Analysis of Fissile Materials by High-Energy Neutron-Induced Fission Decay Gamma Rays

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Thermal neutrons from the Budapest Research Reactor and fast neutrons from the Berkeley Neutron Generator Facility have been used to analyze fissile materials. It has been shown that both prompt and delayed gamma rays from neutron capture and fission product decays can be used to analyze both uranium concentrations and ^{235}U enrichments. Spontaneous fission neutron emission from ^{238}U decay has also been found to be useful for uranium analysis. Detection of high-energy gamma rays following the decay of short-lived fission products was shown to be a sensitive indicator of fissile material, and the ratio of fission product gamma ray intensities can uniquely determine the concentrations of fission isotopes.

Introduction

The analysis of uranium and plutonium has gathered increasing importance in problems including nuclear waste remediation, nuclear nonproliferation, and battlefield exposure to depleted uranium. It is often desirable to perform these analyses nondestructively with complex sample geometries. Neutron probes are optimal for this analysis because neutrons easily penetrate dense materials producing fission neutrons and high-energy gamma rays that can readily escape. Thermal neutrons induce fission with a high cross section in ^{235}U (583 b)[1] and ^{239}Pu (748 b)[1] producing prompt neutrons up to 10 MeV, delayed neutrons up to 2.5 MeV, prompt gammas up to 5 MeV, and delayed gammas up to 8 MeV. Molnar et al[2] demonstrated quantitative analysis of ^{235}U/^{238}U enrichments with thermal neutrons by detection of prompt and delayed gamma. The more abundant isotopes ^{232}Th and ^{238}U have thermal neutron fission cross-sections of only <2.5 μb and 4 μb respectively[1]. High energy neutrons can be produced by neutron generators and fission ^{232}Th and ^{238}U with cross sections ranging from 0.1-1 b for neutron energies >2 MeV. In this work we have investigated the feasibility of analyzing fissile materials with 2.5 MeV neutrons produced at the Berkeley Neutron Generator Facility[3]. We will also discuss the analysis of ^{238}U by the detection of fast neutrons following the small, 5.45×10^{-5} %, spontaneous fission decay branch[4].

Experimental

Budapest Reactor Experiments

Experiments were conducted at both the Budapest Research Reactor using cold and thermal guided neutron beams. The thermal beam[5] had a thermal equivalent neutron flux of 2×10^{6} cm^{-2} s^{-1}. The cold beam[6] has a thermal equivalent neutron flux of 5×10^{7} cm^{-2} s^{-1}. The gamma rays were detected with a Compton suppressed HPGe detector. A beam chopper[6] was also used in some of the measurements and frequency of the chopper was set to 25 Hz. Each cycle was divided into 20 ms activation and counting intervals and spectra were recorded only during the middle 16 ms of each interval. Targets of U_3O_8 with ^{235}U enrichments of natural, 19.1%, 36%, and ≈95% were irradiated in the thermal and cold neutron beams. Gamma-ray spectra were analyzed using the Hypermet PC program[7]. To obtain emission rates, the intensities were corrected for the counting efficiency[8] and saturation of the activity[9]. Decay spectra were corrected for their different dead times, and for activation and counting time. Absolute gamma-ray cross sections were determined by comparison with the standard H 2223 keV gamma ray cross section, 0.3326 b[1], for targets of $UO_2(NO_3)\cdot6H_2O$ and $UO_2(CH_3COO)_2\cdot6H_2O$ with well known stoichiometry[10,11,12].

Berkeley D+D Neutron Generator Experiments

The Berkeley D+D neutron generator[3] utilizes RF-induction discharge to generate deuterium plasma. The neutron generator is based on a co-axial design, which maximizes the target area in compact outer dimensions of the generator and enables operation at high beam power, thus yielding high neutron fluxes. The generator was typically operated at an acceleration voltage of 120 kV and 50 mA deuterium beam current. This beam power

yielded a ≈2.5 MeV neutron flux of >10^9 n/s. Gamma rays were detected with a 20% efficient, relative to 3”×3” NaI, HPGe detector. The efficiency was calibrated with a certified multinuclide calibration source from Isotope Product Laboratories. Spontaneous fission neutrons were detected with a ^{3}He neutron detector that also served as a flux monitor for the neutron generator. A 0.89 kg target of uranium, depleted to 0.1% in ^{235}U, was counted to obtain the natural ^{238}U gamma ray spectrum. It was then irradiated next to the neutron generator for 10 minutes, manually removed from the irradiation chamber, and counted for 6 minutes, starting 1 minute after bombardment. A larger, 14.65 kg, sample of the same depleted uranium was counted directly with the ^{3}He neutron detector.

