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ABSTRACT

URANIUM ASSAY BY DELAYED GAMMA RAY ANALYSIS
FOLLOWING CF-252 NEUTRON INTERROGATIONG. D. Atkinson, Jr., and S. J. Gage
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A technique for the nondestructive assay of fissionable nuclides is described which utilizes a californium-252 neutron source, cyclic activation, and delayed gamma ray analysis.

The results of preliminary experiments with uranium-235 samples are presented, comparing the effects of various cyclic activation sequences and sample enrichments on the high energy gamma ray spectra of the short-lived fission products. The quantitative accuracies of the method are also presented.

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URANIUM ASSAY BY DELAYED GAMMA RAY ANALYSIS FOLLOWING CALIFORNIUM-252 NEUTRON INTERROGATION

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As part of the Californium-252 Market Evaluation Program, researchers at the Nuclear Reactor Laboratory at The University of Texas at Austin are investigating measurement techniques for the nondestructive assay of special nuclear materials. One technique showing promise is based upon the very discriminating gamma ray spectra of the short-lived fission products, observed following interrogation of fissionable samples by neutrons from a californium-252 neutron source. Cyclic activation is used in this technique to enhance the detection of gamma radiation from the shorter-lived fission products.

In this paper, the bases of the proposed technique will be presented, followed by the results of experiments performed with a moderated spectrum of interrogation neutrons.

Bases of Proposed Technique

The main features of the proposed active interrogation involve use of the following: 1) Cf-252 neutron interrogation, 2) cyclic activation, and 3) gamma ray analysis.

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Californium-252 Neutron Source

Much can be said in behalf of stimulation by neutrons. Certainly the most important benefits include our knowledge of neutron interactions, the ease with which neutrons may be produced, and the relative ease with which neutrons may be shielded.

There are two distinct advantages to using californium-252 as the source of interrogating neutrons: 1) radioisotopic source reliability and 2) a convenient neutron energy distribution.

When nonlaboratory uses are envisioned, the radioisotopic neutron source has distinct advantages over accelerators. A radioisotopic source does not require an operator, nor does it normally require maintenance. Obviously there is no problem with target deterioration or target handling.

The radioisotopic source californium-252, with an effective half-life of 2.65 years, offers considerable potential as a source of interrogating neutrons. Providing 2.3×10^{12} neutrons per second per gram, it yields an essentially constant neutron flux.

The neutron energy distribution from Cf-252 is a fission spectrum somewhat harder than that of U-235. (1) A plot of the Cf-252 neutron spectrum is shown in Figure 1. The maximum occurs near 1 MeV and the average energy is 2.3 MeV. Because the full logarithmic representation is somewhat deceiving, the most probable spectral distribution is also indicated. Thus, it is seen that 50% of the neutrons have energies between 0.95 and 3.2 MeV, 90% between 0.28 and 6.0 MeV, and 98% between 0.09 and 8.9 MeV. Only 2% of the neutrons have energies above 8.9 MeV or below 0.09 MeV.

Figure 2 shows this distribution superimposed on the fission cross sections of U-235 and U-238. The raw californium-252 spectrum would thus be suitable for stimulation of these two isotopes (as well as other fissile and fertile isotopes). There are no resonances in this region, nor are there any naturally occurring nuclides with cross sections above 10 b. Self-shielding is thus minimized.

Californium-252 is also favorable as a source of thermal neutrons, simply by moderating the fission spectrum. The softness of the resultant spectrum may be varied by the use of moderators such as W, Be-9, and D₂O. (2)

Cyclic Activation

Cyclic activation is the repetitive irradiation and counting of a single sample. The technique may be performed mechanically, by transferring the sample between the source and the detector, or electronically, by use of a pulsed accelerator source. If the total analysis time is significantly longer than the half-life of the nuclide being counted, then more total counts will be detected by cyclic activation than by conventional activation. Once saturation activity is reached in the cyclic technique, the total counts detected increases linearly. (3)

Cyclic activation may be used as a type of band-pass filter, determined by the total number of analysis cycles and the lengths of time spent in each sample irradiation, transfer to the counting area and cooling, counting, and transfer back to the irradiation area. Examples of the band-pass effect are the analysis sequences used in the experiment to be

described. Table 1 shows the timing sequences used in this study. Figure 3 shows the total counts detected as a function of half-life for each of the three major sequences. Note that in sequence 3 the addition of an 8 second cooling time had the effect of shifting the lower cut-off toward longer half-lives.

