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NON-DESTRUCTIVE ANALYSIS OF URANIUM IN GRAPHITE FUEL ELEMENTS BY NEUTRON ACTIVATION

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ABSTRACT

A method is presented for the determination of uranium (as U-238) in uranium-loaded graphite fuel elements by a non-destructive or direct radioactivity analysis technique. A 200-channel pulse-height analyzer, equipped with a 3 in. x 3 in. thallium-activated sodium iodide crystal, is used to measure the neptunium-239 (2.33 d) radioactivity of the neutron-irradiated samples. The amount of U-238 in the test samples is determined quantitatively by comparing the Np^{239} radioactivity induced in each sample with the Np^{239} radioactivity induced into known standards of U-238 and processed under the same conditions as the test samples. The limit of detection for U-238 in samples of normal uranium composition for this method is about 1.0×10^{-4} μg.

This nondestructive analysis technique involving gamma spectrometry by pulse-height analysis[4] has been used to supplement quantitative radiochemical analytical procedures. Its use has made it possible to obtain very rapid and positive qualitative identification of radionuclides and very accurate quantitative determination of radionuclides. By combining this and conventional analytical procedures according to the requirements of a particular analysis problem, it is possible to realize significant benefits in speed, simplicity, accuracy and precision in making such determinations. The nondestructive technique employed, as reported here, has all of the above benefits. It consists of neutron irradiation followed by gamma spectrometry of the induced radionuclide. To obtain quantitative results, the gamma-spectral data are converted to grams/gram data by use of a known standard and absolute methods of calculation.[2]

Procedure

The procedure consists of five parts: (a) preparation of sample for irradiation, (b) irradiation, (c) preparation for analysis, (d) analyzing, and (e) calculations.

(a) The preparation of a sample for irradiation consists of weighing out portions of the dry U-graphite test sample into a small plastic vial (10 mm dia x 30 mm height). The sample is then placed into a 6 oz polyethylene bottle together with U^{238} standard solutions of known U concentration.

(b) The irradiation of the samples and standards was made in Hole No. 71 of the Oak Ridge Graphite Reactor. This is a water-cooled, dry hole, irradiation facility, maintaining a temperature of $\sim 70^{\circ}F$; the flux in this position is $\sim 7 \times 10^{11}$ n/cm^2/sec. Irradiation times of 62 hours were used in order to

produce a measurable amount of Np^{239} radioactivity. Np-239 is produced as the result of an (n,γ) reaction upon U^{238}.

$$U^{238}(n,\gamma)U^{239} \xrightarrow[23.4m]{\beta^-\gamma} Np^{239} \quad t_{1/2} = 2.346d$$

(c) After the irradiation, the samples are returned from the reactor to a "hot" laboratory facility. The U-graphite material and U-standard solution contained in the small plastic vials are removed from the polyethylene bottle, surveyed for safe radioactivity work level and visually inspected for defects which would or could cause area and personnel contamination. If safe, the samples are then removed from the "hot" laboratory facility to a work area hood and prepared for counting. This preparation consists of transferring the irradiated test samples into a pyrex culture tube (10 mm x 75 mm). Suitable aliquots of standard U-solution are pipetted into other culture tubes. The aliquot is adequate enough to provide an activity of the same order of magnitude as the samples in question.

(d) The nondestructive analysis of the samples is performed by use of a 200-channel pulse-height analyzer (Model 34-2)* equipped with a 3 in. x 3 in. thallium-activated sodium iodide scintillation crystal. The samples and standard are counted under the same conditions, i.e., counting time, geometry, and instrument gain. The photopeak of interest in this analysis is the gamma ray photopeak energy 0.105 Mev of Np-239. (Figure 1) Thus, an instrument gain of 0-1 Mev for the 200 channels is used for the radioactivity measurements; the count rate per sample includes the total count of seven channels 0.090-0.120 Mev over the 0.105 Mev photopeak.

