J Radioanal Nucl Chem (2009) 282:901–904
DOI 10.1007/s10967-009-0283-4

Using low resolution gamma detectors to detect and differentiate ^{239}Pu and ^{235}U fissions

E. Swanberg · E. B. Norman · H. Shugart · S. G. Prussin · E. Browne

Received: 1 July 2009 / Published online: 30 July 2009

Abstract When ^{239}Pu and ^{235}U undergo thermal neutron-induced fission, both produce significant numbers of beta-delayed gamma rays with energies in the several megaelectron volt range. Experiments using high energy-resolution germanium detectors have shown that it is possible to distinguish the fission of ^{239}Pu from that of ^{235}U. It is desirable to detect the presence of ^{235}U or ^{239}Pu using detectors that are less expensive and more rugged than high purity germanium detectors. To this end we demonstrate how differences in the energy spectrum and decay rates of the beta-delayed gamma rays can be used to identify ^{239}Pu and ^{235}U using low resolution plastic and liquid scintillator detectors. Experimental data are used to identify differences in the spectra and also to test the identification algorithms. Results to date are very promising.

E. Swanberg (✉) · E. B. Norman · H. Shugart · S. G. Prussin
University of California at Berkeley, Berkeley, CA, USA
e-mail: swany@nuc.berkeley.edu

E. B. Norman
e-mail: ebnorman@lbl.gov

H. Shugart
e-mail: hshugart@physics.berkeley.edu

S. G. Prussin
e-mail: prussin@uclink4.berkeley.edu

E. B. Norman · E. Browne
Lawrence Berkeley National Lab, Berkeley, CA, USA

E. Browne
e-mail: ebrowne@lbl.gov

Keywords Fission · Beta-delayed gamma · Plastic scintillator · Liquid scintillator · ^{235}U · ^{239}Pu

Introduction

When irradiated with thermal neutrons, both ^{235}U and ^{239}Pu undergo fission. Most fission products are unstable and decay by beta emission accompanied by gamma rays. There is a delay between fission and the emission of gamma rays due to the half lives of the fission products. These gammas are referred to as beta-delayed gamma rays. The half life and the gamma energies vary by isotope. ^{235}U and ^{239}Pu fragment into a wide variety of fission products, but some products are more likely from one or the other of ^{235}U or ^{239}Pu [1]. It is this difference in fission products, and hence differences in gamma rays, that can be used to determine if ^{235}U, ^{239}Pu, or both are present.

Previous studies [2] have demonstrated that High Purity Germanium (HPGe) gamma detectors can identify the contributions of individual fission products soon after neutron irradiation. This is possible because HPGe detectors have good energy resolution and photopeak efficiency. Earlier studies [3, 4] measured delayed gammas using NaI and plastic scintillators, but no attempt was made to use the information to differentiate ^{235}U from ^{239}Pu.

Plastic and liquid scintillators have poor resolution and, owing to the low atomic number of their constituents, they have essentially no photopeak. To employ these detectors in finding ^{235}U and ^{239}Pu requires the use of much broader differences in the emitted gamma spectra than used in the HPGe studies. It is desirable to use low resolution scintillators to detect ^{235}U and ^{239}Pu because of their lower cost, lower maintenance requirements, and increased ruggedness.

Experimental

The 88 inch cyclotron at Lawrence Berkeley Lab was used to generate neutrons by bombarding a thick water cooled beryllium target with 16 MeV deuterons. The neutrons were moderated by iron and polyethylene and then incident on the target. The average thermal neutron flux at the irradiation site was about 1.5×10^6 cm^{-2} s^{-1}. Four different targets were used: a blank; 0.19 g of ^{235}U; 0.568 g of ^{239}Pu; or 3 g of latite, which is a common mineral containing a variety of nuclei including Si, O, Al, K, Ca, and Fe, among others. The targets were irradiated for 30 s and then pneumatically shuttled to the detection system in a separate experimental cave. Detectors included a plastic scintillator and a liquid scintillator. The transit time of the targets was approximately 1 s.

