonium. If the item contains a nonrecoverable quantity of plutonium, then it would be sent to a waste storage area and perhaps analyzed by a more sensitive technique to determine the type of storage required. If the item contains recoverable quantities of plutonium and the recovery was to be accomplished at a separate facility, then the recovery facility may also have a large item thermal neutron detection well to verify incoming shipments. The waste containers are usually 30-gal (114 liter) or 55-gal (208 liter) barrels to satisfy shipping container requirements. Because the large well counters measure small quantities of plutonium and the detectors are usually heavily shielded, the type of electronic circuit employed does not significantly affect the results of the assay.

The largest thermal neutron detector wells are proposed for vehicle monitors at nuclear facilities. A prototype unit has been built to test the feasibility of monitoring small vehicles such as automobiles and light trucks. The well measures 3.05 m wide, 2.44 m high, and 6.10 m long for a total volume of 45 307 liters. The proposed detection scheme is to count total neutrons (above background) as a signature for nuclear material in the vehicle.

5. Expected Performance. This report compares various instruments in terms of their detectability limit. The detectability limit is defined as that quantity of material that yields a response three standard deviations above
background during a 1000-s assay. A discussion of the mathematical formulas used to compute detectability limits is found in the Appendix.

The thermal neutron detector wells measure the neutrons emitted by naturally occurring reactions. The neutron emission rate for plutonium isotopes in various chemical forms is listed in Table VI. The plutonium will be considered to be in one of two categories, weapons grade or reactor grade. A typical isotopic composition and neutron emission rates for the two categories are noted in Table VI. In addition to the neutron emission rates listed in Table VI, multiplication or self-interrogation can increase the observed neutron emission rates. Coincidence count rates are very sensitive to multiplication and sophisticated algorithms have been derived to extract the spontaneous fission rate. However, at the detection limit, multiplication effects are negligible because of the small quantities of plutonium present.

The neutron emission rates listed in Table VI constitute the available signal for the detection of plutonium by passive thermal neutron detector wells. Parameters in the operation of a well that affect the usable signal

<table>
<thead>
<tr>
<th>Table VI</th>
</tr>
</thead>
<tbody>
<tr>
<td>PLUTONIUM NEUTRON EMISSION RATES</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Isotope or Mixture</th>
<th>Spontaneous Fission Rate (/s/g)</th>
<th>Spontaneous Fission Multiplicity (v)</th>
<th>Alpha-n Neutrons Rate (/s/g)</th>
<th>Total Neutron Rate Rate (/s/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>238Pu</td>
<td>1100</td>
<td>2.26</td>
<td>1.4 x 10^6 2.1 x 10^6</td>
<td>2500 1.6 x 10^6 2.1 x 10^6</td>
</tr>
<tr>
<td>239Pu</td>
<td>0.010</td>
<td>2.2</td>
<td>45 4300</td>
<td>0.022 45 4300</td>
</tr>
<tr>
<td>240Pu</td>
<td>471</td>
<td>2.17</td>
<td>170 1.6 x 10^4</td>
<td>1020 1190 1.7 x 10^4</td>
</tr>
<tr>
<td>241Pu</td>
<td>0.011</td>
<td>2.2</td>
<td>~4 700</td>
<td>0.024 ~4 700</td>
</tr>
<tr>
<td>242Pu</td>
<td>800</td>
<td>2.16</td>
<td>~6 1000</td>
<td>1730 1740 2700</td>
</tr>
<tr>
<td>Weapons Grade</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Plutonium</td>
<td>30.1</td>
<td>2.17</td>
<td>56 5400</td>
<td>65 121 5500</td>
</tr>
<tr>
<td>Reactor Grade</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Plutonium</td>
<td>78.1</td>
<td>2.17</td>
<td>83 9100</td>
<td>169 252 9300</td>
</tr>
</tbody>
</table>

a)238Pu-0.02%, 239Pu-93.37%, 240Pu-6.30%, 241Pu-0.28%, and 242Pu-0.03%.
b)238Pu-0.15%, 239Pu-81.36%, 240Pu-15.11%, 241Pu-2.72%, and 242Pu-0.66%.
Include the neutron detection efficiency of the well ($\varepsilon$), the die-away time of the well ($\tau_d$), the gate time for coincidence counting ($\tau_g$), the deadtime for the detectors ($\tau_D$), the deadtime for the electronics ($\tau_e$), and the neutron background count rate ($b$). The background count rate can consist of single counts ($b_s$) and coincidence counts not caused by accidental coincidences ($b_c$).

Figure 9 shows a well counter that was part of the instrumentation used with the Mobile Nondestructive Assay Laboratory (MONAL). As shown in Fig. 9 the well counter was used for assaying 55-gal barrels. The well counter employed BF$_3$-filled detectors to obtain an efficiency of 14% and the die-away time of the well was measured at about 100 $\mu$s. The well counter used at the LASL plutonium facility (TA-55) is shown in Fig. 10. The assay chamber designed for 30-gal barrels is surrounded with 78 BF$_3$-filled detectors, yielding a neutron detection efficiency of 18% and a die-away time
The detectability limit by coincidence counting is about one gram of weapons grade plutonium for the barrel-size units shown in Figs. 9 and 10. Improvements in the detectability limit are achieved by using more efficient $^{3}$He-filled detectors and by increasing the exterior neutron shielding to reduce the background count rate.

If the thermal neutron detector well uses the total neutron count as the plutonium signature and the deadtimes associated with the detectors and electronics have a negligible effect on the total count rate, the signal count rate ($r_s$) is plutonium neutron emission rate (see Table VI) times the efficiency. The data collection scheme consists of a 1000-s count; however, background data with the assay chamber empty also need to be collected. The frequency of background measurements would depend on the degree of shielding provided by the instrument. For a heavily shielded chamber, one background measurement per day might be adequate. On the other hand, if the shielding is insufficient, resulting in background fluctuations attributed to the movement of nuclear materials in the vicinity of the instrument, then a background check would be required between each assay resulting in a decreased throughput. Assuming a 25% efficient well, the detectability limit by total neutron counts for various forms of plutonium is given in Table VII.

The electronic circuits permit coincidence data to be accumulated along with the total events used for the singles count rate analysis. Thus, using a coincidence circuit to collect data from a thermal neutron detector well does not interfere with the detectability limit obtainable by counting the total number of neutrons. Coincidence circuits permit accidental coincidences to be measured concurrently with the real coincidences. However, background coincidence rates associated with lead shielding must be measured separately.

The number of coincidence counts per spontaneous fission for the shift register (Fig. 5) is given by

$$F = \frac{\varepsilon}{2}[1 - \exp(-\tau_g/\tau_d)]\exp(-\tau_e/\tau_d)[v^2 + v(v - 1)]$$

where,

$$v = \text{average fission multiplicity (v = 2.17)},$$

of 80 $\mu$s.$^{63}$
### TABLE VII
DETECTABILITY LIMITS FOR A PASSIVE THERMAL NEUTRON WELL COUNTER
OPERATED IN THE SINGLES COUNTING MODE

<table>
<thead>
<tr>
<th>Isotopic Composition</th>
<th>Gamma-Ray Shielding</th>
<th>Detectability Limit^a (mg)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Metal</td>
</tr>
<tr>
<td>Weapons Grade^b</td>
<td>no^d</td>
<td>27</td>
</tr>
<tr>
<td></td>
<td>yes^e</td>
<td>53</td>
</tr>
<tr>
<td>Reactor Grade^c</td>
<td>no^d</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>yes^e</td>
<td>20</td>
</tr>
</tbody>
</table>

^a) Assay consists of a 500-s count with the item and a 500-s background count with the assay chamber empty. The quantity of material listed yields a signal three standard deviations above background.

^b) $^{238}$Pu-0.02%, $^{239}$Pu-93.07%, $^{240}$Pu-6.30%, $^{241}$Pu-0.28%, and $^{242}$Pu-0.03%.

^c) $^{238}$Pu-0.15%, $^{239}$Pu-81.36%, $^{240}$Pu-15.11%, $^{241}$Pu-2.72%, and $^{242}$Pu-0.66%.

^d) Nominal background rate of 5 counts/s.

^e) Nominal background rate of 20 counts/s.

\[ \sigma = \text{standard deviation about } \nu (\sigma = 1.08), \]

\[ \tau_e = \text{predelay in coincidence circuit (} \tau_e = 4 \, \mu\text{s).} \]

Equation (A-12) is used to determine the detectability limit for plutonium. The coincidence count rate ($r_s$ in Eq. (A-12)) is obtained by multiplying the spontaneous fission rate in Table VI by the number of countable coincidences ($F$ defined by Eq. (1)). The accidental coincidence count rate is explicitly included in Eq. (A-12) and is computed from the plutonium total count rate ($r_t$) and the background count rate ($b_s$). The plutonium total count rate ($r_t$) is obtained by multiplying the total plutonium neutron emission rate given in Table VI by the detection efficiency. An efficiency ($\varepsilon$) of 25%, a die-away time ($\tau_d$) of 100 $\mu$s and a gate time ($\tau_g$) of 128 $\mu$s were choosen as typical values for a 55-gal barrel size well counter. The coincidence detectability limit for various forms of plutonium is given in Table VIII.
### TABLE VIII
DETECTABILITY LIMITS FOR A PASSIVE THERMAL NEUTRON COUNTER OPERATED IN THE COINCIDENCE COUNTING MODE

<table>
<thead>
<tr>
<th>Isotopic Composition</th>
<th>Gamma-ray Shielding</th>
<th>Detectability Limit&lt;sup&gt;b&lt;/sup&gt;(mg)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Metal</td>
</tr>
<tr>
<td>Weapons Grade&lt;sup&gt;c&lt;/sup&gt;</td>
<td>no&lt;sup&gt;e&lt;/sup&gt;</td>
<td>5.3</td>
</tr>
<tr>
<td></td>
<td>yes&lt;sup&gt;f&lt;/sup&gt;</td>
<td>69</td>
</tr>
<tr>
<td>Reactor Grade&lt;sup&gt;d&lt;/sup&gt;</td>
<td>no&lt;sup&gt;e&lt;/sup&gt;</td>
<td>2.0</td>
</tr>
<tr>
<td></td>
<td>yes&lt;sup&gt;f&lt;/sup&gt;</td>
<td>26</td>
</tr>
</tbody>
</table>

<sup>a</sup>) Without shielding operation to 1 R/h, with shielding to 1000 R/h.

<sup>b</sup>) Three standard deviations above background for a 1000-s assay.

<sup>c</sup>) 238Pu-0.02%, 239Pu-93.07%, 240Pu-6.30%, 241Pu-0.28%, and 242Pu-0.03%.

<sup>d</sup>) 238Pu-0.15%, 239Pu-81.36%, 240Pu-15.11%, 241Pu-2.72%, and 242Pu-0.66%.

<sup>e</sup>) Nominal background rate of 5 counts/s.

<sup>f</sup>) Nominal background rate of 20 counts/s with a twofold coincidence background rate of 2.34 counts/s.

---

**B. Fast Neutron Scintillation Detector Wells**

1. **Measurement Principle.** Fast neutron scintillation detector wells measure the naturally occurring energetic neutron and gamma-ray radiation emitted in coincidence from a radioactive item. The detectors are plastic or liquid scintillators with PMs used to convert the scintillation light into electrical signals (see Sec. II.A.1). The scintillation detector's sensitivity to fast neutrons and gamma rays is an asset as well as a liability. In the fission process about six or seven high-energy gamma rays are emitted in coincidence with the two energetic neutrons. Thus, the scintillation detector wells have the potential for measuring much higher order coincidence events than do the thermal neutron wells. Moreover, the scintillation wells, which have a single particle detection efficiency comparable to the thermal neutron...
wells, can measure twofold coincidences with a much higher efficiency because the probability of detecting at least two radiation particles out of eight or nine is much higher than detecting two out of two or three.

The time interval for forming coincidences with a scintillation well is several tens of nanoseconds, making the resolving time a thousand times faster than for thermal neutron detector wells. The fast resolving time eliminates the requirement for the sophisticated coincidence circuits and the scintillation detector wells operate with coincidence logic similar to the OS circuit shown in Fig. 1 but without corrections for deadtime or accidental coincidences.

Because gamma rays are emitted during most radioactive decay processes, the gamma rays detected by a scintillation counter are by no means a unique signature for either uranium or plutonium. The scintillation counters use coincidence counting to make the response more sensitive to the fission process; however, radioactive decays often occur by gamma-ray cascades where several gamma rays are emitted in coincidence without being a part of a fission process.\footnote{72}

The sensitivity of scintillation detectors to gamma rays restricts their use to samples with low levels of beta-gamma activity ($\leq 1$ mR/h). Lead shielding can be used to reduce the radiation to tolerable limits with the penalty of producing a coincidence background (see Sec. III.A.3). Lead shielding can also be used for eliminating coincidences from low-energy cascade gamma rays. However, if the cascade gamma rays are above 1 MeV, as is the case for neutron captures in cadmium, the shielding approach to eliminating them is much less effective. The shielding also loses its effectiveness because pair production results in an increase in the gamma-ray absorption above 2 MeV so that increasing the shielding reduces the gamma-ray detection efficiency with little improvement in the ratio of spontaneous fission gamma rays to energetic cascade coincidence gamma rays.