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*Radiation Instrument Detection Laboratories (RIDL), Northlake, Illinois

(e) The calculations for determining the U^{238} content of the test samples are very simple. Since all of the terms in the activation analysis equation,

$$W = \frac{AM}{(6.02 \times 10^{23})(f)(\bar{\sigma}ac)(\theta)(1-e^{-\lambda t})(e^{-\lambda t})} \qquad (1)$$

A = Radioactivity, dps

M = atomic weight of the target element

6.02×10^{23} = Avogadro's number

f = neutron flux, $n/cm^2/sec$

$\bar{\sigma}ac$ = activation cross-section for the isotope, barns, 10^{-24} cm^2

θ = fractional abundance of the target isotope

$1-e^{-\lambda t}$ = saturation factor

$e^{-\lambda t}$ = decay factor

W = weight in grams

are the same for both the comparator and the unknown samples, except those for weight (W) and activity (A), then a simplified equation can result:

$$\frac{\text{W of } U^{238} \text{ in Test Sample}}{\text{W of } U^{238} \text{ in Comparator Sample}} = \frac{Np^{239} \text{ Radioactivity in Test Sample}}{Np^{239} \text{ Radioactivity in Comparator Sample}} \qquad (2)$$

Discussion

The high sensitivity of the activation analysis method[3] makes it possible to see low concentrations of many elements including uranium. In this report the limit of sensitivity is not approached from the concentration of the uranium in the graphite, but rather from the sample weight used (as shown from the results presented in Table I). Nondestructive methods of

TABLE I

URANIUM IN GRAPHITE

Graphite Sample	Uranium-238 Concentration, Micrograms per gram*
1	512, 516
2	505, 507
3	493, 495
4	532, 540

*Sample portions in the range from 0.060 to 0.072 gram were irradiated in this work.

analysis in neutron activation analysis are useful when employing a sample matrix such as graphite that has a low neutron absorption cross-section and will produce only very low-energy radiations or a minimal amount of radioactivity during an irradiation. Thus, an analysis of the type reported here may be completed by a direct measurement without chemical separation and with a high degree of sensitivity, speed, accuracy, and reproducibility.

Since the gamma ray spectrums of the test and standard samples are compared to detect any impurities in the test samples, the non-destructive method may easily be employed. This technique may be applied to any single radionuclide in either a solid or a liquid sample, where it is directly neutron induced or fission produced. Also, this method may be utilized where the desired radionuclide has the highest gamma energy present in a sample of several radionuclides. When the gamma energy or radionuclide of interest has other gamma energies higher or near the desired gamma energy, the technique of complement subtraction may be applied successfully to strip out the desired radionuclide.