Results and Discussion

Budapest Reactor Data

The prompt and delayed gamma ray spectra produced following the irradiation of uranium enriched to 36% in ^{235}U is shown in Figure 1. Prompt gamma rays are observed up to 10 MeV, and the spectrum is dominated by a plethora of fission and continuum capture gamma rays. Several prompt gamma rays from neutron capture and fission were resolved and can be used for the quantitative analysis of ^{235}U and ^{238}U. Their cross sections are given in Table 1. In Figure 2 we show that the ^{235}U/^{238}U enrichment varies linearly with the ^{235}U(6296)/^{238}U(4060) prompt capture gamma ray intensity ratio, and the ratios of prompt fission gamma rays from ^{100}Zr and ^{134}Te. In the delayed spectrum gamma rays are observed up to 8 MeV, and many transitions are well resolved. These

fission product gamma rays would have unique relative intensities for each fissile material and will be discussed in detail below. We also note that the high-energy gamma rays in the delayed spectrum are intense and occur well above the highest energy background transitions <3 MeV. Detection of gamma rays 3-8 MeV with low resolution, high efficiency scintillation detectors is an attractive way to detect ^{235}U in ordinary materials and fissile materials in general.

Berkeley D+D *Neutron Generator Data*

The ^{238}U decay spectrum, counted for 60 minutes, is shown in Figure 3a. The prominent 1001-keV gamma ray is clearly visible and can be used for uranium analysis. Figure 3b shows a spectrum of the same sample, counted for 6 minutes, one minute after a 10-minute bombardment with ≈2.5 MeV neutrons. Gamma rays are observed up to 5 MeV, well above the highest transitions observed in the background. This spectrum cuts off at a lower energy than that observed in Budapest because most of the high-energy gammas are from short-lived fission products that decayed before counting.

In Table 2 the measured fission product gamma ray yields for ^{90}Rb ($t_{1/2}$=258 s), $^{90\text{-}m}$Rb ($t_{1/2}$=158 s), and ^{95}Y($t_{1/2}$=10.3 min) from ^{235}U (E_n=thermal) and ^{238}U (E_n=2.5 MeV) are shown. They are compared with tabulated fission product yields from ENDF-349[13] for ^{235}U, ^{238}U, and ^{239}Pu where the gamma-ray intensities are normalized using data from the Table of Isotopes[4]. Significant differences are observed in the experimental values for ^{235}U, ^{238}U, and the compiled values for ^{239}Pu suggesting that combined analysis with

thermal and fast neutrons can determine uranium enrichments and the relative abundance of all fissile material. The ^{235}U experimental and ENDF-349 thermal neutron induced fission yields agree well, but the ^{238}U experimental values agree poorly with compilations indicating possible problems with the database.

The neutron count rate, with and without the depleted uranium source, is shown in Figure 4, where the average count rate with the source present was 9.1 cpm with a background rate of 1.0 cpm. Spontaneous fission occurs in 5.45×10^{-5} % of all ^{238}U decays[4], corresponding to 410 fissions per kilogram per minute. Assuming that 3 neutrons are emitted per fission we expect the source to emit ≈1230 n/m. The detector was placed close to the extended source and the absolute neutron detection efficiency can be estimated as 1-2%, which is consistent with the expected source rate.

Conclusions

Several complementary methods for the detection of uranium and other fissile materials by neutron activation have been demonstrated. The observation of high-energy decay gamma rays >3 MeV is a unique signal of fissile material that can be exploited for quantitative uranium analysis. Detection of prompt neutron capture and fission product gamma rays can be used to accurately determine uranium depletion or analyze complex mixtures of fissile materials. A neutron generator can produce a broad range of neutron energies with different moderators, which is important for uranium analysis because thermal neutrons are uniquely sensitive ^{235}U while fast neutrons are sensitive to ^{238}U in

normal materials. Pulsing the neutron source on a short time scale has been shown to increase sensitivities to the short-lived fission products that provide the best gamma-ray signatures. It has also been shown that ^{238}U can be detected by its emission of spontaneous fission neutrons. This suggests that significant sensitivity can also be achieved by detection of delayed fission product neutrons that are emitted in 4.4% of ^{238}U fissions, but only 1.7% of ^{235}U fissions[14].