In the proposed analysis the fission products whose activity yields high energy gamma rays are expected to have half-lives on the order of seconds. It then follows that in order to maximize the total counts detected in experiments lasting several minutes it is advantageous to use cyclic activation.

Gamma Ray Analysis

The advantage in using gamma ray analysis is that the gamma ray response following neutron-induced fission is measurably unique with respect to time and energy for the fissionable nuclides of interest. (4)

The energy resolution with gamma ray techniques makes it possible to discriminate among fissionable isotopes by virtue of the spectral differences alone. This means that only a single spectrum of interrogating neutrons may be needed to determine the masses of all fissile and fertile isotopes present in a sample.

It is desirable to obtain gamma rays with energies above 511 keV, because of the decrease in the gamma ray attenuation coefficient with increasing energy. With high energy gamma rays (above 511 keV), self-absorption within the sample is minimized, a desirable trait for an assay technique.

Since the gamma ray spectra for fission products are harder in the "delayed" time domain closest to the fission event, cyclic activation would be useful to enhance detection of this effect. It is also known that post-fission phenomena generally are more characteristic of the fissioning nuclides in the time regions closer to the fission event. Thus, the most distinctive nature of the delayed gamma ray spectra is detected by analysis of the short-lived fission products, with the accompanying desirable increase in the hardness of the gamma spectrum.

Collective Advantages

Collectively these features of the proposed assay technique offer considerable potential, especially under non-laboratory conditions.

The interrogating source is intense, constant, dependable, and maintenance free. It is also easy to shield. Interrogation may be performed with a fission spectrum, a thermal spectrum, or an intermediate "reactor-like" spectrum.

With cyclic activation the samples emit the maximum photon intensity with a minimum amount of irradiation and counting. Long term emission of radiation from the sample is thus also kept at a minimum.

With gamma ray analysis the spectral response is so distinctive that potentially only a single interrogation spectrum need be used. With a tailored interrogation spectrum, the discriminating power of the method may be further enhanced.

Experimental Components

As shown in Figure 4, the experimental components include a Californium-252 neutron source and shield, a pneumatic transfer system, a Ge(Li) gamma ray detector, and a

multichannel analyzer.

The californium-252 neutron source contained approximately 1 milligram of californium-252. The source was suspended within the central cavity of an experimental facility made of water-extended polyester resin. The source was moderated by a paraffin cylinder which enclosed the pneumatic transfer tube.

A pneumatic transfer system was used to transport polyethylene sample vials a distance of 8 ft. between the irradiation port and the counting port. Transfer times as short as 100 milliseconds have been used, but 2 second transfers were used in this study to minimize damage to the transfer vials.

The Ge(Li) detector had an active volume of 37 cc. Its full-width at half-maximum resolution for the 1332 keV Co-60 peak was 3.9 keV. The peak-to-Compton edge ratio was 12.5. To reduce the spectra contribution from gamma radiation below 511 keV, 0.2 in. of lead was placed at the face of the detector. Not shown is a 4 in. lead castle which surrounded the detector, nor a shadow shield consisting of 8 in. of lead and 15 in. of concrete.

Spectra were recorded with a 2048 channel analyzer. The ADC had a built-in baseline restorer, and was timed by a 100 MHz clock.

Experimental Results.