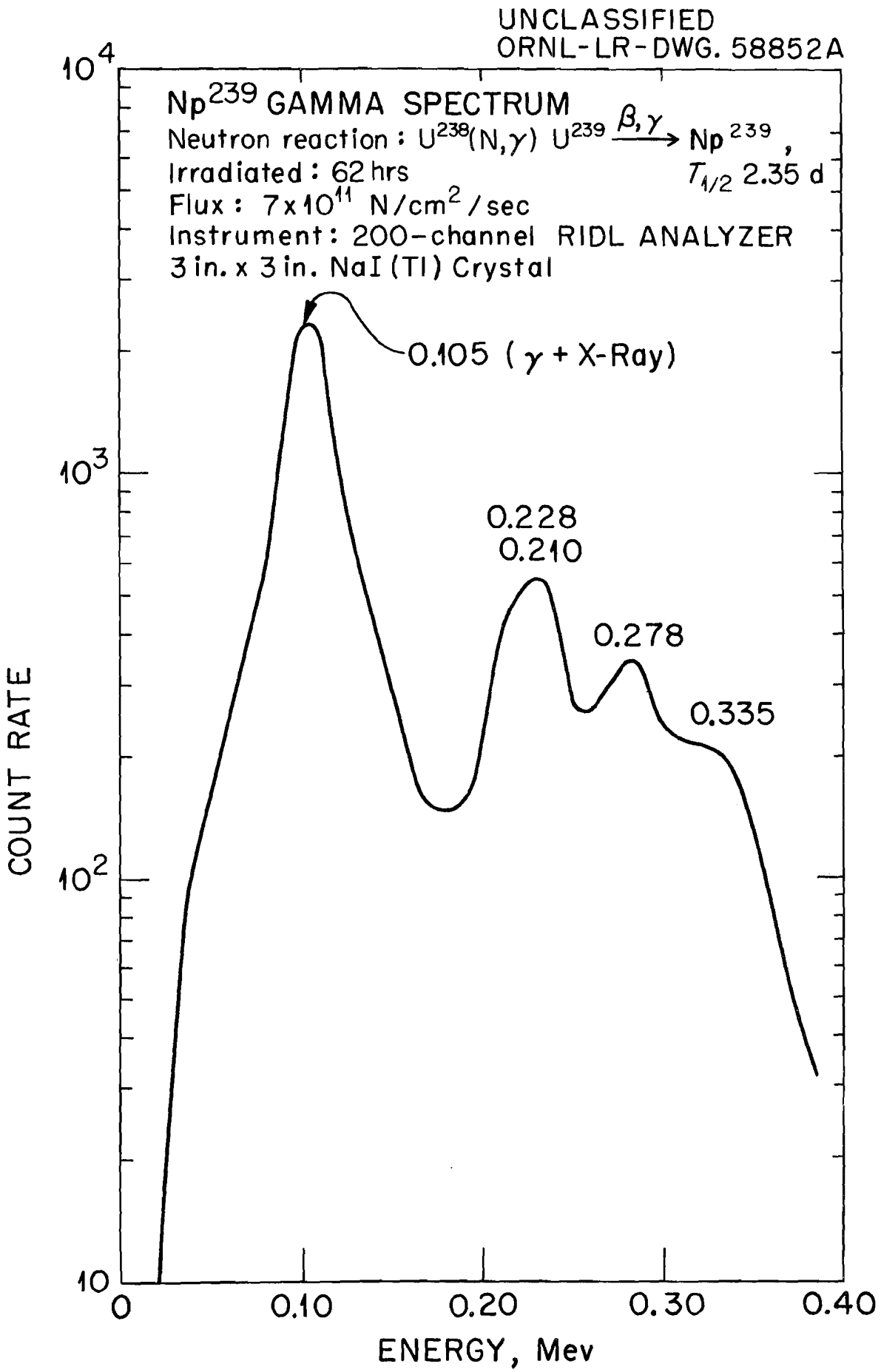
References

1. ORNL-2866, Analytical Chemistry Progress Report - Neutron Activation and Radioactivity Measurements, pp. 22 and 33 (1959).

2. Boyd, G. E., "Method of Activation Analysis," Anal. Chem., Vol. 21 (1949), p. 335.

3. Leddicotte, G. W., "The Use of Neutron Activation in Analytical Chemistry," 2nd UN Geneva Conference on the Peaceful Uses of Atomic Energy, 1958, p. 927 (1959).

4. Bate, L. C., and Leddicotte, G. W., "The Quantitative Analysis of Complex Radionuclide Mixtures By Gamma-Ray Spectrometry," ORNL-2917 (In preparation).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Np239 GAMMA SPECTRUM
Neutron reaction : U238(N,γ) U239 β,γ → Np 239 ,
T1/2 2.35 d
Irradiated : 62 hrs
Flux : 7x10^11 N/cm^2 / sec
Instrument : 200-channel RIDL ANALYZER
3 in. x 3 in. NaI (Tl) Crystal
0.105 (γ + X-Ray)
0.228
0.210
0.278
0.335
COUNT RATE
10^4
10^3
10^2
10
0
0.10
0.20
0.30
0.40
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