Ten measurements, each 2.5 s, were made immediately after the neutron irradiation. Making 10 separate measurements yields temporal information in addition to energy information. 223 sets of measurements were made, including 90 of ^{239}Pu, 94 of ^{235}U, 20 of a blank, and 19 of latite, for a total of 2,230 spectra each for the plastic and liquid scintillators. The 10 spectra in the series comprise the set of spectra for that sample.

To perform an energy calibration a water loop was installed that flowed between the neutron source and the detectors. The oxygen in the water was activated by the neutrons via an (n,p) reaction to produce ^{16}N, which quickly decays ($t_{1/2} = 7.13$ s) and emits gamma rays at 7.1 and 6.1 MeV. These high energy gammas, along with gammas from ^{60}Co and ^{137}Cs, were used to perform an energy calibration of the detector systems. Since no photopeaks were present, the Compton edges were used to determine energies in the spectra.

Data analysis

The low resolution spectra from these scintillators are nearly featureless (see Fig. 1). In a log plot the spectra look like a monotonically decreasing line. The main difference in the energy spectra is that ^{235}U has more high energy gammas. Their temporal behavior is also different. Figure 2 shows the decay for the energy range between 2.5 and 3.5 MeV. The ^{235}U tends to decay faster than the ^{239}Pu.

Since photopeaks are not present broader features of the spectra were used for analysis. All 10 time bins were used for analysis. Energies below 1.2 MeV were not used due to high background levels. Energies above 4 MeV were not used due to low count rates and poor statistics. The energy range between 1.2 and 4 MeV proved very useful during analysis.

To analyze the data, all the spectra were normalized, and then individual sets of spectra were compared to known basis sets of spectra for ^{235}U, ^{239}Pu, and latite spectra. Latite was used to represent a typical material not containing uranium or plutonium. The basis sets of spectra were made by summing all the sets of spectra of a given type. For example all 94 ^{235}U sets of spectra were summed to form the ^{235}U basis set, resulting in improved statistics over the individual sets of spectra. The same was done with ^{239}Pu, blank, and latite spectra. All spectra, including the basis spectra and individual spectra, were normalized by dividing each channel by the highest count channel in the set of spectra. Thus all spectra consist of normalized counts between 0 and 1, reducing dependence on absolute count rate and allowing comparisons between individual sets of spectra and basis sets of spectra. To test the analysis method, all of the individual sets of spectra, including ^{235}U, ^{239}Pu, blank and latite, were compared to the basis sets.

