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TITLE: ANALYSIS OF FISSIONABLE MATERIAL USING
DELAYED GAMMA RAYS FROM PHOTOFISSION

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ANALYSIS OF FISSIONABLE MATERIAL USING
DELAYED GAMMA RAYS FROM PHOTOFISSION

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ABSTRACT

The energetic gamma-ray spectra from the fission products of photofission have been investigated to determine whether photofission can identify heavily shielded fissionable material. Target samples of natural thorium, 93% enriched ^{235}U , natural uranium, and 93% enriched ^{239}Pu were irradiated with bremsstrahlung gamma rays produced by 10-MeV electrons from a small linear accelerator. The gamma-ray spectra for each of the four isotopes studied reveals a distinctive intensity distribution. For example, the intensity ratio of the pair of gamma rays at 1436 keV (^{138}Cs) and 1428 keV (^{94}Sr) is 1.9 for ^{235}U , 2.4 for ^{238}U , 1.7 for ^{232}Th and 1.4 for ^{239}Pu .

1. Introduction

A problem in assaying fissionable material arises when the material is well shielded. Conventional passive gamma-ray techniques use low-energy (less than 500 keV) gamma rays to identify ^{235}U and ^{239}Pu and are not applicable. The attenuation of gamma rays of these energies by lead, for example, is such that about 1% penetrate 2.54 cm of lead shielding. Higher energy gamma rays are needed, which can be produced by the fission products resulting from photofission. About 22% of 1500-keV gamma rays would penetrate 2.54 cm of lead.

Kahane and Wolf (1) have recently reported the gamma-ray spectra resulting from photofission of ^{238}U having an energy up to 1131 keV. The Belgium group (2) has made extensive studies of the mass distributions resulting from photofission of uranium and plutonium isotopes. The centroid for the high mass distribution is nearly constant, while that for the low mass distribution changes almost linearly with the mass of the target. This suggests that unique gamma-ray intensity distributions exist for each fissionable isotope. Gamma rays from short-lived fission products were not observed, because counting did not begin until 6 min (see Ref. 1) or 5 sec (see Ref. 2) after photofission. Walton et al. (3) measured the integrated intensity of delayed gamma rays above 510 keV from photofission of ^{235}U , ^{238}U , and ^{232}Th from 100 μsec to 7 sec. Fisher and Engle (4) performed similar measurements for fast neutron fission of ^{232}Th , ^{233}U , ^{235}U , ^{238}U , and ^{239}Pu during the time interval 0.2 to 45 sec after fission. The investigations described in Ref. 3 and 4 show that the rate of gamma-ray emission is more than an order of magnitude larger in

the time period from 10^{-3} sec to 1 sec than for time intervals later than 5 sec after fission.

We report results of measurements of energetic gamma rays obtained with a high-resolution germanium detector as early as 14 msec after photofission. Several gamma rays from fission products are observed with energies greater than 1000 keV. These energetic gamma rays can be used to identify shielded fissionable material.

2. Experimental method

Bremsstrahlung gamma rays produced by 10-MeV electrons from a small transportable linear accelerator irradiated samples of natural thorium (1-kg disk, 15 cm in diameter and 0.32 cm thick), 93% enriched ^{235}U (1-kg disk, 15 cm in diameter and 0.32 cm thick), natural uranium (2-kg disk, 15 cm in diameter and 0.32 cm thick), and 93% enriched ^{239}Pu (2-kg cylinder, 5.1 cm in diameter and 5.1 cm long). A target of ^{208}Pb (1-kg disk, 15 cm in diameter and 0.32 cm thick) was used to identify room background gamma rays, both passive and bremsstrahlung-induced gamma rays in the shielding material.

The arrangement of the experimental apparatus is shown in plan view in Fig. 1. The electron beam had a macro-structure of 10- μ sec duration pulses at a 10-Hz repetition rate. The peak current in each pulse was 80 mA. A thick tungsten electron converter produced the uncollimated photon beam, which irradiated the targets placed approximately 60 cm away. The targets and the linac were placed inside a 20-cm-thick concrete cave. Lead shielding as detailed in Fig. 1 was placed between the tungsten target and the gamma-ray detector. The gamma-ray spectrometer was an 11% efficient coaxial, high-purity

germanium detector equipped with a standard resistive feedback preamplifier.