Five uranium samples were analyzed using the cyclic analysis sequences previously described. The samples each had uranium masses of approximately 1 gm, with uranium-235 enrichments varying from 23 ppm to 93%. The uranium-235

masses were verified by passive gamma spectrometry before the samples were subjected to any interrogating radiation.

Qualitative Results

Shown in Figure 5 is the fission product spectrum obtained from sample U-030* in analysis sequence 3, from 0 to 4400 keV. This spectrum is generally characteristic of all the spectra obtained in this study. The most significant peaks in the spectrum are found in the energy range from 511 keV to 1427 keV. Other conspicuous characteristics are the interference from the combined continua resulting from Compton distributions, the fission product gamma ray continuum, and the bremsstrahlung induced within the sample by beta decays; and the host of small, interfered peaks.

In Figure 6 are the lower energy halves of the spectra from samples U-930, U-200, and U-020 for sequence 1. Note that the noise increases as the uranium-235 enrichment is decreased. In Figure 7 it is seen that the spectra for sequence 1 degrade completely for uranium-235 enrichments below 20%.

In sequence 2 the signal-to-noise ratio is improved, as is shown in Figure 8. Again, the spectra degrade for lower enrichments.

In sequence 3 (see Figure 9) the signal-to-noise ratios were best, with sample U-930 exhibiting 44 gamma ray peaks with net peak areas significant at the 3 σ confidence limit. Resolution for the 93% enriched sample was poor, however, due to count-rate dependent losses and gain and zero shifts.

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*The uranium-235 enrichments of the samples are given in the sample designations, U-nnn, where nnn is the enrichment in tenths of percent.

The energies of the various gamma ray peaks were compared with those stored in two reference tables, one for fission products (5,6) and one for radioisotopes produced in reactor irradiation (7). The fission product reference indicated that in most cases individual peaks observed in the spectra were not from gamma rays from a single nuclide: instead, peaks were found to be composites from not only more than one nuclide, but also from the three types of peaks detectable, total absorption peaks and single and double escape peaks. This further emphasized the point that a single exponential may be a poor approximation for the decay of a single peak.

Shown in Table 2 are the energies of the ten most significant peaks found in sequence 3. These energies, as well as those of the other significant peaks in the spectra, were compared with those found in the reference for reactor irradiation. In no case were the gamma ray energies of reactor produced radionuclides close enough to those of fission products that the technique might be deceived by substitution of nonfissionable nuclides. The nearest imposter for the large 1427 keV peak, for example, is the 1434 keV total absorption peak of vanadium-52. As longer analysis sequences are used, however, the fission products emitting the most significant gamma rays in the spectrum may tend to be close to the line of nuclear stability and therefore produceable by (n, γ) reactions.

As was expected, the gamma ray spectra did indeed harden for the shorter analysis sequences. In Table 3 it is seen

that the mean energy of the most significant peaks increases markedly in going from sequence 3 to sequence 1. Unfortunately, peak statistics were poorer in sequence 1 and 2 than in sequence 3.

Extending the total analysis time while maintaining short cycle times, however, did not provide both high peak energies and better peak statistics. Instead, as is shown in Figure 10 and 11, the peak statistics were degraded. The total number of counts obtained per channel increased, and so did most peak areas. The continuum contribution also increased, which added to the noise in the spectra and raised the peak baselines.

Quantitative Results

High count-rate losses for samples U-930 and U-200 and poor peak statistics for samples U-030 and U-020 prevented the direct acquisition of quantitative results typical of this technique. The results expected in a system with optimum electronics and repetitively analyzed standards, however, were approximated. The net peak areas of the ten most significant peaks in each spectrum observed were used to describe standards. It was then assumed that the samples being assayed would have similar peak statistics. The results from simulated least-squares fits of such data are indicated in Table 4.

A wide range of accuracies are possible for the technique, depending upon the analysis sequence chosen and the size or enrichment of the sample. The accuracies would obviously improve if more than 10 of the significant peaks were used to describe the standards.