2. Instrument Examples. Fast neutron detector wells employing either plastic or liquid scintillators are available from commercial vendors. The well employing a plastic scintillator is marketed as an active assay system (see Sec. IV.B); however, it is readily converted to a passive unit by removing the neutron interrogation sources.\footnote{73} The unit employing the liquid scintillator also uses a NaI gamma-ray energy spectrometer.\footnote{74}
measures the 400-keV region characteristic of \(^{239}\text{Pu}\). When sufficient quantities of plutonium are present the NaI measurement can be combined with the coincidence measurement to estimate the isotopic composition of the plutonium. Both units use electronic circuits that permit selection of the coincidence order.

3. Expected Performance. The two commercially available fast neutron scintillation detector wells have approximately the same \(^{240}\text{Pu}\) detection limit (60-90 mg). The performance of these coincidence detectors would be improved by using a 4π detector configuration. The improvement would yield a more uniform response throughout the sample volume and a lower detection limit. An improvement factor of 2.2 is obtained as the efficiency is increased from 15% to 25%. Thus, the passive measurement with fast neutron scintillation counters can expect to obtain a 27-mg detectability limit for \(^{240}\text{Pu}\).

IV. ACTIVE ASSAY TECHNIQUES

α Alpha-n Interrogation and Thermal Neutron Detection

1. Measurement Principle. An NDA instrument employing alpha-n interrogation with thermal neutron detection is generically referred to as an Active Well Coincidence Counter (AWCC).\(^{75}\) The measurement principle of the AWCC is based on neutron interrogation and detection of the induced fission neutrons by coincidence counting. Small \(^{241}\text{AmLi}\) neutron sources installed in a standard coincidence counter effectively convert the passive assay unit to an AWCC. The technique is best suited for measuring \(^{233}\text{U}\) and \(^{235}\text{U}\) because of their high fission cross section and the low spontaneous fission rate for all the uranium isotopes. For low-level plutonium measurements passive assay is the preferred choice.

The AmLi sources are mounted in an assembly that tailors the interrogating neutron energy spectrum for the particular measurement application. For waste measurements the tailoring assembly moderates the neutrons to near thermal energies where the fission cross section is largest for the fissile uranium isotopes. The thermal neutron interrogation maximizes the fission rate for a given source strength and thereby improves the instrument's detectability limit. The singles background from the source is reduced by designing the neutron detectors and source tailoring assembly to yield a higher detection
efficiency for the induced fission neutrons than for the interrogation neutrons. For small sample systems a detection discrimination ratio of 5 to 1 can be achieved. However, for large items such as 55-gal barrels, where a 4π geometry is used to maximize efficiency, improving the detection ratio is difficult. Even after reducing the relative interrogation neutron efficiency the singles background rate from the source still limits the detectability limit for the technique.

2. Instrument Example. The AWCC assay technique was developed to permit passive coincidence counters employed for field measurements of plutonium to be modified for the assay of $^{233}$U and $^{235}$U. The modification consists of inserting end plugs that contain a properly tailored AmLi source while using the same detectors and electronic package developed for passive plutonium assay. Figure 11 shows a schematic diagram of a well counter designed specifically for use as an AWCC unit. The system is mounted on a hand truck for mobility. Specifications for the unit shown in Fig. 11 are given in Table IX.

3. Expected Performance. For a 13-liter sample volume the AWCC unit in the thermal mode has a detectability limit of about one gram of fissile material. In extrapolating to the expected performance for a barrel-size unit, some assumptions will be necessary because neither test measurements nor calculations have been made to optimize the detectability limit of 55-gal barrel AWCC unit.

The background coincidence rate for an AWCC is dominated by

Fig. 11. Schematic diagram of the Active Well Coincidence Counter (AWCC) (LASL Neg. #79-2547).
TABLE IX
ACTIVE WELL COINCIDENCE COUNTER (AWCC)
SPECIFICATIONS

<table>
<thead>
<tr>
<th>Specification</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of $^{241}$AmLi Sources</td>
<td>2</td>
</tr>
<tr>
<td>Total neutron source strength</td>
<td>$10^5$ neutrons/s</td>
</tr>
<tr>
<td>Detector type</td>
<td>$^3$He-filled tubes</td>
</tr>
<tr>
<td>Detection efficiency</td>
<td>30%</td>
</tr>
<tr>
<td>Signal</td>
<td>Coincident neutrons</td>
</tr>
<tr>
<td>Electronics</td>
<td>Shift register circuit</td>
</tr>
<tr>
<td>Coincidence gate time</td>
<td>64 μs</td>
</tr>
<tr>
<td>Sample volume</td>
<td>4 liters</td>
</tr>
<tr>
<td>Detectability Limit (thermal interrogation)</td>
<td>$1 \text{g } ^{235}\text{U}$</td>
</tr>
</tbody>
</table>

accidental coincidences from the $^{241}$AmLi source. Neglecting backgrounds other than from the source and assuming a shift register module is being used, the coincidence background rate is given by

$$b_c = (S\epsilon_b)^2\tau_g$$  \hspace{1cm} (2)$$

where

$S$ = AmLi source strength (neutrons/s),
$\epsilon_b$ = Source neutron detection efficiency, and
$\tau_g$ = coincidence gate time.

The coincidence signal rate from fissions induced by the source is given by
where $e_1$ is the number of induced fissions per source neutron per gram of fissile material, and $F$ is the number of coincidence counts per induced fission as defined by Eq. (1). Using the expressions in Eq. (2-3) and Eq. (A-10) the detectability limit for the AWCC is given by

$$m = \frac{3e_b(2\tau_g/T)^{1/2}}{\epsilon_1 F}.$$  

From inspection of Eq. (4) the detectability limit is independent of the source strength. The assumptions used in deriving Eq. (4) are valid as long as the source is intense enough to dominate all the other backgrounds but not so intense as to result in count rate losses caused by pulse pileup. An interrogation source in the range of $10^4$ to $10^5$ neutrons/s satisfies the two bounding criteria.

The lowest detectability limit for the AWCC is achieved by minimizing the efficiency for detecting source neutrons while maximizing the interrogation efficiency and detection efficiency for fission neutrons. The optimum detectability parameters must also be achieved uniformly over the volume of the barrel. In general optimizing a particular parameter comes at the expense of another parameter and an actual significant improvement in the detectability limit is difficult. In estimating the detectability limit for 55-gal barrels the following data is used. The interrogation efficiency ($\epsilon_1$) for uranium is $1.8 \times 10^{-4}$ fission per source neutron per gram $^{235}$U, as estimated from the $^{239}$Pu fission rate in 55-gal barrels and the ratio of the $^{235}$U to $^{239}$Pu thermal fission cross sections. The detection efficiency for interrogation neutrons ($\epsilon_b$) and fission neutrons ($\epsilon$) is assumed equal at 25%. A coincidence count rate per fission ($F$) is computed as 0.16 based on $\nu = 2.43$ for $^{235}$U. The expected detectability limit for the AWCC for a 55-gal barrel is then estimated at 23 g $^{235}$U. The use of lead shielding does not affect the above estimate. Some optimization may be possible with calculational or test bed studies; however, an improvement of better than a factor of two is not expected.
If the AWCC were used in both an active and passive mode, then 500 s could be allotted to each measurement. The active assay would yield a detectability limit of 33 g $^{235}$U and for plutonium 19 and 23 g for weapons grade and reactor grade, respectively. The plutonium detectability limit is based on the $^{239}$Pu content of the two grades (see Table VI). The passive neutron assay would have detectability limits a factor of square root of two (1.4) larger than those listed in Tables VII and VIII because of the shorter assay time.

B. Alpha-n Interrogation and Fast Neutron Detection

1. Measurement Principle. A moderated alpha-n neutron source is used to interrogate the waste barrel, and fast neutrons from the induced fissions are detected by proportional counters filled with either helium, methane, or hydrogen (see Sec. II.A). The implementation of the measurement scheme is simple enough to permit the technique to be used for a hand-held instrument that verifies the $^{235}$U content of reactor fuel assemblies or measure in-plant holdup. However, matrix effects are potentially quite serious and the technique is not recommended for general purpose waste assay.

The alpha-n neutron source yields a considerable fast neutron background in the detectors even after moderation. The background rate from the source can in principle be determined by measuring a barrel that contains only the matrix material of the waste category being measured. When a barrel of waste is assayed, the additional count rate above background can be interpreted as the presence of fissile material. However, a difference in the matrix material of the waste can affect the background rate. If the waste item has a less moderating matrix, then the waste item assay will indicate the presence of fissile material when actually only the background increased. On the other hand, if the waste item moderates more than the standard, then the fast neutron background will be reduced. The reduced background causes the fissile assay to be low and perhaps even indicate a negative quantity. The potential for a false low or high assay caused by something as innocent as a change in the moisture content makes the technique questionable for waste assay applications.

2. Instrument Example. The NDA technique of alpha-n interrogation and fast neutron detection was developed for verification of the $^{235}$U content of light water reactor (LWR) fuel assemblies. A photograph of the instrument is
shown in Fig. 12. The unit has a weight of 18 kg and the AmLi source strength is $5.5 \times 10^5$ neutrons/s. The instrument was designed for field use by the IAEA to detect the removal or substitution of fuel rods in an assembly.

3. Expected Performance.
Although the technique of alpha-\(n\) interrogation and detection of fast neutrons from the induced fissions is not recommended for waste assay, the detectability limit will be estimated for comparison with the other techniques. In addition the data used to calculate the detectability limit can be employed to estimate the magnitude of matrix effects.

The signal rate from the induced fissions is given by

$$r_s = S\varepsilon_1\nu\varepsilon$$  \hspace{1cm} (5)

where

- \(S\) = AmLi source strength ($10^6$ neutrons/s),
- \(\varepsilon_1\) = induced $^{235}$U fission rate ($1.8 \times 10^{-4}$ fissions/source neutron/g, see Sec. IV.A.3),
- \(\nu\) = fission multiplicity (2.43 for $^{235}$U), and
- \(\varepsilon\) = fast neutron detection efficiency (0.0025).

The fast neutron detection efficiency ($\varepsilon$) assumes 50-mm-diam, 18-atm $^4$He.
tubes covering half of the available detection area. The resulting signal rate \( (r_s) \) is 1.1 counts/s/g \(^{235}\text{U}\).

The fast neutron background for a barrel-size system is assumed to originate from unmoderated source neutrons. For the AmLi source neutrons the macroscopic cross section for hydrogen neutron scattering in polyethylene is about 0.3/cm. Even with a relatively thick moderator (5 cm polyethylene) 20% of the source neutrons are unscattered. The detection efficiency for the AmLi source neutrons is taken to be one-third of the fission neutron value to account for the difference between the \(^4\text{He}\) scattering cross section at the AmLi alpha-n neutron energy and the fission neutron energy. Ignoring the moderation effects of the item being assayed, the background rate is then estimated at 170 counts/s. Thus, the signal-to-background ratio is 0.6% per gram of \(^{235}\text{U}\). If the background rate is well characterized by repeated assays of standards, then the detectability limit is 1.4 g \(^{235}\text{U}\).

A rough estimate of the effect of the matrix material on the source background will now be made. As an example of matrix effects, a variation in the moisture content of the waste will be considered. For a barrel with 100 kg of waste, the addition or loss of 1 kg (1 liter) of water represents a 1% change in the matrix material by weight. (The variation of moisture content of earth is typically much larger than 1%). The macroscopic scattering cross section at the mean AmLi source neutron energy for a liter of water uniformly dispersed throughout a 55-gal barrel is \(1.3 \times 10^{-3}/\text{cm}\). If the diameter of the barrel (56 cm) is taken as the path length and only half of the source neutrons, which contribute to the background, traverse the barrel, then the change in the background rate associated with the liter of water in the matrix material is 3.6%. The reduction in the source background with the addition of 1 kg \(\text{H}_2\text{O}\) is equivalent to about 5.6 g \(^{235}\text{U}\) signal. Thus, for even a modest change in the matrix material, the assay value for the fissile content can be shifted by an amount several times the detectability limit.

Although no instruments have been built for passive neutron assay employing recoil proportional gas-filled neutron detectors, an estimation of the detectability limit with these detectors will be made to check the feasibility of using the instrument in an active/passive mode. An estimation of the background rate is based on the cosmic-ray-induced neutron background in thermal well counters but only one-hundredth as large to account for the difference in efficiency. Thus, the background is estimated at 0.15 counts/s.
With the passive plutonium total emission rate for PuO$_2$ given in Table VI, the detectability limit is calculated using Eq. (A-10) at 174 mg and 83 mg for weapons grade and reactor grade plutonium, respectively. Coincidence counting is possible but the largest contribution to the errors is a one count uncertainty added in quadrature with the standard error estimations of the total counts. The coincidence detection limit is 38 g for weapons grade plutonium and 15 g for reactor grade.