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Table 1. Elemental partial gamma ray production cross sections (σ_γ) for representative gamma rays detected during the activation of natural $UO_2(NO_3)\cdot 6H_2O$.

Target	Isotope	Transition Type	Energy	s_g (b)
^{238}U	^{239}Np	Decay daughter	278	0.382(6)
^{235}U	^{134}Te	Prompt fission γ-ray	297	0.22(2)
^{235}U	^{140}Ba	Prompt fission γ-ray	537	0.066(3)
^{235}U	^{134}Te	Prompt fission γ-ray	1279	0.20(1)
^{238}U	^{239}U	Prompt γ-ray	4060	0.191(2)
^{235}U	^{236}U	Prompt γ-ray	6395	0.0038(2)

Table 2. Comparison of measured and semi-empirical fission product gamma ray yields. Cold neutron measurements were performed at the Budapest Reactor and D+D measurements at the Berkeley Neutron Generator Facility. Semi-empirical values were obtained from the compilation of England and Rider (ENDF-349)[13].

		Relative Fission Yields				
		Experiment		ENDF-349		
Fission Product	Eg (keV)	^{235}U(cold)	^{238}U(2.5 MeV)	Thermal	Fast	239Pu(thermal)
^{90}Rb(258 s)	3317.00(17)	77	145	70	44	33
^{95}Y(10.3 m)	3575.84(20)	147	325	162	226	88
^{90}Rb(158 s)	4135.47(20)	100	100	84	84	84
^{90}Rb(158 s)	4365.82(24)	95	96	100	100	100

Figure Captions

Figure 1. Spectrum of prompt (upper) and fission product decay (lower) gamma rays produced following the irradiation of Uranium, enriched to 36% ^{235}U, with cold neutrons from the Budapest Reactor. The decay spectrum was acquired with a neutron beam chopper set to a 20 ms activation phase and 16 ms counting interval.

Figure 2. Analysis of ^{235}U/^{238}U ratios with prompt and fission gamma rays produced by bombardment with cold neutrons at the Budapest Reactor. The enrichment is observed to vary linearly with gamma-ray intensity.

Figure 3. Spectra of natural decay and fission product decay gamma rays produced before (a) and after (b) irradiation of depleted Uranium with ≈2.5 MeV neutrons from the D+D Berkeley Neutron Generator Facility. The natural decay spectrum represents one hour of counting, and the activation spectrum was counted for 6 minutes following a 10-minute bombardment.

Figure 4. Comparison of background and spontaneous fission rates measured with a ^{3}He detector 10 cm from 14.65 kg of depleted uranium. ^{238}U spontaneously fissions with a rate of ≈6 fissions per second per kg.