Summary

The active interrogations exhibited the following characteristics: 1) highly activated samples, 2) complex gamma ray spectra degraded by continua, 3) gamma ray peaks unique to fission products, 4) gamma ray peaks of higher energy for short cycle times, and 5) marginal quantitative analyses.

With a 1 mg californium-252 neutron source and a 37 cc Ge(Li) gamma ray detector, a 1 gm sample of uranium-235 activated sufficiently to exceed the data handling ability of the gamma ray detection system. The analyzer count rate losses for sample U-930 indicated that for the time periods studied the maximum amount of high energy gamma ray information was being obtained.

The complex gamma ray spectra obtained were found to be composed of a multitude of unresolved peaks. Peak analysis was hampered by the presence of strong continuum interference.

The gamma ray peaks comprising the elements of the standards were found to be unique to fission products. Potential imposters would be detected by the improper energies of their peaks and the presence of peaks in areas expected to be barren.

The mean, weighted energy of the most significant gamma rays was found to shift to higher energies as the irradiation-counting cycle times were shortened. This shift was accompanied by an increase in the continuum interference, which in turn reduced the significance of the peaks detected.

The quantitative results were marginal, caused not entirely by effects intrinsic to the technique, but by the systematic effects of count-rate losses and gain-zero shifts--

with improved instrumentation these limitations could have been overcome.

In conclusion, the technique investigated provides another source of signatures unique to each fissionable nuclide of interest, using a dependable and versatile interrogation source and potentially short analysis times.

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Table 1.

CYCLIC ACTIVATION TIMING SEQUENCES

<u>Designation</u>	<u>Trips</u>	<u>Time in Seconds</u>						
		<u>Transfer In</u>	<u>Irradiate</u>	<u>Transfer Out</u>	<u>Cool</u>	<u>Count</u>	<u>Cycle</u>	<u>Total</u>
1	10	2	2	2	0	2	8	80
2	10	2	10	2	0	10	24	240
3	10	2	60	2	8	60	132	1320
4*	30	2	2	2	0	2	8	240
5**	55	2	10	2	0	10	24	1320

*Sample U-930 Only

**Sample U-200 Only

TABLE 2

**ENERGIES OF THE MOST SIGNIFICANT PEAKS,
STANDARD U-200, SEQUENCE 3**

Energy, keV

540.4

589.8

602.6

724.0

831.1

973.7

1117.9

1312.4

1426.9

1564.2

TABLE 3.

**AVERAGE WEIGHTED ENERGIES OF PEAKS USED IN QUANTITATIVE ANALYSES,
STANDARDS U-930 and U-200.**

<u>Sequence</u>	<u>Energy in keV</u>	
	<u>U-930</u>	<u>U-200</u>
1	1720	2066
2	1371	1693
3	913	961

TABLE 4.
LOWER ERROR LIMITS*

<u>standard</u>	<u>Sequence 1</u>	<u>Sequence 2</u>	<u>Sequence 3</u>
U-930	12	9.5	3.5
U-200	14	10	3.8
U-030	n.a.	20	6.9
U-020	26	13	8.5

*Estimated standard deviation, given in percent.

1. Californium-252 Neutron Spectrum

2. Californium-252 Neutron Distribution and Fission Cross Sections

3. Total Detected Counts Per Total Sequence vs. Half-life

Figure 4. Block Diagram of the Ge(Li) Spectrometry System

5. Fission Product Spectrum, Sequence 3,
Sample U-030

6. Fission Product Spectra, Sequence 1,
Samples U-930, U-200, and U-020.

7. Fission Product Spectra, Sequence 1,
Samples U-030 and U-000.

8. Fission Product Spectra, Sequence 2,
Samples U-930, U-200, and U-020.

SEQUENCE 3
U-930, U-200, U-020

9. Fission Product Spectra, Sequence 3,
Samples U-930, U-200, and U-020.

10..Fission Product Spectra, Sequences 3 and 5,
Sample U-200.

11. Fission Product Spectra, Sequences 2 and 4,
Sample U-930.