C. Alpha-n Interrogation and Neutron Detection with Scintillators
   1. Measurement Principle. The NDA instruments employing an alpha-n neutron interrogation source and detecting fission neutrons with scintillation counters are generically referred to as Random Drivers. An $^{241}$AmLi source is used to induce fissions in the sample and prompt fission neutrons are counted in coincidence with the scintillation counters. Because energetic gamma rays are released in the fission process and the scintillation counters are also sensitive to gamma rays, some Random Driver units use the coincidences from combinations of energetic gamma rays and neutrons. Other units attempt to reduce the gamma-ray contribution to the coincidence total. Because fission gamma rays are emitted with a multiplicity of 6-7, counting both gamma rays and neutrons considerably improves the detectability limit by increasing the available signal. The 1-2 MeV fission gamma rays have more attenuation in dense samples than do neutrons; for low-density hydrogenous materials the opposite is true.

   The coincidence requirement eliminates the singly emitted neutrons from the AmLi source. Because gamma rays and fast neutrons are detected, the coincidence resolving time is determined by the differences in the time-of-flight. For gamma rays, which always travel at the speed of light, the time differences result from the differences in the path lengths to the various detectors and the optical path length within the scintillators. Barrel-size systems can expect about a 10 ns resolving time for gamma rays. The resolving time for neutrons is longer because their velocity is always less than gamma rays and depends on the neutron energy. The resolving time for neutrons is about 100 ns in barrel-size systems. Because the coincidence resolving time is about a thousand times shorter than for the thermal neutron wells, the source contributes a much smaller accidental coincidence background and a more intense source may be used to increase the signal rate.
2. Instrument Example. An instrument with a 55-gal barrel assay chamber is available from National Nuclear Corporation (NNC). Their unit, called a Jumbo Random Driver, is shown in Fig. 13. Two $^{238}\text{PuLi}$ sources yielding about $10^6$ neutrons/s are used to achieve a uniform interrogation of the barrel without employing a turntable. The scintillation detectors surround the assay chamber on four sides. Each side is operated as a single detector to allow coincidences of 2/4, 3/4, and 4/4. The detectors count both neutron and gamma-ray signals; however, an electronic coincidence circuit designed to eliminate acceptance of coincidences between gamma rays is optional. The response has a root mean square (rms) variation of 9% over the assay chamber, based on data given in the NNC descriptive literature.

3. Expected Performance. The Jumbo Random Driver for 55-gal barrels has a detectability limit of 230 mg $^{235}\text{U}$ and 175 mg $^{239}\text{Pu}$ calculated from data supplied by the manufacturer. In certain field applications a somewhat better performance has been observed, with detectability limits of 70 mg $^{235}\text{U}$ and 115 mg $^{239}\text{Pu}$. Because scintillation detectors are used, the instrument is suitable for assaying only cold waste.

If the Jumbo Random Driver is used as an active/passive assay system, then half of the assay time (500 s) is spent in each mode. Thus, the passive detectability limits based on the equivalent $^{240}\text{Pu}$ content is 1.3 g weapons grade plutonium and 0.5 g reactor grade plutonium. The active assay detectability limits are 330 mg $^{235}\text{U}$, 265 mg weapons grade plutonium, and 300 mg reactor grade plutonium.

Fig. 13. Jumbo Random Driver (photograph courtesy of National Nuclear Corporation).
D. Photoneutron Interrogation and Thermal Neutron Detection

1. Measurement Principle. There are two approaches available for separating the photoneutron interrogation neutrons from fission neutrons when gas-filled thermal neutron detectors are employed. The approaches can rely on either the multiplicity of the fission neutrons or on the greater penetrability of the fission neutrons compared to photoneutrons. If the coincidence approach is used, then the instrument is very similar to the AWCC described in Sec. IV.A with the exception that the photoneutron source replaces the $^{241}$AmLi source. However, the high gamma-ray fields and short half-life of the photoneutron source are much less convenient than the $^{241}$AmLi source for the AWCC application.

The greater penetrability of the fission neutrons compared to $^{124}$Sb-Be photoneutrons has been employed for small sample assay of irradiated uranium-thorium pebble bed reactor fuel. Figure 14 schematically shows the assay technique. The 6-cm-diam carbide sphere is irradiated with neutrons from the $^{124}$Sb-Be source. A solution of boric acid between the sample and detectors preferentially absorbs the source neutrons. The net effect is that the signal rate is 200 counts/s/g $^{235}$U while the background from the source is only 1 count/s. Because the irradiated fuel sphere has a fission product activity of 100 Ci, the 100 Ci $^{124}$Sb source places a little additional burden on the gamma-ray shielding requirements.

2. Instrument Examples. The technique of photoneutron interrogation with detection of penetrating fission neutrons with thermal neutron detectors has been employed at the Kernforschungsanlage (KFA) Julich, Institute of Chemical Technology. A

Fig. 14. Assay system constructed within a 55-gal barrel for measurement of irradiated pebble bed reactor fuel (EG&G Neg. #10257).
unit designed to assay both small and medium sized items is shown in Fig. 15. Small samples with up to a 6 cm diam are inserted in the top access hole. Medium sized samples are scanned horizontally. The cross-section opening for the scanning measurements is 21 cm high by 28 cm wide.

3. Expected Performance. The proposed barrel-size assay unit shown in Fig. 16 was designed primarily for safeguard's measurement of waste rather than environmental monitoring. Thus, the detectability limit was not optimized nor was a detectability limit given other than an implied limit of better than one gram fissile. Based in part on data available in the literature, a detectability limit will be estimated. An engineering study of the problem is beyond the scope of this report.

The signal rate is given by

$$I_S = S_{\epsilon_1} \nu_{\epsilon_1} / A_f$$  \hspace{1cm} (6)
where

\[ S = \text{neutron source strength (}3.2 \times 10^8/\text{s for 100 Ci }^{124}\text{Sb, ref. 86)},\]
\[ \varepsilon_i = \text{induced }^{235}\text{U fission rate (}1.8 \times 10^{-4}/\text{source neutron, see Sec. IV.A.3)},\]
\[ \nu = {^{235}\text{U fission multiplicity (2.43)},}\]
\[ \epsilon = \text{detection efficiency (25%), and}\]
\[ A_f = \text{attenuation of fission neutrons (22, ref. 85).}\]

The background rate from the source is given by

\[ b = \frac{Se}{A} \quad (7) \]

where

\[ A = \text{attenuation of the source neutrons (80 000, ref. 85).} \]

Thus, the signal is \(1.6 \times 10^7\) counts/s/g \(^{235}\text{U}\), and the background rate from the source is \(1.0 \times 10^3/\text{s}\). Other contributions to the background are negligible. Equation (A-10) is used in estimating the detectability limit where it is assumed that half the assay time is spent measuring the source background. The detectability limit for \(^{235}\text{U}\) is then estimated at 3.8 mg. The detectability limit for \(^{239}\text{Pu}\) is slightly lower at 2.5 mg. Thus, the method of photoneutron interrogation with penetrating fission neutron detection appears to have the potential for a low detectability limit.

Because the interrogation neutrons contribute a sizable fraction of the background, the matrix material can affect the background rate and in turn result in erroneous estimations of the fissile content (see Sec. IV.B). If the matrix variation is taken to be the addition of one liter of water and the neutron absorption length of 2.2 cm for the 26 keV interrogation neutrons is used, then about 10% of the source neutrons traversing the barrel are absorbed. Assuming half of the source neutrons traverse the barrel, then the background rate is decreased by 5% (50 counts/s). The corresponding \(^{235}\text{U}\) quantity is 81 mg. The matrix effect can be reduced by increasing either the quantity or concentration of boric acid (or other low energy neutron absorbing material) between the barrel of waste and the detectors or by additional moderation of the source neutrons (see Sec. IV.G). In addition the matrix effects might be
measured by placing some detectors closer to the barrel where the effect is more pronounced. Thus, the matrix variation appears to be a manageable problem.

Employing a 100 Ci $^{124}\text{Sb}$ gamma-ray source will necessitate lead shielding for the detectors. The detectors within 0.3 m of the source will experience approximately a 1000 R/h radiation dose rate. Thus, the unit will require about 15 cm of lead shielding around the interrogation source. If the barrels are to be manually inserted into the assay chamber, then additional shielding would be required. However, if the facility were remotely handling high-level radioactive waste then the extra personnel shielding would not be required. Because of the high gamma-ray activity accompanying the source, the technique of photoneutron interrogation and detection of penetrating neutrons is best suited for facilities handling high beta gamma wastes, reprocessing plants.

The $^{124}\text{Sb-Be}$ interrogation system could be operated as an active/passive unit; however, the absorptive shielding surrounding the detectors reduces their efficiency resulting in a degraded passive detectability limit. For a 500-s passive assay (the $^{124}\text{Sb-Be}$ source is withdrawn) the singles detectability limit is 235 mg for weapons grade plutonium and 115 mg for reactor grade plutonium ($\text{PuO}_2$). The detectability limits for the coincidence counts including the effects of the lead shield are estimated at 15 and 10 g $\text{PuO}_2$ for weapons and reactor grade, respectively. The passive counts add little to the utility of the $^{124}\text{Sb-Be}$ unit because of the poor detectability limits compared to the active mode. Thus, an active/passive operation is not recommended.

The short half-life of the $^{124}\text{Sb}$ source makes renormalization of the calibration constants important because both the signal and background rates change with the decaying source. The renormalization procedure should include standards measurements several times a shift coupled with either a computation of the source strength or a measurement of the source strength performed as part of the assay. In addition to the radioactive decay of the source, the background and signal rates are affected by density changes caused by temperature drifts in the boric acid surrounding the assay chamber. Because of the massiveness of the system the drifts will be slow and can be monitored by standards measurements with additional corrections based on temperature measurements.
E. Photoneutron Interrogation and Fast Neutron Detection

1. Measurement Principle. A photoneutron source, usually $^{124}$Sb-Be, is used to interrogate a sample and $^4$He (or CH$_4$ or H$_2$) gas-filled detectors count fast neutrons from fission. The $^4$He detectors are biased to count only neutrons with energies above the 26- and 360-keV source neutrons.

2. Instrument Examples. A variety of NDA instruments have been built employing the method of photoneutron interrogation and fast neutron detection. A plan view of a system designed to assay small diameter samples such as fuel pellets or rods is shown in Fig. 17. The nickel cylinder surrounding the source reflects interrogating neutrons into the sample region to increase the neutron flux. The nickel cylinder drilled for the detectors shown in Fig. 17 reflects fission neutrons back toward the $^4$He neutron detectors.

Figure 18 shows a system with a $^{226}$Ra-Be photoneutron source employed to inventory fuel elements after the termination of the Nuclear Propulsion Reactor Testing Program. The fuel was packaged in cardboard tubes 70 mm in diameter and 1.37 m long. The fuel from 14 reactors totaling about 731 kg uranium was inventoried and the agreement between the measured value and the book value was better than 0.2%. $^{28,87,88}$

A prototype system was developed at Karlsruhe to measure LWR fuel assemblies. The unit employed a $^{124}$Sb-Be source and methane-filled fast neutron detectors. The $^{235}$U
Fig. 18. Assay system for the measurement of irradiated Rover fuel. Top: Data acquisition and analyses electronics located outside the hot cell. Bottom: Fuel tube positioned in front of the entrance to the $^{226}\text{Ra-Be}$ neutron interrogation region. The $^3\text{He}$-filled fast neutron detectors are located on the sides of the interrogation assembly. The hot cell environment protects the operator from the dose from the $^{226}\text{Ra-Be}$ source and the irradiated fuel (LASL Neg. #CN76-6811).

Fig. 19. Uranium borehole logging sonde (EG&G Neg. #10202).
measurement accuracy for various fuel rod configurations was approximately 1%.\textsuperscript{89}

In addition to the application to fuel elements the technique of photoneutron interrogation and fast neutron detection is being developed for uranium borehole logging.\textsuperscript{90} Figure 19 shows a schematic diagram of the sonde. The sonde is expected to log the boreholes at a rate of approximately one meter per minute and to detect ore concentrations as low as 200 parts per million (ppm).

3. Expected Performance. An instrument designed for assaying barrels with a photoneutron interrogation and fast neutron counting has not been built. The signal rate will be based on thermal neutron interrogation of barrels and the efficiency of fast neutron detectors. The background rates will be estimated from the existing smaller NDA instruments.

The signal count rate is given by

$$r_s = S \varepsilon_1 \nu \epsilon$$  \hspace{1cm} (8)

where:

- $S$ = neutron source strength (3.2 x 10\textsuperscript{8} s for 100 Ci \textsuperscript{124}Sb),
- $\varepsilon_1$ = induced \textsuperscript{235}U fission rate (1.8 x 10\textsuperscript{-4} fissions/source neutron/g, see Sec. IV.A.3),
- $\nu$ = fission multiplicity (2.43 for \textsuperscript{235}U), and
- $\epsilon$ = fast neutron detection efficiency (0.0025).