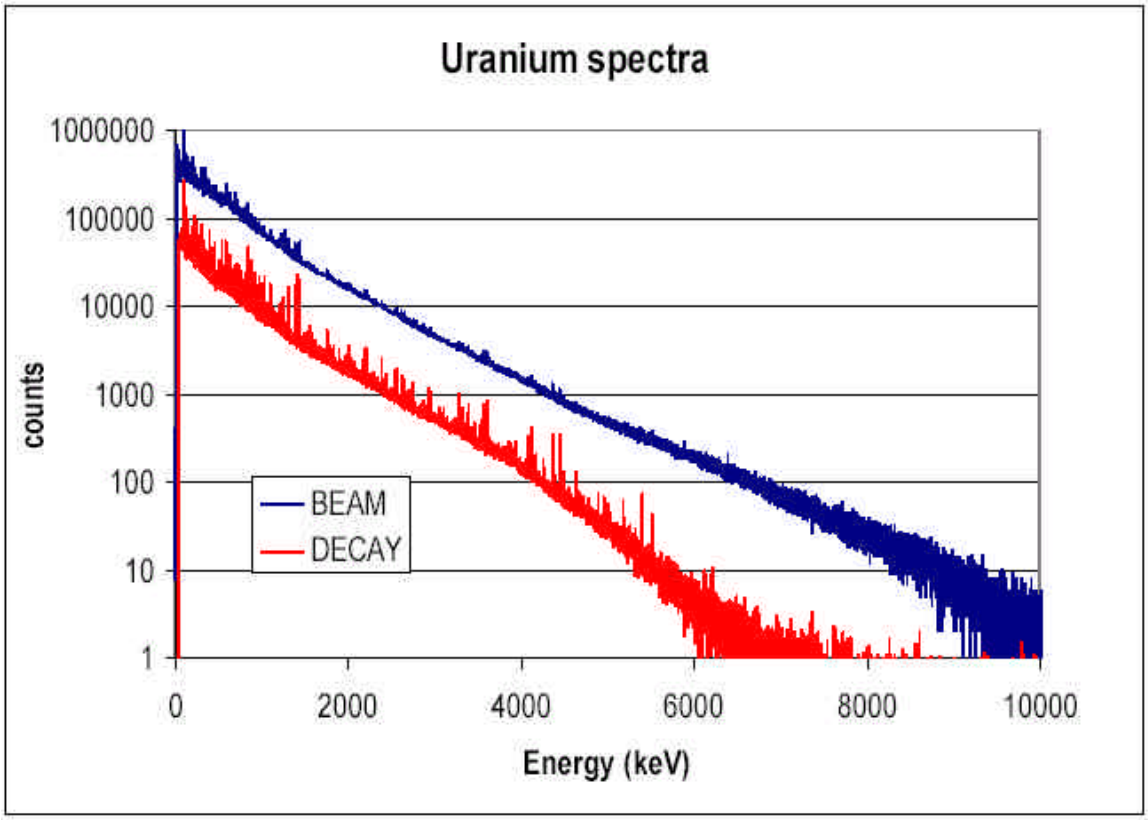
Uranium spectra
1000000
100000
10000
1000
100
10
1
counts
BEAM
DECAY
0
2000
4000
6000
8000
10000
Energy (keV)

Figure 1.

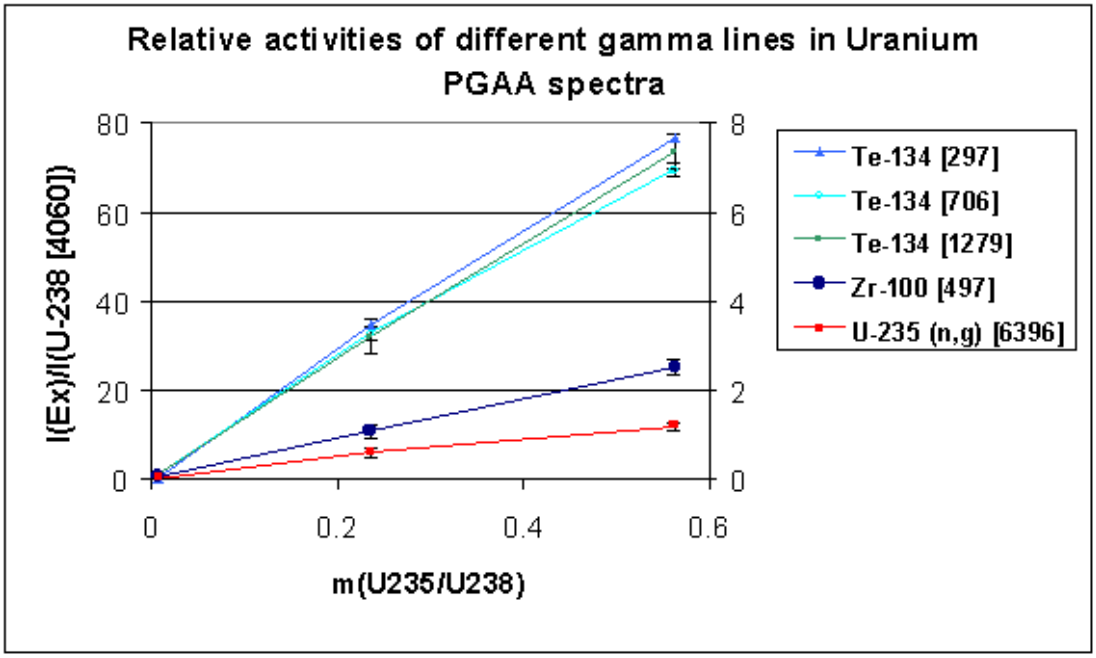
Relative activities of different gamma lines in Uranium PGAA spectra
I(Ex)/I(U-238 [4060])
80
60
40
20
0
8
6
4
2
0
0
0.2
0.4
0.6
m(U235/U238)
Te-134 [297]
Te-134 [706]
Te-134 [1279]
Zr-100 [497]
U-235 (n,g) [6396]

Figure 2.

