THE DETERMINATION OF $^{235}\text{U}/^{238}\text{U}$ RATIOS BY ACTIVATION ANALYSIS UTILIZING HIGH RESOLUTION $\gamma$-SPECTROMETRY

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The application of high resolution $\gamma$-spectrometry to the direct determination of $^{235}\text{U}/^{238}\text{U}$ isotopic ratios in small uranium samples after neutron activation is reported. The consistency obtained relative to mass spectrometrically determined isotopic ratios is better than 5% for isotopic ratios between 0.06885 and 1.095. The method is not subject to the main disadvantages of the known radiochemical methods for $^{235}\text{U}/^{238}\text{U}$ isotopic ratio determinations, and should be appreciably more rapid compared to the solid-source mass spectrometric method giving the same precision.

Introduction

A knowledge of the $^{235}\text{U}/^{238}\text{U}$ isotopic ratios of uranium is of great importance in nuclear technology, as well as in miscellaneous studies in which enriched or depleted uranium is used. Isotopic analysis is also required in determinations of the uranium content of samples by isotope dilution analysis. This is almost invariably done with a solid-source mass spectrometer. A high degree of precision (well below 1%) is obtainable by this method, depending on the time spent on the analyses and provided no interfering peaks are encountered during mass analysis. Less accurate radiochemical methods of determining $^{235}\text{U}$ abundances by neutron activation have also been published, but they suffer the serious disadvantage that the total amount of uranium irradiated has to be known exactly. Radiochemical isolation of a certain fission product, chemical yield determinations, absolute radioactivity measurements, and the necessity to treat a known amount of a standard of known isotopic abundance analogously, render these methods time-consuming and subject to additional errors. Even when precision and accuracy are not required, the mass spectrometric method is usually still far superior. Impurities in samples may, however, occasionally hamper the mass spectrometric approach.

In recent years, the advent of semiconductor detectors in $\gamma$-spectrometry, particularly the Ge(Li) detector, has aroused considerable interest on account of their excellent resolving power. The Ge(Li) detector permits studies of complex radioactive mixtures, such as fission products, in much greater detail than
was previously possible with the conventional NaI(Tl) scintillation spectrometers. It is already applied as a powerful tool in nuclear decay-scheme studies, in multiple-element analysis by activation analysis, in range and yield determinations of fission products, and in the burn-up analysis of nuclear reactor fuels. Although gross fission products have been studied with Ge(Li) detectors, little attention has been given to the possibility of determining $^{235}\text{U}/^{238}\text{U}$ ratios directly from the $\gamma$-spectra after irradiation in a reactor. The isotopic analysis of relatively large uranium samples without activation by $\gamma$-spectrometry with high resolution detectors to within $1-2\%$ has, however, been demonstrated. This paper describes our investigations into the feasibility and scope of measuring the intensities of resolved $\gamma$-peaks of $^{239}\text{Np}$ relative to the intensity of a high yield fission product, as a rapid means of determining $^{235}\text{U}/^{238}\text{U}$ ratios in small uranium samples (a few $\mu$g) by neutron activation analysis. Hitherto, the low $\gamma$-energy spectrum from about 100 keV to 400 keV, of interest in this respect, could not be adequately studied as a result of the poor resolution of scintillation spectrometers.

**Experimental procedure**

For the evaluation of the method, uranium stock solutions of various $^{235}\text{U}/^{238}\text{U}$ ratios were prepared. The isotopic ratios varied between 0.00726 (natural U) and 1.095, and were determined mass spectrometrically with a precision of better than 0.5%. Samples from these solutions, containing approximately 30 $\mu$g of uranium, were evaporated to dryness in separate quartz ampoules, sealed and irradiated simultaneously for about 10 minutes in a thermal neutron flux of $\sim 3 \times 10^{13}$ n cm$^{-2}$ sec$^{-1}$ in the hydraulic rabbit of *Safari 1*, an ORR-type reactor. The Wescott's epithermal index, $r$, for this irradiation position is 0.0472, and the fast neutron flux ($> 0.1$ MeV) beyond 8% of the total flux.

$\gamma$-Spectrometry of the irradiated sample ampoules was done between 2 to 3 days after irradiation. The ampoules were placed 1 cm below the Ge(Li) detector, except where the effect of distance from the detector on the $\gamma$-spectrometric peak ratios was investigated. Counting times were usually 10 minutes. The detector used was a 5-cm$^3$ coaxial Ge(Li) diode (Princeton Gamma Tech, Princeton, N. J., U.S.A.) mounted in a cryostat and cooled with liquid nitrogen. An uncooled TC 130 Tennelec preamplifier, a TC 200 Tennelec amplifier and a Ridl bias amplifier (only the cut amplifier section), were used to obtain low noise amplification of the diode signals. The spectrum analysis was done on an Intertechnique 400 channel analyser (Model SA 40B) bypassing the built-in linear amplifier of the analyser. The resolution of the system is 4.2 keV (f.w.h.m.) for the 662 keV photopeak of $^{137}\text{mBa}$. The graphical analysis method reported by us was used to obtain the true peak counts under the photopeaks of interest.