Thus, the signal count rate ($r_s$) is 350 counts/s/g \textsuperscript{235}U. The $^4$He detectors are biased above the 26 keV and 360 keV neutrons from the source; however, a fraction of the 360 keV source neutrons and neutrons with energies up to 1 MeV produced by small branching ratio gamma rays yield recoil interactions that require a rather high threshold setting that reduces the signal from the fast neutrons from the fission process. In addition gamma-ray pulse pileup from the $^{124}$Sb source makes the selection of the threshold setting more difficult. The lower threshold settings result in increasing the background, and higher threshold settings decrease the signal rate. The background rate for the barrel-size system is estimated by extrapolating from the optimum conditions.
arrived at for smaller systems and multiplying by the ratio of the total detector size. From the small sample assay unit shown in Fig. 17 the estimated background rate is 250 counts/s and from the uranium borehole logging sonde shown in Fig. 19 the background rate is estimated at 150 counts/s. These two estimations are remarkably close and the background is taken as the lower of the two estimations.

An additional background results from the 1.4 ppm concentration of $^3$He found in natural helium. The small concentration of $^3$He yields a signal from the interrogation source of about 500 counts/s. Wrapping the detectors with cadmium has been observed to reduce this background approximately an order of magnitude. Thus, combining the background sources the background rate ($r_b$) for a barrel-size sample is estimated at 200 counts/s. Using Eq. (A-10) the detectability limit is estimated at 8 mg $^{235}$U and for $^{239}$Pu the limit is 5 mg.

When the detection efficiency for fast neutrons was estimated, the potential effect of the matrix material moderating the fast fission neutrons below the detection threshold was not considered. A detailed examination of this problem should be made with a test bed or Monte Carlo calculations. Because both of these approaches are beyond the scope of this report, some simplifying assumptions will be made in order to estimate the size of the effect. First we will assume the pulse height in the detector is equally probable for all values up to the value corresponding to the maximum energy allowed by the kinetics. This assumption makes the detection probability proportional to the difference between the neutron energy and the detection threshold ($\sim 0.5$ MeV). The second assumption is that the primary moderating material is hydrogen for which the "average" scattering cross section is taken as 4 b for the fast fission neutrons. If the typical path length is taken as the barrel radius (28 cm), then the average number of collisions is $3.6 \times 10^{-2}$/liter H$_2$O. Using the assumed model for the fast neutron detection the one liter of water results in a detection efficiency loss of about 4%. Thus, for the technique to be reasonably accurate the hydrogenous content of the standards should closely match the typical material. The total hydrogenous content should be limited to less than the equivalent of about 10 liters of uniformly dispersed water per 55-gal barrel and matrices containing plastics should be avoided.
F. Californium-252 Interrogation and Delayed Thermal Neutron Detection

1. Measurement Principle. The fissile isotopes present in the waste material are measured by counting delayed neutrons after the item has been irradiated by a $^{252}$Cf neutron source. Instruments that assay containers consist of an assay chamber and an isolated source storage position. The source is positioned in the storage container for background and delayed neutron counting and the source is transferred to the assay chamber for the neutron irradiation of the item. During the assay the source is cycled between the assay chamber and storage position. Because of the many transfers of the source, this particular instrument type is often generically referred to as a "$^{252}$Cf Shuffler."

Figure 20 shows a schematic diagram of a $^{252}$Cf Shuffler for 55-gal barrels. The assay chamber is completely surrounded by $^3$He-filled detectors to maximize the efficiency and uniformity of the neutron detection. Detector tubes filled with $^{10}$BF$_3$ gas cannot be used in the $^{252}$Cf Shuffler because
of the build-up of electronegative gases (fluorine) during the irradiation by the $^{252}\text{Cf}$ source.\textsuperscript{17,18}

Two irradiation positions are shown in Fig. 20. The purpose of the two positions is to provide a more uniform interrogation of the barrel. Implementation of the scheme could be accomplished by using two sources (one for each position) or by having one source alternate between the two positions. A different approach to achieving a uniform interrogation would have the source translate vertically along the edge of the barrel as shown in Fig. 21. This latter option would be more flexible in terms of optimizing the uniformity for different container types or matrix materials. Other Shuffler instruments have been designed to translate long items past the interrogation-counting region to achieve a uniform assay.\textsuperscript{95} The systems shown in Figs. 20 and 21 rotate the barrel during the assay to make a more uniform neutron interrogation in the radial dimension.

The neutrons emitted from the californium source have a fission energy spectrum with an average energy of about 2.3 MeV. Moderating material surrounds the source at the interrogation position (indicated in Fig. 21) so that the barrel is irradiated with thermal- as well as high-energy neutrons. The low-energy neutron irradiation enhances the response because of the increased fission cross section at thermal energies. The high-energy neutrons can penetrate to the central regions of a barrel before thermalizing. The response can also be increased by using a more intense $^{252}\text{Cf}$ source. Practical tradeoffs between the desired detectability limit for a reasonable assay time and the quantity of radiation shielding needed for personnel safety result in a $^{252}\text{Cf}$ source size of about 2-5 mg or a maximum neutron emission rate of $10^{10}$/s. The $^{252}\text{Cf}$ half-life is 2.65 years and the initial source strength is usually selected to provide about five years of service.

![Fig. 21. Conceptual drawing for a $^{252}\text{Cf}$ Shuffler assay unit. The californium source is scanned axially past the barrel during interrogation (EG&G Neg. #10248).](image-url)
The lead liner as shown in Fig. 20 is 15 cm thick to allow the unit to measure items with gamma-ray levels as high as 1000 R/h. As with the passive well counters, the lead shield can contribute to the overall background and thus no more lead than is required for shielding should be installed. Figure 21 shows a lead shield 5 cm thick for measurement of 10 R/h material. Shielding beyond the detector banks reduces the room neutron background in the assay chamber and it reduces the exterior radiation dose from the $^{252}\text{Cf}$ source during the sample irradiation.

The storage position for the $^{252}\text{Cf}$ source is surrounded by massive quantities of neutron and gamma-ray shielding material. The shielding is usually designed to keep the personnel exposure dose to less than a few mR/h on contact. In addition the shield minimizes the neutron leakage back to the assay chamber. One or more bends are incorporated in the transfer tube to reduce the number of neutrons streaming to the assay chamber.

The assay begins with the source in the storage position and the barrel in the assay chamber. The data accumulated in this initial portion of the assay is equivalent to a passive neutron assay and both the total and coincidence neutron rates can be measured. The passive phase of the assay accounts for about one-third of the total assay time. The active portion of the assay is cyclical with the irradiation time of each cycle being about 16 s and the delayed neutron counting time being about 8 s. A delayed neutron counting time shorter than the irradiation time is used to increase the assay precision by improving the delayed neutron signal-to-background ratio. The source is transferred with a high-speed, computer-controlled stepping motor. The transfer time is typically 0.5 s in either direction and results in a total time of 25 s for one complete active assay cycle. For low-level counting about 30 active cycles are used.

2. Instrument Examples. A $^{252}\text{Cf}$ Shuffler was installed at the Savannah River Plant (SRP) reactor fuel fabrication facility for measurement of a range of scrap and waste materials. The material is packaged in 7.6-liter (18 cm diam, 30 cm height) cans that contain up to 3 kg $^{235}\text{U}$. Figure 22 shows a photograph of the system before installation. The unit functions primarily for accountancy and quality control so that a greater emphasis was placed on the linearity of the response at the expense of the detection limit. The operating characteristics of the unit are given in Table X.
TABLE X
SAVANNAH RIVER PLANT 252Cf SHUFFLER
PERFORMANCE DATA

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>252Cf source strength</td>
<td>0.8 mg</td>
</tr>
<tr>
<td>Assay time (typical)</td>
<td>8 minutes</td>
</tr>
<tr>
<td>(maximum)</td>
<td>19 minutes</td>
</tr>
<tr>
<td>Assay precision (typical)</td>
<td>0.3%</td>
</tr>
<tr>
<td>(achievable)</td>
<td>0.1%</td>
</tr>
<tr>
<td>Delayed neutron count rate</td>
<td>1.6%</td>
</tr>
<tr>
<td>Delayed neutron linearity (stacked SRP U-Al disks to 3 kg 235U)</td>
<td>+1.6%</td>
</tr>
<tr>
<td>Detectability (3σ, 19 minutes)</td>
<td>1 gram 235U</td>
</tr>
</tbody>
</table>

Fig. 22. Savannah River Plant (SRP) 252Cf Shuffler for the assay of scrap and waste in the uranium fuel fabrication facility (LASL Neg. #CN78-B6496).

Fig. 23. Neutron interrogator designed for the Idaho Chemical Processing Plant (LASL Neg. #CN79-4576).
Figure 23 shows a unit that is being built for the Fluorinel and Storage (FAST) facility at the Idaho Chemical Processing Plant (IClP). Spent high enrichment fuel elements and solid waste produced during the fuel dissolution will be measured at the assay port specifically designed for each of the two material types. The fuel elements and waste canisters are translated past the neutron interrogation and counting region to achieve longitudinal uniformity. In addition the $^{252}\text{Cf}$ source is moved around the fuel element region (handling procedures preclude rotation) to obtain radial uniformity.$^{95,99}$

A borehole logging service that employs the technique of $^{252}\text{Cf}$ neutron interrogation and delayed neutron counting is commercially available. The probe is 57 mm (2-1/4 in) in diameter and 5.2 m (17 ft) long. Helium-3 detectors are located at both ends and the $^{252}\text{Cf}$ source is internally transferred back and forth between the ends of the probe. The logging consists of a background measurement when the probe is lowered into the borehole and an active measurement as the probe is withdrawn. During the active portion, the two sets of detectors permit neutron interrogation at one end of the probe and delayed neutron counting at the opposite end. In this mode of operation the source does "double duty." The uranium ore sensitivity is 100 ppm at a logging rate of 1 meter per minute. The sonde can also be used for detecting plutonium and americium migration through rock formations and ground waters in the vicinity of nuclear waste disposal sites.$^{101}$

The first Shuffler-type NDA instrument is shown in Fig. 24. This instrument featured two interrogation modes. A thermal mode was used to obtain a sensitive assay for samples containing only small quantities of fissile material. A fast interrogation mode was used for samples containing more than several grams of fissile material or high enrichment uranium. The thermal mode employed a lead and polyethylene neutron spectrum tailoring assembly and a bare polyethylene counting well, whereas the fast mode employed a tungsten, beryllium, and nickel neutron spectrum tailoring assembly and a cadmium liner was inserted into the assay chamber. The characteristics of the fast and thermal modes of the unit shown in Fig. 24 are summarized in Table XI.$^{94}$

An assay chamber for 55-gal barrels was assembled for the prototype Shuffler shown in Fig. 24.$^{102}$ A schematic diagram of the test setup is shown in Fig. 25. The test bed included a 15-cm-thick lead shield to study effects of the massive quantity of lead (4800 kg) on the neutron detection characteristics and to observe the cosmic-ray-induced neutron background. A
TABLE XI
PROTOTYPE $^{252}$Cf SHUFFLER CHARACTERISTICS
WITH A 0.5 mg $^{252}$Cf SOURCE

<table>
<thead>
<tr>
<th>Neutron Interrogation</th>
<th>Thermal</th>
<th>Fast</th>
</tr>
</thead>
<tbody>
<tr>
<td>Net Signal Rate $^{235}$U</td>
<td>$1500$ counts/s/g $^{235}$U</td>
<td>$13$ counts/s/g</td>
</tr>
<tr>
<td>Background Rate</td>
<td>$20$ counts/s</td>
<td>$14$ counts/s</td>
</tr>
<tr>
<td>Detectability Limit</td>
<td>$0.4$ mg $^{235}$U (6 ppm U ore)$^a$</td>
<td>$40$ mg $^{235}$U</td>
</tr>
</tbody>
</table>

$^a$Four-liter sample

Fig. 24. Prototype Shuffler (LASL Neg. #CN74-5700).
Fig. 25. Barrel-size $^{252}$Cf Shuffler test bed schematic diagram (EG&G Neg. #10252).
total of 60 $^3$He-filled detectors (2.54 cm diam, 91 cm length) were placed on three sides of the assay chamber yielding an average neutron detection efficiency of 10%. Figure 26 shows a photograph of the barrel-size Shuffler test bed. The simulated waste matrix materials were available with insertion tubes for placement of fissile material at different radial and longitudinal positions within the barrel. The placement of samples at various positions permitted the comparison of the observed characteristics with those calculated by three-dimensional Monte Carlo neutron transport codes.

The Monte Carlo calculations were made in two steps. First the number of fissions induced in the barrel per source neutron was estimated. The fission rate was tallied by position in the barrel to check uniformity. Because of the single point irradiation used on the test bed the fission rate varied by nearly a factor of four over the barrel volume. In the second step of the Monte Carlo calculation the delayed neutron detection efficiency was obtained. The average detection efficiency was 10% but varied by a factor of three from the center to the ends of the barrel because the detectors were placed only on the sides, and end losses were enhanced by the 15-cm-thick lead shield. The agreement between the Monte Carlo calculations and the observed performance for the test bed was quite good, typically within ±15%. The slight differences are attributed largely to "simplifications" used in the calculation geometry.