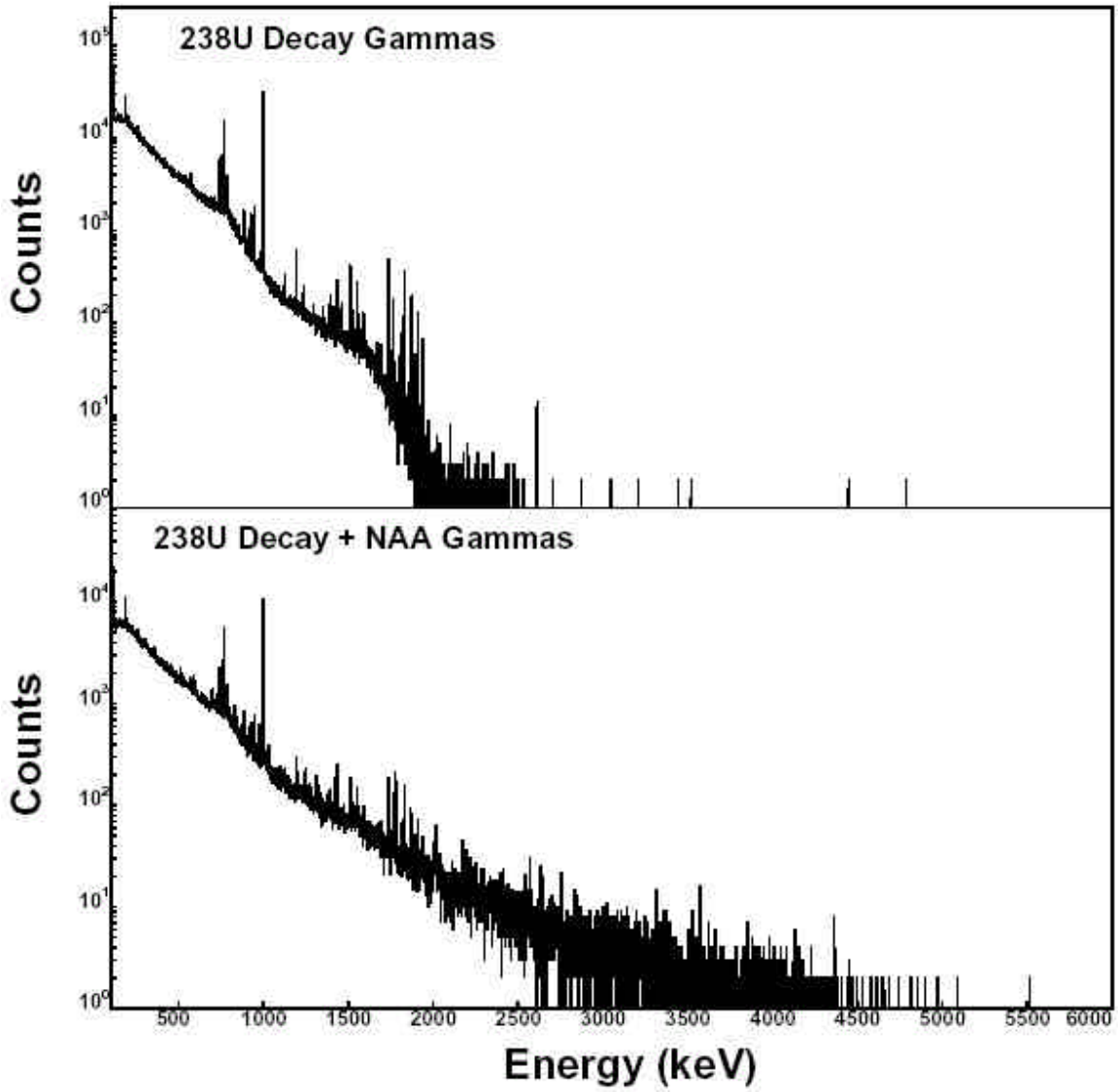
238U Decay Gammas
238U Decay + NAA Gammas
Counts
Counts
Energy (keV)
500
1000
1500
2000
2500
3000
3500
4000
4500
5000
5500
6000

Figure 3.