Even though shielded by 70 cm of lead from the direct bremsstrahlung gamma rays, the germanium detector was paralyzed from the beginning of the beam pulse for 13 msec. The multichannel analyzer was gated off for 13.6 msec by a gate pulse derived from the electron beam burst and remained on for 86.4 msec until the next burst. Each target was irradiated approximately 2 hours, with spectra saved after irradiation periods of 5 minutes, 15 minutes, 30 minutes, and 2 hours.

3. Results

The gamma-ray spectra are very rich and complex. The portion of the gamma-ray spectra from 1000 to 1500 keV, shown in Fig. 2, reveals a distinctive intensity distribution for the four isotopes studied. The stronger gamma-ray lines associated with fission products have been identified using the tabulations of Blachot and Fiche (5) and of Reus and Westmeier (6).

The intensities of the lines in each spectrum depend on many factors. The mass distribution of the fission products and the detailed time history of the measurement are important considerations. The time history is important because of the various half lives of the nuclides in the decay chains. For spectra with similar times histories on the four target materials, Table 1 lists intensity ratios for the stronger lines relative to the intensity for the 1436-keV gamma ray from ^{138}Cs . These ratios can be used to identify the fissionable material as illustrated by the pair of gamma rays at 1436 keV (^{138}Cs) and 1428 keV (^{90}Sr). The intensity ratio of this pair is 1.9 for ^{235}U , 2.4 for ^{238}U , 1.7 for ^{232}Th , and 1.4 for ^{239}Pu .

Each spectrum for each isotope can be considered the response function of that isotope to the bremsstrahlung gamma rays. The isotopic composition of an unknown sample of material can be obtained by performing a linear fit of a mixture of the individual response functions to the gamma-ray response function of the unknown material.

Additional measurements are needed on other fissionable isotopes to obtain their response functions. We also intend to extend these measurements to shorter time periods after fission. Preliminary measurements indicate that a fast recovery preamplifier on a germanium detector, such as a transistorized reset preamplifier, will allow measurements to be made within 100 μ sec after fission.

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REFERENCES

1. S. Kahane and A. Wolf, Phys. Rev. C32 (1985) 1944.
2. D. De Frene et al., Phys. Rev. G28 (1984) 1908, and references contained therein.
3. R. B. Walton et al., Phys. Rev. 134 (1964) 8824.
4. P. C. Fisher and L. B. Engle, Phys. Rev. 134 (1964) 8796.
5. J. Blachot and C. Fiche, At. Data Nucl. Data Tables 20 (1977) 241.
6. U. Reus and W. Westmeier, At. Data Nucl. Data Tables 49 (1983) 13.

Table 1

Intensity ratios of the fission product gamma rays indicated in Fig. 1.

All ratios are compared to the 1436.0-keV gamma ray from ^{138}Cs .

Fission Product	Gamma-Ray Energy (keV)	Intensity Ratios			
		^{239}Pu	^{238}U	^{235}U	^{232}Th
^{138}Cs	1436.0	100.0	100.0	100.0	100.0
^{94}Sr	1428.3	136.4	235.7	186.8	171.3
^{136}I	1313.0	72.7	240.5	123.0	109.3
^{89}Rb	1248.2	13.1	85.4	92.7	228.0
^{90}Kr	1118.7	39.5	110.5	108.5	171.0
^{97}Y	1105.0	113.7	148.1	115.2	68.9
^{89}Rb	1031.9	66.5	116.4	135.9	265.1

FIGURE CAPTIONS

- Fig. 1. Experimental arrangement showing the germanium detector, fissionable target material, lead and concrete shielding, and electron beam.
- Fig. 2. Gamma rays between 1000 and 1500 keV from fission products resulting from photofission of ^{232}Th , ^{235}U , ^{238}U , and ^{239}Pu . Selected peaks are labeled with the energy and associated fission products.