Although the test bed was constructed to test the feasibility of low-level measurements (<1 g fissile), a request was made to use the instrument to assay highly enriched uranium (93% $^{235}$U) intermixed with furnace parts and reduction residues. The material was packaged in 30-gal (114 liter) barrels and the uranium content had been estimated using the "by difference" technique. In addition to the test bed a Segmented Gamma Scanner...
TABLE XII

$^{235}\text{U}$ CONTENT OF TWELVE 30-GAL BARRELS CONTAINING FURNACE PARTS AND REDUCTION RESIDUES MEASURED BY THE SGS AND THE BARREL-SIZE SHUFFLER TEST BED

<table>
<thead>
<tr>
<th>Drum No.</th>
<th>Net Weight (kg)</th>
<th>Estimated $^{235}\text{U}$ (g)</th>
<th>Measured $^{235}\text{U}$ a</th>
</tr>
</thead>
<tbody>
<tr>
<td>70</td>
<td>92</td>
<td>65</td>
<td>59.1 ± 2.2</td>
</tr>
<tr>
<td>79</td>
<td>31</td>
<td>9</td>
<td>53.8 ± 1.4</td>
</tr>
<tr>
<td>80</td>
<td>23</td>
<td>6</td>
<td>1.1 ± 1.0</td>
</tr>
<tr>
<td>81</td>
<td>56</td>
<td>16</td>
<td>4.8 ± 1.4</td>
</tr>
<tr>
<td>82</td>
<td>47</td>
<td>12</td>
<td>2.1 ± 1.0</td>
</tr>
<tr>
<td>83</td>
<td>126</td>
<td>61</td>
<td>55.9 ± 1.4</td>
</tr>
<tr>
<td>84</td>
<td>69</td>
<td>36</td>
<td>14.8 ± 0.8</td>
</tr>
<tr>
<td>85</td>
<td>68</td>
<td>35</td>
<td>19.2 ± 1.0</td>
</tr>
<tr>
<td>86</td>
<td>68</td>
<td>35</td>
<td>20.8 ± 0.9</td>
</tr>
<tr>
<td>87</td>
<td>69</td>
<td>30</td>
<td>21.6 ± 1.0</td>
</tr>
<tr>
<td>88</td>
<td>69</td>
<td>28</td>
<td>21.3 ± 0.9</td>
</tr>
<tr>
<td>89</td>
<td>39</td>
<td>33</td>
<td>18.8 ± 1.0</td>
</tr>
</tbody>
</table>

aErrors indicate statistical precision only (1-$\sigma$ level).

(SGS)$^{109}$ was used for passive counting of the 185-keV gamma rays from $^{235}\text{U}$. Both techniques used a single standard containing 46.4 g $^{235}\text{U}$ intermixed with paper in a 30-gal barrel to approximate the barrels of waste. The SGS used a gamma-ray transmission measurement to correct for the matrix differences between the standard and the waste barrels; however, no matrix corrections were made for the neutron-based test bed measurements. Table XII shows the results of the two NDA assay techniques along with their one-standard-deviation statistical precision. For the 10 barrels assayed by both the SGS and the test bed, the total $^{235}\text{U}$ inventory was 177.6 g and 165.9 g, respectively, a reasonable agreement for this type of material. (Two barrels were not assayed by the neutron technique because they were too heavy to transport to the test bed.) The higher count rate for the neutron technique gave better precision than the gamma-ray measurement. For the standard, the neutron NDA test bed count rate was 7.4 counts/s/g $^{235}\text{U}$; whereas, the SGS count rate was 1.8 counts/s/g.
However, because the waste barrels were more dense than the standard, the SGS count rate was reduced by about an order of magnitude.

3. Expected Performance. The test bed unit shown in Fig. 26 had an observed detectability limit of 42 mg $^{235}$U with a 0.2 mg $^{252}$Cf source. Figures 20 and 21 show systems designed to optimize the detectability limit in both the active and passive assay modes. Features in an optimized system include neutron detectors that completely surround the assay chamber, a large californium source (2-5 mg), thick external shielding (20-30 cm) to reduce room background, and a source transfer tube designed to minimize the number of neutrons from the source at the storage position reaching the assay chamber. In addition, if lead shielding is required (gamma-ray levels $>1\ R/h$), then as much overhead shielding as practical should be installed to eliminate cosmic rays.

The delayed neutron signal rate ($r_s$) is given by:

$$r_s = S \epsilon I_k \epsilon$$

where

- $S = 2^{252}$Cf source strength ($2\\text{mg}\ 2^{252}$Cf yields $5 \times 10^9$/s),
- $\epsilon_I = $ interrogation efficiency ($1.8 \times 10^{-4}$ fissions/source neutron/g-$^{235}$U; $2.3 \times 10^{-4}$ fissions/source neutron/g-$^{239}$Pu, see Sec. IV.A.3),
- $k = $ the fraction of fissions resulting in delayed neutron emitted during the counting portion of the assay ($k = 3.1 \times 10^{-3}$, $^{235}$U; $k = 1.5 \times 10^{-3}$, $^{239}$Pu), and
- $\epsilon = $ delayed neutron detection efficiency ($\epsilon = 0.25$).

The delayed neutron kinetics are based on the equilibrium values for an active assay cycle of 16-s irradiation, 8-s delayed neutron counting, and 0.5-s source transfer time in both directions. Thus the signal rates are 700 counts/s/g $^{235}$U and 430 counts/s/g $^{239}$Pu.

The background rates can be extrapolated from the test bed unit with allowance made for improvements in the design. The background contributions arise from ambient room neutrons, the $^{252}$Cf source at the storage position, and cosmic-ray interactions in the lead shielding. The room neutron rate in the test bed was 5 counts/s. Because more exterior shielding will be required to reduce the personnel exposure when the larger californium source is used,
the room background should decrease to about 1-3 counts/s. The transfer tube length in the test bed unit was 1.37 m long with one 3-deg bend midway to prevent a line-of-sight neutron path yielding 10 counts/s from the source. With the bends indicated in Figs. 20 and 21, better than an order of magnitude improvement is possible. Thus, with the larger source (2 mg $^{252}$Cf) a background count rate estimate of 10 counts/s is conservative. The cosmic-ray-induced background rate is difficult to estimate because it depends on the location selected for the instrument. A reasonable estimate would be the rate observed for the test bed of 15 counts/s. Thus, the total background rate is estimated at 28 counts/s. If the assay is assumed to consist of a 300 s background count followed by 28 active interrogation cycles for a total assay time of 1000 s, then Eq. (A-10) may be used to compute the detectability limit at 2.0 mg for $^{235}$U and 3.4 mg for $^{239}$Pu.

The background measurement performed at the beginning of each assay is equivalent to a passive assay with a well counter. The detectability limit for the passive portion of the assay can be estimated from the formulas developed for the passive well counters (see Sec. III.A.5). For the 5-min background count the Shuffler with a 15-cm-thick lead shield has a singles neutron detectability limit of 32 and 15 mg for weapons and reactor grade plutonium oxide, respectively. The coincidence counting modes yield detectability limits of 110- and 42-mg Pu for weapons and reactor grade, respectively.

In terms of stability and reliability for low-level detection measurements, delayed neutron counting has a definite advantage over the prompt neutron active interrogation techniques described here. For the prompt detection techniques the isotopic interrogation source generally dominates the background rate and the matrix material can affect the apparent background rate as well as the induced signal. Thus, as the quantity of fissile material in the barrel approaches the detectability limit, the matrix effects on the background and the induced signal must be corrected through standards that closely match the material being assayed both in composition and fill height. On the other hand, for the delayed neutron approach the induced signal (delayed neutrons) is measured under conditions identical to those before the signal was induced (background). Thus, no correction based on the barrel's effect on the background is necessary because a direct measurement is made at the start of each assay. With the delayed neutron technique, correction factor data collected during the assay (density, fill height, moderation, neutron
absorption, etc., see ref. 98) can be applied directly to the induced signal instead of the combination of induced signal and background rate as would be required for the prompt neutron counting approaches described here. Because of the simpler analysis of the assay data, the delayed neutron technique requires fewer calibration standards and matching the standards to the waste material being assayed is less critical.

G. Californium-252 Interrogation and Fast Neutron Detection

1. Measurement Principle. A strongly moderated $^{252}$Cf is used to interrogate the item, and fast neutrons from the induced fissions are detected with recoil type gas-filled proportional counters. The measurement principle is similar to that used for the instruments discussed in Sec. IV.B. However, because a fission source ($E' = 2.3$ Mev) is used instead of the $^{241}$AmLi source ($E = 0.5$ MeV), moderating the neutrons below the detection threshold of the fast neutron counters is even more difficult. Generally a specialized moderating material such as heavy water ($D_2O$) must be used. If hydrogenous material like polyethylene is used the neutrons that have been thermalized are absorbed by the hydrogen ($H$) before the remaining high-energy neutrons can be moderated below the detection threshold.

The use of less efficient moderators and the large degree of moderation required result in a thick moderating assembly surrounding the source. The moderation assembly increases the size of the instrument and results in the interrogation position being moved farther from the barrel being assayed.

2. Instrument Example. The technique of using a moderated $^{252}$Cf neutron source and detection of fast neutrons has been employed in fuel rod scanners. Figure 27 shows the placement of the source in the center of a ring of fuel rod assay channels to allow for a more efficient use of source neutrons by having up to six rods assayed simultaneously. The $^4$He-filled neutron detectors are placed close to the fuel rods to improve the fast neutron detection efficiency. The placement of the $^4$He tubes relative to the source and fuel rod was selected to yield a uniform response across the fuel rod.

3. Expected Performance. The advantages of this technique, which has been successfully employed for fuel rod scanners, are considerably diminished
when large sized barrels of waste with low fissile loadings are assayed. The exchange of the $^{252}\text{Cf}$ source for the $^{241}\text{AmLi}$ described in Sec. IV.B would not be expected to improve the detectability limit because the additional source strength available from $^{252}\text{Cf}$ would be offset by a higher background rate.

A possible alternative approach would be to use a moderator with a high moderation to absorption ratio such as heavy water ($D_2O$). To moderate a substantial fraction of the $^{252}\text{Cf}$ neutrons below the detection threshold of the $^4\text{He}$ proportional counters a heavy-water moderator would have to be quite thick ($\approx 30\text{ cm}$). With a thick moderating assembly the source must be positioned farther from the barrel resulting in a decreased interrogation efficiency that will require a larger source to achieve the desired induced fission rate. The use of a large quantity of heavy water has a serious drawback in that it increases the potential for a criticality accident. Two possible scenarios are 1) an item placed in the assay chamber becomes critical because of the highly reflective moderator or 2) the tank leaks and the heavy water collects in an area containing fissile material.

The detectability limit for the $^{252}\text{Cf}$ interrogation and fast neutron detection with recoil proportional gas counters will be estimated from the design and performance of the fuel rod scanner. The thick deuterium moderator assembly requires the $^{252}\text{Cf}$ source to be placed about 15 cm farther from the barrel than for the assemblies proposed for either $^{124}\text{Sb-Be}$ or $^{241}\text{Am-Li}$ neutron interrogation. With the larger separation distance the induced fission efficiency ($e_i$) is halved yielding $9 \times 10^{-4}$ fissions/source neutron/g for

![Fig. 27. Schematic diagram showing a cross section of the cylindrical LWR fuel rod assay system (EG&G Neg. #10263).]
and 1.2 × 10⁻⁴ fissions/source neutron/g for ²³⁹Pu. Assuming a 1-mg ²⁵²Cf source (2.3 × 10⁹ neutrons/s), Eq. (8) may be used to estimate the prompt neutron count rate (rₜ) at 1260 counts/s/g for ²³⁵U and 2000 counts/s/g for ²³⁹Pu.

The background rate includes energetic source neutrons being counted in the detectors as well as epi-cadmium neutrons interacting with the 1.4 ppm concentration of ³He found in natural helium. Comparing the neutron interrogation fluxes from the ²⁵²Cf and the ¹²⁴Sb-Be source (see Sec. IV.E.3) the background attributed to the ³He impurity is 200 counts/s. For each assay channel in the fuel rod scanner (two helium-filled detectors) the fast neutron background from the 0.1-mg ²⁵²Cf source was about 600 counts/s. Extrapolating to a barrel-size unit with a 1-mg ²⁵²Cf source, the fast neutron background rate is estimated at 2 × 10⁴ counts/s. Employing Eq. (A-20) the detectability limit is 11 mg for ²³⁵U and 7 mg for ²³⁹Pu. The high background rate makes this technique sensitive to the matrix variations that were noted for the similar method employing the ²⁴¹Am-Li source interrogation (see Sec. IV.B.3). The optimum moderator thickness for waste measurement may be considerably greater than that used in fuel rod scanners. However, an optimization study is beyond the scope of this report.

H. Californium-252 Interrogation and Delayed Neutron Detection with Scintillators

1. Measurement Principle. This particular instrument type employs a ²⁵²Cf source for interrogation, and delayed neutrons are detected with scintillation counters. As described, no instrument of that exact type has been built. However, NDA instruments have been built using ²⁵²Cf interrogation and delayed gamma-ray counting with NaI scintillation detectors. Because the plastic scintillation counters used for neutron detection also detect gamma rays the two methods are similar.