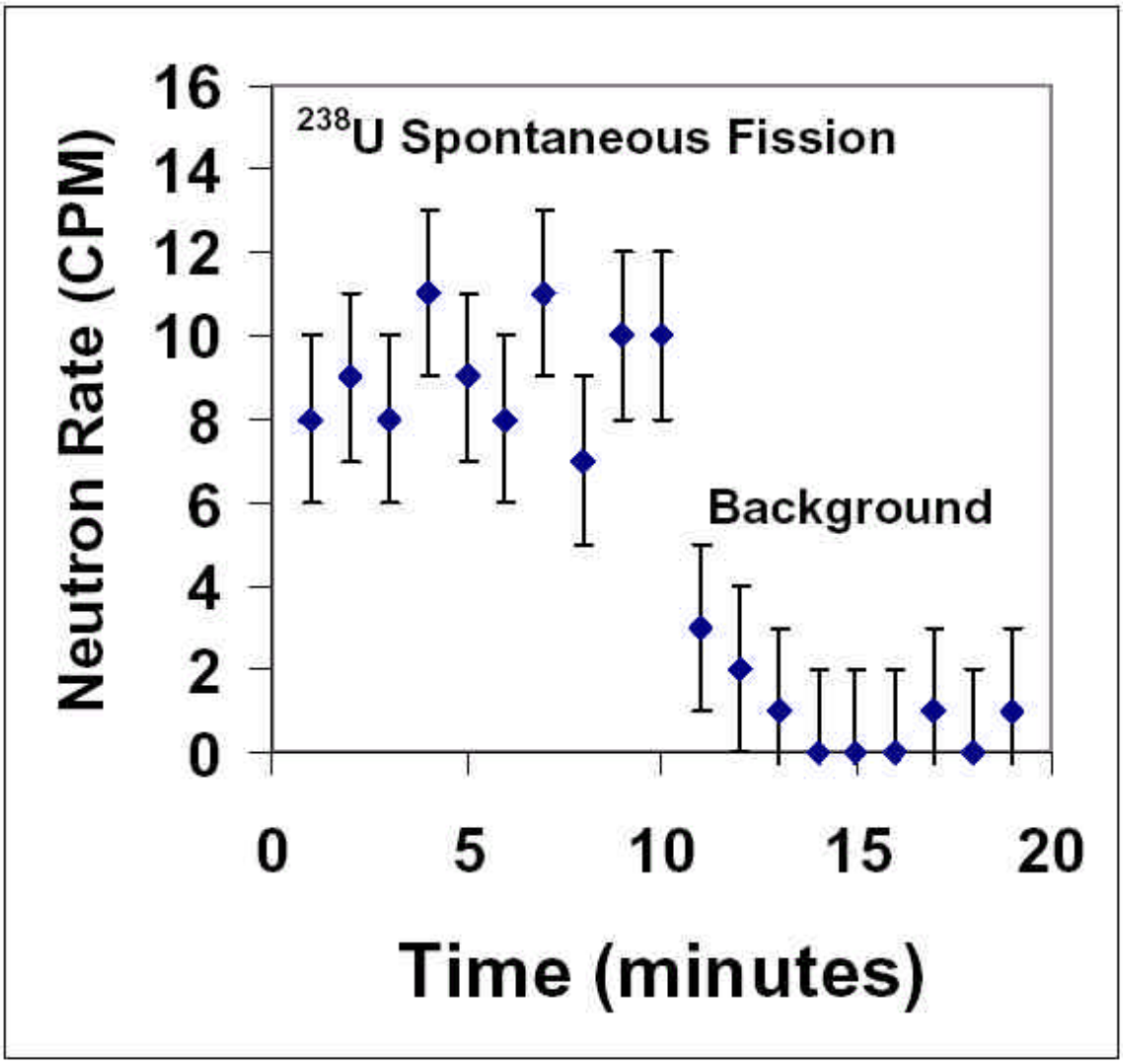
238U Spontaneous Fission
Background
Neutron Rate (CPM)
16
14
12
10
8
6
4
2
0
0
5
10
15
20
Time (minutes)

Figure 4.