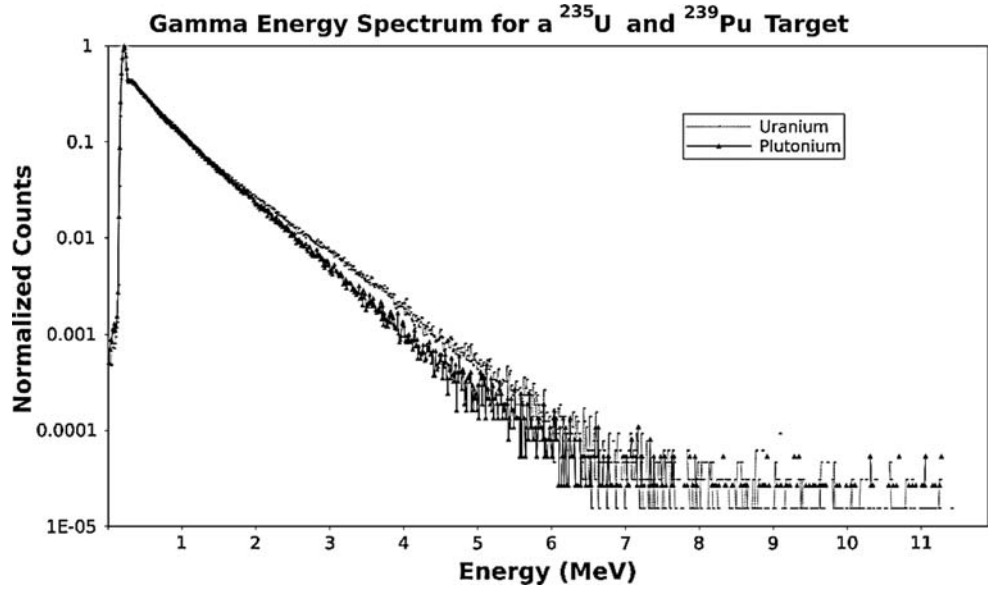

Fig. 1 A plot of normalized counts versus energy for a Uranium (*top line*) and a Plutonium (*bottom line*) sample. This plot is from the liquid scintillator. The plastic scintillator looks identical

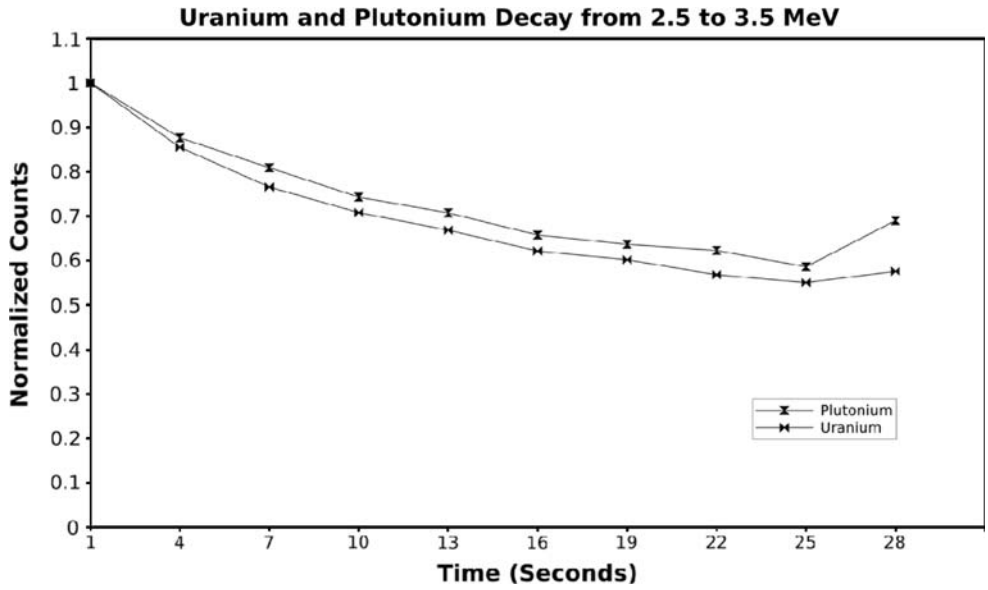

Fig. 2 The decay of counts from a Plutonium (*top line*) and a Uranium (*bottom line*) sample over the energy range 2.5–3.5 MeV. This is from the liquid scintillator. The plastic scintillator looks the same

The sets of spectra were compared by summing the absolute value of the differences at each point in energy and time for energies between 1.2 and 4 MeV and for all time bins. This sum is the residual. In equation form:

$$R = \sum_{E=1.2}^{4} \sum_{t=0}^{9} \left| B_{E,t} - I_{E,t} \right|$$

where R is the residual, B represents a basis set of spectra, I represents an individual set of spectra, E is the energy and t is the time bin. Whichever basis yields the lowest residual with a given individual set of spectra is assumed to be the content of that set.

This analysis method was run in two ways. In the first, the desired outcome was to label the individual spectra as either ^{235}U, ^{239}Pu, or neither. The first method did not consider the possibility of a mixed sample of ^{235}U and ^{239}Pu. Each set of spectra was compared to three basis sets (^{235}U, ^{239}Pu, and latite) to determine its composition. If the outcome matched what the individual set of spectra actually was then the method was successful.

