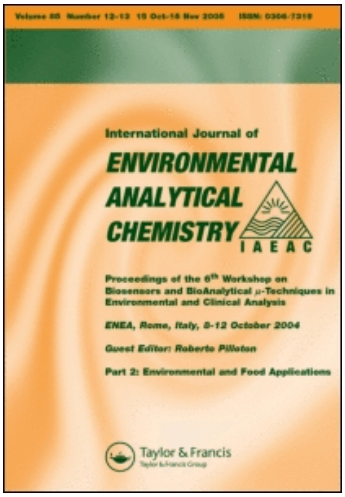


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# Epithermal Neutron Activation Analysis of Uranium Using the 106.1 keV Peak of Neptunium-239

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# Epithermal Neutron Activation Analysis of Uranium Using the 106.1 keV Peak of Neptunium-239

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The possibility of the most intensive gamma-peak of $Np^{239}$ (106.1 keV) for epithermal neutron activation analysis (ENAA) of uranium is evaluated. A procedure for the peak area calculation of the partially overlapping peaks of $Sm^{153}$ and $Np^{239}$ is discussed. It proved to be suitable for determination of uranium in phosphate ore samples by the 106.1 keV peak of $Np^{239}$ after epithermal irradiation using only one standard. The accuracy of the uranium determination is about 95% on average for the applied method.

## INTRODUCTION

One of the ways for epithermal activation analysis of uranium is by gamma-spectroscopy (106.1 keV peak) of $Np^{239}$. Practically the only interference is the 103.2 keV of the nuclide $Sm^{153}$.[1-8] To eliminate this bias, Kuleff and Kostadinov[9] proposed a method using only the interference-free half of the overlapping gamma-peaks, see Figure 1(b), which shows an amplified part of the gamma-spectrum of the

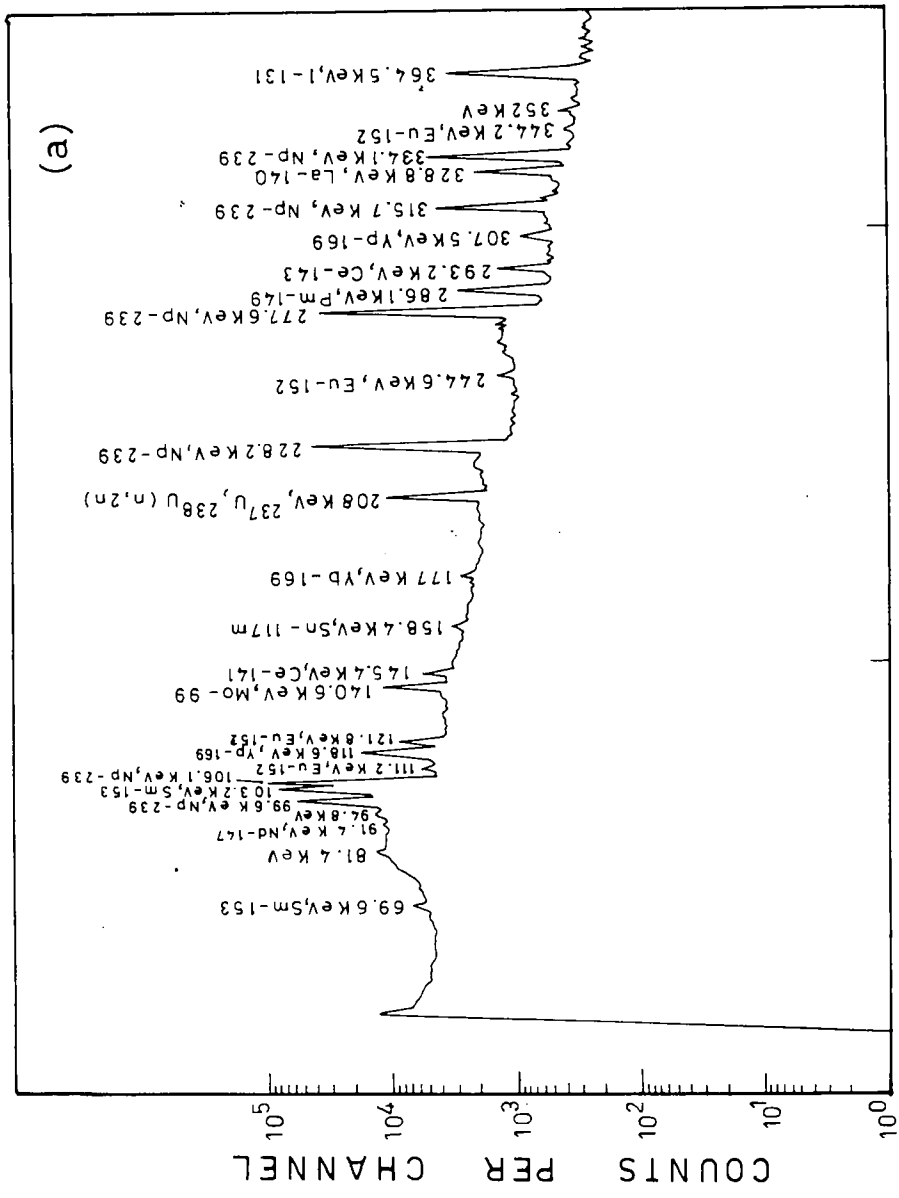
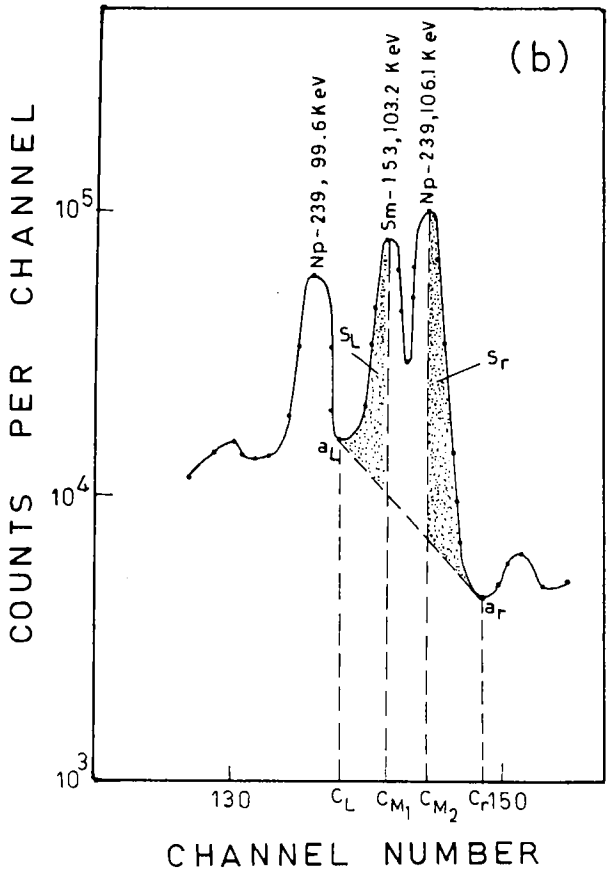