Delayed gamma rays are far more numerous than delayed neutrons. In the time interval (0.2 to 45 s) following the induced fission about 200 times more delayed gamma rays are available than neutrons from ²³⁵U. For ²³⁹Pu the factor is even larger, about 465. Thus, the scintillation counter used for neutron detection will primarily count gamma rays if no shielding is employed. The average delayed gamma-ray energy is about 1 MeV and the maximum...
energy is 6.5 MeV. Therefore, a considerable thickness of lead shielding (10-15 cm) would be required to make the neutrons the dominant signal.

Although delayed gamma rays yield a strong signal from the induced fissions, they are not an unique signal. Neutron captures in many isotopes result in beta unstable nuclei that emit energetic gamma rays that could be misinterpreted as delayed gamma rays from fission.\textsuperscript{115} For example neutron capture in fluorine ($^{19}$F) results in gamma-ray emissions of 0.6 and 1.6 MeV with a half-life of 11.4 s.\textsuperscript{116} Other elements emitting delayed gamma rays after thermal neutron capture include sodium, aluminum, chlorine, calcium, silicon, iron, nickel, and copper. For a particular waste matrix category, the delayed gamma-ray background would have to be determined by calculational methods or by test measurements of representative matrix materials free from fissionable materials.

2. Instrument Examples. Figure 28 shows the Multienergy Californium Assay System (MECAS).\textsuperscript{117,118} This instrument has the capability of both subthreshold neutron irradiation (below the fission threshold of fertile materials) and superthreshold irradiation. By tailoring the neutron spectrum during the assay, both fissile $^{235}$U and fertile $^{238}$U content of the sample can be measured, and hence the enrichment determined.

The basic assay method consists of neutron irradiation of the sample and measurement of delayed gamma rays from induced fissions in the sample material. The gamma rays are detected by a 127-mm-diam by 127-mm-long NaI detector with a 25-mm through-hole in which the sample is counted. The sample is transferred from the irradiation region at the center of the unit into the detector for counting. The neutron spectrum is tailored by rotating the $^{252}$Cf source and the tungsten-beryllium core within the nickel reflector with respect to the sample position. When the $^{252}$Cf source is nearest the sample position, superthreshold neutrons interrogate the sample. When the source is rotated away from the sample the interrogating neutrons are primarily subthreshold.

All operations and calculations are performed by an integral minicomputer linked to the mechanical structure by custom interfaces that permit hands-off operation of all assay sequences. Complete operator control of the system for special research applications can also be selected.\textsuperscript{119}

Fuel rod scanners have been modified to provide a pellet-to-pellet uniformity scan.\textsuperscript{120,121} A collimated NaI detector placed at the exit of the assay
channel measures the delayed gamma rays. Figure 29 shows a thermal neutron 252Cf fuel rod assay system with modifications for pellet-to-pellet scan. The delayed gamma-ray counting rate is 90 000/s for a 3.3% enriched fuel rod scanned at a rate of 4 cm/s. The resulting statistical precision indicates that a pellet that differs by 7.5% from its neighbors can be detected with a 97% confidence limit.  

3. Expected Performance. The estimation of the detectability limit will follow the method used in Sec. IV.F.3 for the 252Cf interrogation and delayed neutron counting with 3He-filled detectors. Assuming the density of the matrix material is 0.5 g/cm$^3$ then about 25% of the delayed gamma rays escape the barrel yielding a delayed gamma-ray plus neutron counting rates of $3.5 \times 10^4$/s/g for $^{235}$U and $5 \times 10^4$/s/g for $^{239}$Pu. The background rate is more difficult to estimate because it depends on the area of the scintillators, the number and type of phototubes, and the method used with the
electronic logic for defining an event in the detector. In addition a background occurs for neutron captures in materials other than the fissionable isotopes of interest. The background rate for a small system, four phototubes and a scintillator surface area of 2 sq ft (9.3 \times 10^{-2} \text{ m}^2), is approximately 1000 counts/s.\(^{124}\) Extrapolating by surface area only the background rate for a barrel-size system is 3.4 \times 10^4 \text{ counts/s}. Employing Eq. (A-10) the detectability is estimated at 1.4 mg for \(^{235}\text{U}\) and 1.0 mg for \(^{239}\text{Pu}\).

The technique of \(^{252}\text{Cf}\) interrogation and delayed neutron and gamma-ray counting appears to have the potential for a low detectability limit. However, there is a need for investigation of the possible difficulties with this technique. Already mentioned are the delayed gamma rays from matrix materials. The recovery time of the phototubes and scintillators from the neutron irradiation should be determined. The Random Drivers, which also employ scintillation detectors, have a temperature of drift in the count rate of about 0.7\%/\text{C}.\(^{125}\) For temperature drifts between the background and active portion of the assay, bias in the assay corresponds to 7 mg/\text{C} for \(^{235}\text{U}\) and 5 mg/\text{C} for \(^{239}\text{Pu}\) if no temperature corrections are made. Attenuation of the delayed gamma rays by the matrix material can be corrected for uniformly mixed samples by weight or perhaps by transmission measurement made with the \(^{252}\text{Cf}\) source. If the matrix is nonuniform, then lumping will cause serious problems. Gamma-ray shielding designed to attenuate the less penetrating gamma rays will lessen the matrix sensitivity at the expense of the detection limit. In addition this technique is extremely sensitive to background gamma-ray radiation. If the background assumed for estimating the detectability limit were to come solely from radiation emitted by the waste barrel (500 keV gamma rays), then the corresponding gamma-ray dose from the barrel would be \(3 \times 10^{-4} \text{ mR/h},\) an extremely low activity level. If the dose level were 1 mR/h, then the background rate would be \(1.25 \times 10^8 \text{ counts/s}.\) Assuming the scintillators could still count with that high a rate, then the detectability limit would be about 60 times higher than previously estimated. In view of the sensitivity of this technique to even modest radiation levels and troublesome induced signals from common isotopes, the NDA method of \(^{252}\text{Cf}\) neutron interrogation with delayed gamma-ray and neutron detection with scintillation counters is not recommended for general waste measurements.
Gamma-ray and x-ray counting instruments have been developed and tested for a variety of waste measurement applications. In general measurements are made employing the photon emission with the highest energy that has sufficient intensity to make the measurement possible in a reasonable counting time. Because the higher energy emissions have a greater penetrability, the use of the higher energy photons requires smaller attenuation and shielding corrections or alternatively a larger or more dense item can be measured.

Table XIII lists the major gamma-ray activity for the uranium and plutonium isotopes and $^{241}$Am. In the case of $^{235}$U the photon choice for assay measurements is straightforward, the 185-keV gamma ray. However, in the case of $^{239}$Pu assay different portions of the spectrum are used because no single region of the spectrum is adequate for making both low-level and high accuracy measurements. The 414.7-keV gamma ray is used for assaying quantities of $^{239}$Pu exceeding several grams. The gamma-ray emissions in the 50-400 keV region are used for isotopic and total plutonium determination of small samples, usually solutions, closely coupled to a high-resolution detector. To obtain detectability limits below 10 nCi/g the L x rays have been used for low-density materials.

The applicability of passive gamma-ray and x-ray counting to various waste categories will be discussed using the existing instruments to show the advantages and limitations of each technique.

A. Segmented Gamma-ray Scanner (SGS)

The SGS refers to an instrument as well as a measurement concept. During an assay the item is rotated and translated past the detector. The passive gamma-ray counting rate and gamma-ray transmission through the item are recorded for vertical segments (pancake shaped). The thickness of the segment is adjusted by varying the opening of the collimator. Typically about 10 to 20 segments are used. A computer is used to control the rotation, translation, and actuation of the shutter on the transmission source. The computer also performs the data reduction. An assay requires about 1000 seconds and for well-characterized, homogeneous samples such as incinerator ash, assay accuracies better than 5% can be achieved on a routine basis.
<table>
<thead>
<tr>
<th>Isotope</th>
<th>Half-life (years)</th>
<th>Activity Level (Ci/g)</th>
<th>Principal Gamma rays (keV)</th>
<th>Emission Rate&lt;sup&gt;a&lt;/sup&gt; (gamma rays/s/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td>235&lt;sup&gt;U&lt;/sup&gt;</td>
<td>$7.04 \times 10^8$</td>
<td></td>
<td>185.7</td>
<td>$4.5 \times 10^4$</td>
</tr>
<tr>
<td>238&lt;sup&gt;U&lt;/sup&gt;</td>
<td>$4.47 \times 10^9$</td>
<td></td>
<td>766.4</td>
<td>39</td>
</tr>
<tr>
<td>238&lt;sup&gt;Pu&lt;/sup&gt;</td>
<td>87.79</td>
<td>17.1</td>
<td>766.4</td>
<td>$1.5 \times 10^5$, 30, $^b$ 225&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td>1000.1</td>
<td></td>
<td>1000.1</td>
<td>$8.2 \times 10^3$, 1.6, $^b$ 12&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>239&lt;sup&gt;Pu&lt;/sup&gt;</td>
<td>24082</td>
<td>0.0621</td>
<td>129.3</td>
<td>$1.4 \times 10^5$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>375.0</td>
<td>$3.6 \times 10^4$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>413.7</td>
<td>$3.5 \times 10^4$</td>
</tr>
<tr>
<td>240&lt;sup&gt;Pu&lt;/sup&gt;</td>
<td>6537</td>
<td>0.228</td>
<td>160.4</td>
<td>$3.5 \times 10^4$, 2200, $^b$ 5300&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>642.3</td>
<td>$1.2 \times 10^3$, 76, $^b$ 180&lt;sup&gt;c&lt;/sup&gt;</td>
</tr>
<tr>
<td>241&lt;sup&gt;Pu&lt;/sup&gt;</td>
<td>14.35</td>
<td>103.4</td>
<td>148.6</td>
<td>$7.3 \times 10^6$, 2 $\times 10^4$, $^b$ 2 $\times 10^5$, $^c$</td>
</tr>
<tr>
<td>242&lt;sup&gt;Pu&lt;/sup&gt;</td>
<td>$3.79 \times 10^5$</td>
<td>0.0039</td>
<td>none</td>
<td></td>
</tr>
<tr>
<td>241&lt;sup&gt;Am&lt;/sup&gt;</td>
<td>434.1</td>
<td>3.42</td>
<td>368.6</td>
<td>$2.6 \times 10^5$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>662.4</td>
<td>$4.4 \times 10^5$</td>
</tr>
</tbody>
</table>

<sup>a</sup>For pure isotope unless otherwise noted

<sup>b</sup>Based on 1 g weapons grade plutonium

<sup>c</sup>Based on 1 g reactor grade plutonium
Fig. 30. SGS for barrel-size samples (LASL Neg. #77-17401).

shows an SGS capable of assaying items up to a 55-gal barrel size. The detectability limit for $^{235}$U with the barrel-size unit is 3 g $^{235}$U.108

B. Multiple NaI Barrel Counter

Figure 31 shows an early version of the segmented gamma-ray scanning technique. Here a NaI detector and a transmission source are employed for each segment.127,128 This deployment of detectors eliminates the need for translating the barrel during the scanning, and a lower detectability limit is achieved because of the increased number of detectors.

The opening angles of the collimators were adjusted following an initial transmission measurement to improve the radial response uniformity. For matrices with densities up to 0.8 g/cm$^3$ a response uniformity of ±10% over the entire volume of the barrel could be obtained with two collimator positions.127 In the unit shown in Fig. 31 the collimator adjustment was made manually. This cumbersome procedure could be eliminated by having the collimators positioned with computer-controlled actuators.
A set of barrels containing various quantities of weapons grade plutonium in different "typical" waste matrices was prepared by the Rocky Flats Plutonium Facility for test measurements. The quantity of $^{239}$Pu ranged from 10 to 195 grams. The matrix materials consisted of graphite, raschig rings, washables, combustibles, wet combustibles, resin, and plexiglas. For the 17 test barrels, the standard deviation of the difference between the measured and known $^{239}$Pu was 11.7% and the bias for the total quantity was -7.2%.

In another campaign two hundred 30-gal drums containing $^{238}$Pu residues were assayed. The spacing between the 5- by 5-cm NaI detectors was reduced from 15 to 10 cm to match more closely the geometry of the 30-gal drums. The calibration was obtained using 13 drums for which the $^{238}$Pu content ranged from 0.3 to 45.3 g. The standard deviation was 12% based on measurements of the 766-keV gamma ray. The detectability limit for either $^{238}$Pu or $^{239}$Pu was one gram or better for the two barrel sizes.

C. Multi-Energy Gamma Assay System (MEGAS)

The assay principle used in the Multi-Energy Gamma Assay System (MEGAS) combines gamma-ray and L x-ray detection to measure low-density combustible room trash packaged in 57-liter cardboard cartons. The L x-ray region (~17 keV) is monitored for the sub-10-nCi/g measurements and the higher energy gamma rays are used as the contamination level increases. The NaI detector has a 0.25-mm-thick beryllium entrance window to permit the transmittance of the x rays and the NaI crystal is 51 mm thick to absorb the 400-keV gamma rays from plutonium. The NaI crystal has a 127-mm diameter; however, the viewing area is collimated to a 25-mm width by 102-mm height to improve the response uniformity. Figure 32 shows the MEGAS unit used at the Los Alamos Scientific Laboratory (LASL) plutonium facility (TA-21). Similar units are in use at the LASL waste incineration facility, at the new plutonium facility (TA-55), and at the Nuclear Materials and Equipment Company (NUMEC), Leechburg, PA.