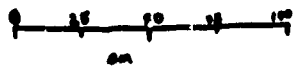
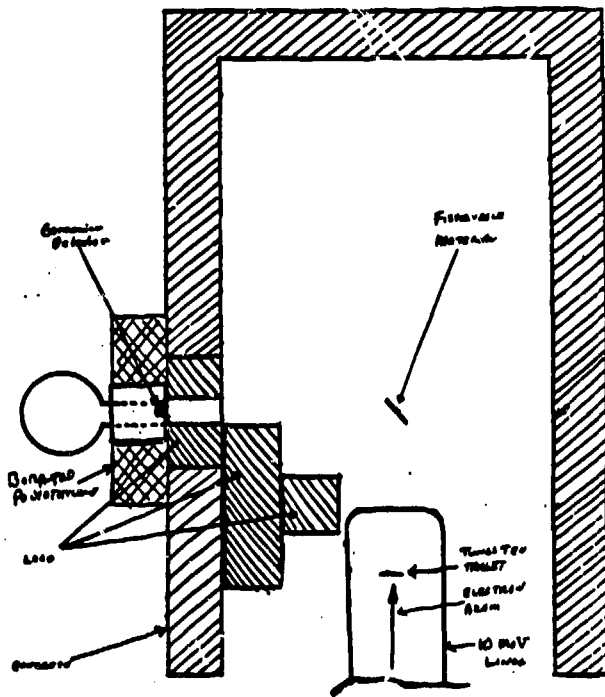


FIG. 3

Approximate final size
3 1/8" width

III. RESULTS

The portion of the gamma-ray spectra from 1000 to 1500 keV for each of the four isotopes (Figure) reveals a distinctive intensity distribution. For example, the intensity ratio of the pair of gamma rays at 1436.0 keV (^{138}Cs) and 1428.3 keV (^{94}Sr) varies with the target. It is 1.9 for ^{235}U , 2.4 for ^{238}U , 1.7 for ^{232}Th , and 1.4 for ^{239}Pu . These and other energetic gamma rays can be used to identify and quantify unknown samples of heavily shielded fissionable material.

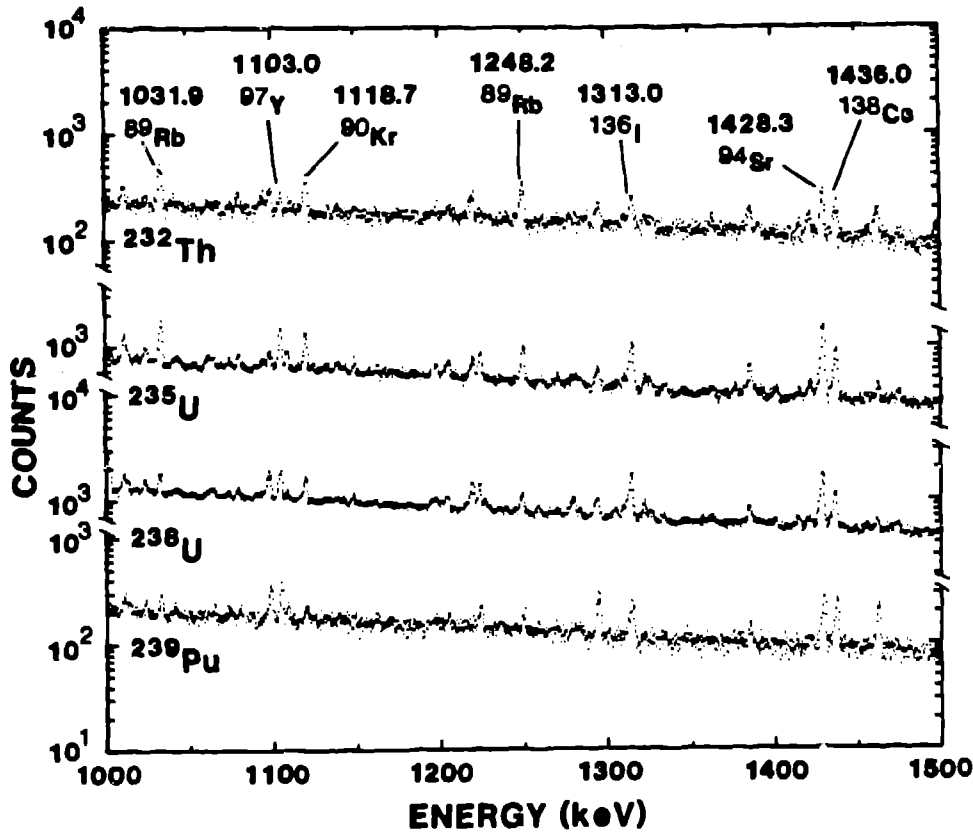


Figure. Gamma rays between 1000 and 1500 keV from fission products from photofission of ^{232}Th , ^{235}U , and ^{238}U , and ^{239}Pu . Selected peaks are labeled with the energy and associated fission product.