The second method considers combinations of ^{235}U and ^{239}Pu. Since none of the actual measurements included combinations of ^{235}U and ^{239}Pu, synthetic sets of spectra were created using the sets of spectra that were collected experimentally. Spectra were created with varying levels of ^{235}U from 0 to 100% in 10% increments, with the remainder ^{239}Pu. The analysis of these synthetic spectra was similar to above, except now different basis sets were generated by mixing the ^{235}U and ^{239}Pu basis sets. B is now a combination of ^{235}U and ^{239}Pu bases with the fraction of ^{235}U varying from 0 to 100% (and ^{239}Pu varying from 100 to 0%) in 1% intervals. The sum of ^{235}U and ^{239}Pu was fixed to 100%. The mix of basis sets of spectra that resulted in the smallest residual with the synthetic set of spectra is the presumed composition of that set of spectra.

Results

When the goal was to determine if an individual set of spectra contained ^{235}U, ^{239}Pu, or neither, the analysis was able to predict the correct result 100% of the time. This is due, at least in part, to the high signal to noise ratio in the experiment. The blank and latite sets of spectra were never predicted to be ^{235}U or ^{239}Pu. This is good because it indicates a high level of background rejection.

Basis sets of spectra, being a sum of many individual spectra, have much smaller uncertainties than the individual sets of spectra. Therefore the uncertainties are dominated by the individual unknown sets of spectra being measured. There was an average of over 23,000 counts in the area of interest for individual sets of spectra. When the uncertainties in the residuals were calculated for the individual sets of spectra the average was $\sigma = 0.23$. The average difference in residuals for ^{235}U and ^{239}Pu sets of spectra was 0.42, for an average confidence level of 1.8σ. The difference between ^{235}U or ^{239}Pu sets of spectra residuals and latite residuals is 6.9 or 27σ. This large confidence shows there are large differences between latite spectra and spectra containing ^{235}U or ^{239}Pu, but is also the result of the high signal to noise ratio in our experiment.

Predicting the composition of a mixed sample was more challenging, but the results were still favorable. Table 1 contains a list comparing the average fraction of ^{235}U measured versus the actual amount of ^{235}U in the synthetic spectra. These are averages over a large number of

Table 1 Measurement of composition

Actual % U	0.0	10.0	20.0	30.0	40.0	50.0	60.0	70.0	80.0	90.0	100.0
Average measured % U	6.9	10.9	16.9	24.2	35.5	48.0	59.1	68.7	76.7	83.4	89.3
Standard deviation of results	10.2	12.9	15.7	18.9	17.1	13.3	11.7	11.9	11.9	10.4	8.5

comparisons, including plastic and liquid scintillators. It can be seen that the measured composition was typically within 15–20% of the actual composition of the synthetic spectra. While not highly accurate, this demonstrates it is possible to distinguish ^{235}U from ^{239}Pu using low resolution detectors.

Conclusions

Despite their low resolution, plastic and liquid scintillators can be used to determine the presence of ^{235}U or ^{239}Pu in a sample following neutron interrogation. Instead of identifying peaks belonging to various isotopes, general shapes in the energy spectrum and temporal behavior are used. Our experimental setup yielded very high accuracy when determining the presence of ^{235}U or ^{239}Pu. In a real world application the signal to noise ratio would likely be much worse and would place limits on the minimum detectable quantity. Detecting the presence of ^{235}U or ^{239}Pu is easier than determining the composition of a mixture of the two. Our analysis allowed measurements of composition to within 15–20% of the actual composition.

References

1. England TR, Rider BF (2004) ENDF-349, LA-UR-94-3106
2. Marrs RE et al (2008) Nucl Instrum Meth A 592:463
3. Berick AC, Evans AE, Meissner JA (1969) Phys Rev 187:1506
4. Maienschein FC, Peelle RW, Zobel W, Love TA (1958) Gamma rays associated with fission. In: Second United Nations international conference on the peaceful uses of atomic energy, USA