The MEGAS box counter is being upgraded to include a large-area intrinsic germanium detector. The additional high-resolution detector increases the versatility of the system by permitting the separation of interfering fission product gamma-ray and x-ray emissions from those of the transuranics. The germanium detector also aids in improving the accuracy by facilitating the implementation of sophisticated analysis techniques. These techniques include transmission measurements that correct for the rapidly varying absorption cross-
secton in the L x region, and for
the gamma rays the increased back-
ground caused by Compton scattering
within the waste container provides
a technique for estimating the attenu-
ation within waste material.\textsuperscript{131}

Table XIV summarizes the detecta-
bility limits quoted for the NaI and
germanium detectors for several
energy regions and isotopic species.
The NaI detector has a better detecta-
bility limit for well-isolated
spectral regions because of its
larger active area. However, for
regions containing emissions from
two or more uncorrelated isotopes,
the germanium detector is the
preferred choice. The important
data to emphasize from Table XIV are
that the MEGAS box counter and the
upgraded version can achieve a detectability limit below the 10-nCi/g fiducial
for a variety of low-density wastes.

The below-10-nCi/g performance of the MEGAS technique cannot be achieved
for 55-gal barrels, because the L x rays are absorbed by the container's steel
walls. Assuming the absorption problem in the container wall can be eliminated
by packaging the waste in something like plastic or paper "trash bags," then
the density restriction for the waste makes the use of large containers
impractical. The upper limit for the MEGAS waste density is about 0.14 g/cm\(^3\)
before absorption of the L x rays in the 30-cm-thick containers becomes a
serious problem.\textsuperscript{131} In employing larger containers the maximum density would
have to be reduced proportionally to the increased thickness. For example,
55-gal barrel-size containers would be restricted to a combustible waste
density less than about 0.07 g/cm\(^3\) (14 kg maximum). This rather low density
limit implies strict administrative control to assure no compaction whatsoever
and "fluffing" of the waste before assay. Thus, no handling savings can be expected for larger size containers.

**TABLE XIV**

MEGAS DETECTABILITY LIMITS

<table>
<thead>
<tr>
<th>Photon Energy Region (keV)</th>
<th>Source</th>
<th>Detectability Limit (nCi/g)(^a)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>NaI Detector without Fission Products</td>
</tr>
<tr>
<td>13.6, 17.2, 20.2 (U L x rays)</td>
<td>Pu(^b)</td>
<td>0.0260</td>
</tr>
<tr>
<td>32.1 (Ba K x ray)</td>
<td>(^{137})Cs</td>
<td>0.01</td>
</tr>
<tr>
<td>51.6</td>
<td>Pu(^b)</td>
<td>2</td>
</tr>
<tr>
<td>59.5</td>
<td>(^{241})Am</td>
<td>0.002</td>
</tr>
<tr>
<td>98.4 (U K x ray)</td>
<td>Pu(^b)</td>
<td>7</td>
</tr>
<tr>
<td>129.3</td>
<td>Pu(^b)</td>
<td>11</td>
</tr>
<tr>
<td>400</td>
<td>Pu(^b)</td>
<td>23</td>
</tr>
<tr>
<td>662</td>
<td>(^{137})Cs</td>
<td>0.004</td>
</tr>
<tr>
<td>1332</td>
<td>(^{60})Co</td>
<td>0.004</td>
</tr>
</tbody>
</table>

\(^a\)Detection limits are at the 3\(\sigma\) level above background, 1000 s\(^2\)counting time, 57 liter carton, 6200 g of waste, average density = 0.11 g/cm\(^3\).

\(^b\)Plutonium with 94\% \(^{239}\)Pu and 6\% \(^{240}\)Pu.

\(^c\)Fission products simulated by 10 \(\mu\)Ci \(^{137}\)Cs point source.

\(^d\)Fission products simulated by 4 \(\mu\)Ci \(^{60}\)Co point source.
D. Pancake Counter

The pancake counter is an uncollimated NaI detector positioned at the top of a glove box to view items placed beneath it. The detector has a 127-mm-diam by 1.6-mm-thick NaI crystal. The thin detector makes the instrument well suited for detecting low-energy x rays and gamma rays, although it is relatively insensitive to high-energy gamma rays. Two spectral regions are accumulated, the L x rays at \( \approx 17 \) keV from transuranic isotopes and the 60-keV region arising from the \(^{241}\text{Am}\) gamma ray. The americium contribution is subtracted from the L x-ray region based on the count rate at 60 keV. Thus, for waste that is known to be contaminated with only plutonium and americium the two components can be estimated separately.

The potential for large spacial variations in the response limits the accuracy to about \( \pm 50\% \). However, the instrument is used in a sorting mode and the waste items tend to be either contaminated or not contaminated with very few intermediate cases found in actual practice. The assay is accomplished with a 10-s count with the control switches placed within the glove box. This procedure permits a trash box to be sorted rather quickly to locate contaminated items. The 10-s count has a detectability limit of 6 \( \mu \text{g} \) \(^{239}\text{Pu}\) and 8.5 ng \(^{241}\text{Am}\). The 2-liter packages typically have a mass of 400 g or less, thus the corresponding contamination levels are 1.1 nCi/g for plutonium and 0.07 nCi/g for americium.

E. Elephant Gun

The elephant gun is a shielded and collimated NaI detector developed for the assay of large containers of combustible waste from low-enriched uranium operations. The 185.7-keV gamma ray from \(^{235}\text{U}\) can lead to unreliable assay results because of the attenuation and can be severe and perhaps impossible to evaluate.

The elephant gun employs detection of the 767- and 1001-keV gamma rays from the decay of \(^{234}\text{m} \text{Pa}\), a daughter of \(^{238}\text{U}\). The decay chain that populates \(^{234}\text{m} \text{Pa}\) is

\[
^{238}\text{U}(4.5 \times 10^9 \text{ yr}) \xrightarrow{\alpha} ^{234}\text{Th}(24.1 \text{ day})
\]

\[
\xrightarrow{\beta^{-}} ^{234}\text{m} \text{Pa}(1.2 \text{ min}) \xrightarrow{\beta^{-}} ^{234}\text{U}(2.5 \times 10^5 \text{ yr})
\]
The half-life of $^{234}$Th (24.1 day) dominates the time required to reach equilibrium. Uranium that has not undergone chemical separation from its daughters for about 3 months is effectively in equilibrium with the $^{234m}$Pa daughter. The equilibrium emission rates for the gamma rays at 767 and 1001 keV are 60 and 100/s/g, respectively. Because of this low specific activity, a large NaI detector is required to obtain a sufficient count rate.

Figure 33 shows the details of the elephant gun detector. The NaI crystal is 127 mm by 127 mm. The detector is operated with a standard commercially available portable electronic unit that includes the high-voltage power supply, amplifier, single-channel analyzer, scaler, and timer. The window on the single-channel analyzer is set wide enough to accept pulses from gamma rays at 767 and 1001 keV.

The $^{238}$U assay with the elephant gun was developed for measurements of 1.07- by 1.22- by 1.22-m (3-1/2- by 4- by 4-ft) plywood boxes used by the General Electric Company, Wilmington, North Carolina, for temporary storage of combustible waste. These containers typically hold 300 kg of rags (density = 0.2 g/cm$^3$). The detector views the box through the smallest dimension at a distance of 2 m from the surface, a second measurement is made viewing the opposite face of the box, and the two results are averaged. Figure 34 shows a waste box, the elephant gun, and electronic unit. In an actual

Fig. 33. Elephant gun detector schematic drawing (EG&G Neg. #10259).

Fig. 34. Elephant gun, electronic unit, and waste box (LASL Neg. #72-12442).
assay the separation between the elephant gun and the box would be greater than that indicated by Fig. 34.

Because the mass absorption coefficients are essentially constant over the range of matrix materials present in the waste, the attenuation correction factor for the gamma rays can be estimated from the weight of the box. Typically the correction factor is modest, about 1.5. In case of nonhomogeneous arrangements of material in the box the assay tends to underestimate the $^{238}\text{U}$ content. For example, if the uranium is located in a thin layer on the bottom of the box the assay is 10% low, if the uranium is surrounded by a 3.2-mm-thick (one-eighth inch) iron plate the assay is 14% low, and if the uranium is in the form of light-water reactor fuel pellets the assay is 55% low because of self-shielding within the pellets. For a 1000-s assay the detectability limit is about 16 g $^{238}\text{U}$. If the uranium has an enrichment of 3%, the quantity of $^{235}\text{U}$ present with the $^{238}\text{U}$ is 0.5 g.

VI. ASSAY TECHNIQUES SUMMARY

Assay techniques based on passive neutron, passive gamma-ray and x-ray, and active interrogation with isotopic sources have been reviewed for their applicability to waste measurements of 55-gal barrels. The emphasis has been placed on the detectability limit and potential difficulties associated with each measurement technique. Table XV lists the techniques and the detectability limits for $^{235}\text{U}$ and weapons and reactor grade plutonium oxide (see Table VI for the assumed plutonium isotopic composition).

From the data displayed in Table XV five techniques appear to be capable of sorting waste at the 10-nCi/g fiducial. These techniques include the two photoneutron interrogation methods with prompt energetic fission neutron detection and three techniques employing $^{252}\text{Cf}$ interrogation. The $^{252}\text{Cf}$-based techniques are delayed neutron counting with $^3\text{He}$ detectors, energetic prompt neutron detection with gas-filled detectors after the source neutrons are heavily moderated, and delayed gamma-ray and neutron counting with scintillation detectors. The passive neutron counting instrument employing $^3\text{He}$ detectors is close to being able to sort waste at the 10-nCi/g fiducial. The three techniques employing an alpha-n ($^{241}\text{AmLi}$) source have detectability limits too high to make the instruments viable for sorting wastes at the 10-nCi/g fiducial. The passive gamma-ray techniques have detectability limits of about 68
### Table XV
DETECTABILITY LIMITS SUMMARY

<table>
<thead>
<tr>
<th>Detector Type</th>
<th>Generic Name (Emission detected)</th>
<th>Interrogation Source</th>
<th>Isotopes Detected</th>
<th>Detectability Limit&lt;sup&gt;a&lt;/sup&gt; Weapon Grade Pu&lt;sup&gt;b&lt;/sup&gt;</th>
<th>Reactor Grade Pu&lt;sup&gt;c&lt;/sup&gt;</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Passive well Counter</td>
<td>$^3$He, $^{10}$Be(n)</td>
<td>none</td>
<td>even Pu &amp; Cm</td>
<td>---</td>
<td>28 mg, 21 nCi/g</td>
<td>6.9 mg, 7.6 nCi/g</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>69 mg, 52 nCi/g</td>
<td>26 mg, 29 nCi/g</td>
</tr>
<tr>
<td>Passive Random Driver</td>
<td>Scint. (n,γ)</td>
<td>none</td>
<td>even Pu &amp; Cm</td>
<td>---</td>
<td>0.43 g, 325 nCi/g</td>
<td>0.17 g, 180 nCi/g</td>
</tr>
<tr>
<td>AWCC</td>
<td>$^3$He (coin. n)</td>
<td>$^{238}$U</td>
<td>fissile</td>
<td>23 000</td>
<td>use passive mode</td>
<td>use passive mode</td>
</tr>
<tr>
<td></td>
<td>$^3$He (fast n)</td>
<td>$^{235}$U</td>
<td>fissile</td>
<td>1400</td>
<td>1.0 g, 756 nCi/g</td>
<td>1.1 g, 1200 nCi/g</td>
</tr>
<tr>
<td>Random Driver</td>
<td>Scint. (n,γ)</td>
<td>$^{238}$U</td>
<td>fissile</td>
<td>70</td>
<td>122 mg, 92 nCi/g</td>
<td>144 mg, 160 nCi/g</td>
</tr>
<tr>
<td></td>
<td>$^3$He (n)</td>
<td>$^{235}$U</td>
<td>fissile</td>
<td>3.8</td>
<td>2.7 mg, 5.0 nCi/g</td>
<td>3.1 mg, 3.4 nCi/g</td>
</tr>
<tr>
<td></td>
<td>$^3$He (fast n)</td>
<td>$^{235}$U</td>
<td>fissile</td>
<td>0</td>
<td>5 mg, 4 nCi/g</td>
<td>6 mg, 7 nCi/g</td>
</tr>
<tr>
<td>$^{252}$ Cf Shuffler</td>
<td>$^3$He (delayed n)</td>
<td>$^{252}$Cf</td>
<td>fissile even Pu &amp; Cm</td>
<td>2.0</td>
<td>3.6 mg, 2.7 nCi/g</td>
<td>4.2 mg, 4.6 nCi/g</td>
</tr>
<tr>
<td></td>
<td>$^3$He (fast n)</td>
<td>$^{252}$Cf</td>
<td>fissile</td>
<td>11</td>
<td>7 mg, 5 nCi/g</td>
<td>9 mg, 10 nCi/g</td>
</tr>
<tr>
<td></td>
<td>Scint. (delayed n,γ)</td>
<td>$^{252}$Cf</td>
<td>fissile even Pu &amp; Cm</td>
<td>1.4</td>
<td>1.0 mg, 0.8 nCi/g</td>
<td>1.3 mg, 1.4 nCi/g</td>
</tr>
<tr>
<td>SOI</td>
<td>Ge, NaI (r)</td>
<td>none</td>
<td>$^{235}$U, $^{238}$U, $^{238}$Pu, $^{239}$Pu, $^{241}$Am</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

---

<sup>a</sup>1000 s assay, 3σ above background and up to 1000 R/m fission products

<sup>b</sup>Conversion factor: for 100 kg of waste 1 mg weapons grade Pu corresponds to 0.176 nCi/g

<sup>c</sup>Conversion factor: for 100 kg of waste 1 mg reactor grade Pu corresponds to 1.106 nCi/g

<sup>d</sup>No fission products, with fission products a "rule-of-thumb" maximum fission product concentration is 1 mCi per gram of U, Pu, or Am one gram for $^{235}$U, $^{238}$U, $^{238}$Pu, $^{239}$Pu, and $^{241}$Am. Thus the passive gamma-counting methods are not practical for 10-nCi/g measurements in 55-gal barrels. However, the gamma-ray measurements are the most reliable methods for isotopic characterization at the 1-g and higher contamination levels.

Of the five methods potentially capable of sorting at the 10-nCi/g fiducial, the technique employing the $^{252}$Cf source with delayed gamma-ray and neutron counting using scintillators should be eliminated from consideration. The technique is quite sensitive to gamma-ray radiation in that the materials to be sorted should have surface dose rates below 1 mR/h. In addition most matrix materials contain common elements such as silicon, sodium, chlorine, aluminum, iron, or fluorine that emit delayed gamma rays following neutron irradiation. Thus, the signature used in detecting uranium and plutonium

---

69
is not unique to fissionable materials and many "false alarms" would occur with the assay results.

The best technique is the $^{252}$Cf interrogation and delayed neutron detection with $^3$He-filled proportional gas detectors. The instruments employing this technique are generically referred to as $^{252}$Cf Shufflers. Although the delayed neutrons are a much smaller signal than the prompt neutrons, the delayed neutrons do not have to override the source during interrogation, and the $^{252}$Cf Shuffler's detection of delayed neutrons is positive proof of the presence of fissionable material. The assay chamber of the $^{252}$Cf Shuffler is optimized for efficient neutron detection so that the unit functions as a passive well counter during the background counting portion of the assay. Passive coincidence neutron counting with the $^{252}$Cf Shuffler is a valuable technique for the assay of quantities of plutonium greater than about 200 mg. The prompt neutron counting methods viable at the 10-nCi/g fiducial are optimized to count only the energetic neutrons from the induced fissions; however, the fast neutron counting schemes are inherently inefficient making passive coincidence counting impractical.

The delayed neutron counting approach is less sensitive to matrix moderating effects in the signal neutrons because the detector banks can be optimized for a nearly flat energy response. In the case of the prompt neutron detection schemes, once the neutron is moderated below the detection threshold (typically 0.5 MeV), it is no longer available for counting.

The three prompt neutron counting instruments capable of 10-nCi/g measurements all have the drawback that differences between the matrix material of the standards and items being assayed can induce either a false high or low assay result by changing the number of detected source neutrons. The instruments can improve their response characteristics by reducing the number of source neutrons counted with the signal. Adding more moderator to the source tailoring assembly will help; however, for the $^{252}$Cf-based system the initial high energy of the source neutrons makes improvements more difficult than with the lower energy $^{124}$Sb-Be neutrons. Of the two $^{124}$Sb-Be-based systems, discriminating against the source neutrons with the detectors favors the $^3$He detector unit employing the low-energy neutron absorber between the detector and the item being assayed. Here the thickness or concentration of boric acid (or other boron-loaded neutron absorber) can be increased to attenuate the source neutrons relative to the fission neutrons. In addition, neutron
detectors can be placed at several depths within the absorbing medium to monitor the source neutron attenuation. However, the unit has the most complicated and expensive assay chamber of any of the systems described in this report. The matrix problems present in the other two prompt neutron counting units are sufficiently severe as to cast doubts on their ability to sort waste routinely at the 10-nCi/g fiducial.

VII. RECOMMENDATIONS

The conservative practice of treating all wastes from plutonium processing areas as potentially contaminated has resulted in retrievable storage (burial) of large quantities of materials that for the most part are uncontaminated. The exhumation of the burial sites increases the quantity of material classified as contaminated waste because previously uncontaminated earth at the burial site is then considered part of the waste. Separating uncontaminated items at the generation point is of prime importance for reducing the quantity of waste to those items that are actually contaminated.

The MEGAS box counter employed at LASL has been successful in sorting low-density combustible waste at the 10-nCi/g fiducial. Of the items sorted, only 10% are found to be contaminated and thus a 90% reduction in the quantity of waste is realized. The yearly savings including the initial cost of the instrument and operational expenses is over $200,000 per year. Similar or perhaps greater cost saving would accrue with the sorting of the high-density waste packaged in 55-gal barrels.

The review of instruments in this report has determined that the $^{252}\text{Cf}$ Shuffler assay technique is easily capable of sorting plutonium-contaminated waste at the 10-nCi/g fiducial as well as being able to detect $^{235}\text{U}$ quantities as small as 2 mg. The $^{252}\text{Cf}$ Shuffler is also the best choice over other instruments considered here with detectability limits estimated to be below 10 nCi/g.

To demonstrate the practicality of sorting high-density waste a test and evaluation program would be required. To test limits of the technique thoroughly, a host facility with a variety and quantity of waste would have to be identified. The unit, after being designed for the categories of waste handled by the host facility, would be delivered for field tests. Locating the instrument near the waste generation site may require installing the unit
in a temporary building or trailer if floor space is unavailable. If the fission product activity is high (somewhere between 10 and 100 mR/h), remote and preferably automatic processing of the barrels may be desired. Automatic handling of the barrels will reduce the manpower operations cost while increasing the throughput.

The $^{252}$Cf Shuffler could serve as one of the components in a waste measurement station. In addition to the $^{252}$Cf Shuffler a screening station and gamma-ray assay station would be included. The screening station would monitor the external gamma-ray and neutron dose rates, check for criticality safety, and check for exterior alpha contamination. The gamma-ray assay station would identify and quantify the fission products present as well as provide assay measurements for thorium, uranium, plutonium, and americium when sufficient quantities of those heavy elements are present. The $^{252}$Cf Shuffler provides the go/no go measurement at 10 nCi/g as well as an assay of the fissile and spontaneously fissioning isotopes.

Upon completing the test and evaluation, additional units with suggested modifications could be obtained from commercial vendors without additional development costs.

ACKNOWLEDGMENTS

I would like to thank the LASL staff members who have contributed their knowledge and expertise to supplement the sometimes sparse published data for the instruments and techniques described in this report. In particular, Mike Baker contributed detailed information on the photoneutron interrogation techniques. George Eccleston provided suggestions for calculational techniques to estimate neutronic characteristics. Norbert Ensslin, my office mate, has been tapped for information on well counters, coincidence circuits, and Random Driver instruments. Jack Parker has discussed the gamma-ray techniques on several occasions. Darryl Smith's notes have provided unpublished background data. Howard Menlove commented on the evaluation of several techniques, particularly the AWCC. Douglas Reilly aided in obtaining information on the barrel-size passive well counter and gamma-ray assay techniques. The support and guidance for this report has come from Roddy Walton, Q-1 Group Leader, and John Umbarger. And I would like to thank especially Joy Clark and Jeanne Hassenzahl for their typing and editing of this document.
APPENDIX
DETECTION LIMIT CALCULATION

The detection limit is defined as that quantity of material that yields a signal three standard deviations above background. This appendix will develop formulas for estimating the detection limit for a wide class of nondestructive assay (NDA) instruments. No attempt will be made to include systematic or correlated errors that might influence the error estimations for a particular instrument. A list of references is given for detailed discussions of error estimation and statistical analysis theory.136-140

When NDA instruments measure an item by nuclear emissions usually some background counts are also accepted along with the desired signal. To obtain an estimation of the background counts accepted with the signal counts a separate measurement of the background is made. The estimated background counts are subtracted from the signal plus background to obtain the actual signal. The estimated background can with some techniques be obtained concurrently with the signal plus background, whereas other techniques require additional measurements. The error for the actual signal includes uncertainty contributions from the signal plus background measurement and the separate background measurement. The estimated number of signal counts is given by

\[ S = (S + B_1) - B_2 \]  \hspace{1cm} (A-1)

and the uncertainty in \( S \) is given by

\[ \Delta S = \left[ \Delta(S + B_1)^2 + (B_2)^2 \right]^{1/2} \]  \hspace{1cm} (A-2)

where

\[ (S + B_1) \] = signal plus background counts,
\[ B_2 \] = estimated background counts,
\( \Delta(S + B_1) \) = uncertainty in signal plus background counts, and
\( \Delta B_2 \) = uncertainty in the estimated background counts.

The number of standard deviations the signal is above the background is given by

\[
D = \frac{S}{\Delta S}. \tag{A-3}
\]

The detectability limit is defined with \( D = 3 \). For a generalized case the counts and their uncertainties are given by

\[
(S + B_1) = m r_S T_1 + m r_b T_1 + b T_1, \tag{A-4}
\]

\[
\Delta(S + B_1) = (S + B_1)^{1/2}, \tag{A-5}
\]

\[
B_2 = (m r_b T_2 + b T_2) \frac{T_1}{T_2}, \tag{A-6}
\]

\[
\Delta B_2 = \left( B_2 \frac{T_1}{T_2} \right)^{1/2}, \tag{A-7}
\]

where

- \( m \) = the quantity of material being assayed,
- \( r_s \) = the signal rate (counts/s/g),
- \( r_b \) = the background from the material (counts/s/g),
- \( b \) = the background not from the material (counts/s),
- \( T_1 \) = time spent counting signal plus background, and
- \( T_2 \) = time spent counting background.

Thus, the number of standard deviations the signal is above background is given by
\[ D = \frac{m_r S_1}{m_r T_1 + m_r T_1 + b T_1 + \left( m_r T_2 + b T_2 \right) \left( \frac{T_1}{T_2} \right)^2} \]. \quad (A-8)

Solving Eq. (A-8) for the mass of material at the detectability limit yields

\[ m = \frac{D^2 \left[ I_s + I_b \left( 1 + \frac{T_1}{T_2} \right) \right] + D^2 \left[ I_s + I_b \left( 1 + \frac{T_1}{T_2} \right) \right]^2 + 4br_T \left[ 1 + \frac{T_1}{T_2} \right] \left( \frac{T_1}{T_2} \right)^{1/2}}{2r_s T_1} \]. \quad (A-9)

In most applications, the count rate is dominated by the system background \( b >> m_r b, m_r \), and the mass at the detectability limit to a good approximation is given by

\[ m = \frac{D \left[ b \left( 1 + \frac{T_1}{T_2} \right) \right]^{1/2}}{r_s T_1^{1/2}} \]. \quad (A-10)

In the case of coincidence counting measurements, Eq. (A-8) is modified to include the possibility of a coincidence as well as a single event neutron background. The single event background contributes to the coincidence total only through "accidental" coincidences. On the other hand, the coincidence neutron background contributes directly to coincidence total. A coincidence background can arise from cosmic-ray interactions in a lead shield (see Sec. III.A.3). The single neutrons (alpha,n) emitted by the nuclear material also contribute "accidental" coincidence counts. If the single event and coincidence background rates are known from previous measurements, the equation for the detectability limit is
\[ D = \frac{m r_s T}{\left[ m r_s T + (b_s + m r_b)^2 \tau_g T + b_c T \right]^{1/2}}, \]  

\( (A-11) \)

where

- \( r_s \) = coincidence count rate (counts/s/q),
- \( r_b \) = single neutron count rate (counts/s/g),
- \( b_s \) = background singles count rate (counts/s),
- \( b_c \) = background coincidence count rate (counts/s),
- \( \tau_g \) = coincidence gate time, and
- \( T \) = assay time.

Solving Eq. (A-11) for the mass at the detectability limit yields

\[ m = \frac{D^2 (r_s + 2r_b b_s \tau_g) + D \left[ D^2 (r_s + 2r_b b_s \tau_g)^2 + 4 (r_s^2 T - D^2 r_b^2 \tau_g) (b_c + b_s^2 \tau_g) \right]^{1/2}}{2 (r_s^2 T - D^2 r_b^2 \tau_g)}, \]  

\( (A-12) \)
REFERENCES


41. Calculator manufactured by Hewlett-Packard, Model HP-27; the complete system (well counter, coincidence circuit and support electronics, and HP-27 calculator modified for direct data input) is available from IRT Corp. Model HLNCC-100.


49. J. Gray, Allied General Nuclear Services, personal communication.


60. Manufactured by National Nuclear Corp. in standard models or to customer specifications.


133. Manufactured by Eberline, Santa Fe, New Mexico, model SAM II.