**Figure 1** (a) Gamma-ray spectrum of epithermal neutron irradiation of uranium compound ($U_3O_8$). Cooling time,




(b) Scheme of the method of half peak area.

uranium compound ($U_3O_8$) containing samarium and uranium, recorded with a Ge(Li) detector.

The following expressions hold:

The half of the total peak area of the higher energy 106.1 keV peak (right peak) is:

$$S_r = \sum_{i=C_{m2}}^{i=C_r} a_i - \frac{C_r - C_{m2} + 1}{2}\left[2a_r + \frac{(a_L - a_r)(C_r - C_{m2})}{(C_r - C_L)}\right] \qquad (1)$$

and the area of the left half of the lower energy 103.2 keV peak (left peak) is:

$$S_L = \sum_{i=C_L}^{i=C_{m1}} a_i - \frac{C_{m1} - C_L + 1}{2} \left[ 2a_L + \frac{(a_r - a_L)(C_{m1} - C_L)}{(C_r - C_L)} \right. \qquad (2)$$

where

$a_i$ = counts in the $i$th channel;
$a_L$ = counts in the outermost left channel of the left peak;
$a_r$ = counts in the outermost right channel of the right peak;
$C_L$ = number of the outermost left channel of the left peak;
$C_r$ = number of the outermost right channel of the right peak;
$C_{m1}$ = number of the channel of the maximum of the left peak;
$C_{m2}$ = number of channel of the maximum of the right peak.

All parameters in $S_r$ and $S_L$ equations may be determined from the numerical record of gamma-spectrum of the analyzed sample.

The proposed equations may be used for area calculation of any partially overlapping peaks such as $^{82}$Br-$^{76}$As, $^{76}$As-$^{122}$Sb and $^{65}$Zn-$^{46}$Sc. The use of epithermal neutron activation analysis (ENAA) to reduce the influence of interfering radionuclides by eliminating the thermal part of the reactor neutron spectrum has been discussed by many authors. The application in ENAA of a monostandard and high resolution Ge(Li) detectors for analysis of multielement samples is discussed by Zaghloul *et al.*[10] In this method the specific photopeak count-rates of the isotopes investigated in the sample are compared with that of a single comparator specified with relatively high resonance integral value ($I_0$), both measured under identical experimental conditions. Their ratio is defined as the $K$-value. The sensitivity and accuracy are improved for many trace elements and led to frequent use of this method in determining the trace elements in geological and biological materials.[11-23]

Quantitative determination of uranium in its ores through its daughter product $Np^{239}$ can be done by measurement of either the most intensive 106.1 keV gamma peak or the interference-free 277.6 keV peak. The drawback of the 106.1 keV peak is its interference with the 103.2 keV peak of $Sm^{153}$. The resolution of overlapping these two peaks is possible in good accuracy using the half peak area method (HPA) by applying Eqs. (1) and (2). To test

the utility and accuracy of this method, the net gamma-peak area of the same isotope ($Np^{239}$) through its well resolved gamma line of 277.6 keV is also estimated from the computerized peak area (CPA) using the PDP-11/34 digital computer connected with the measuring system and utilizing the programme "SPEKTRAN-F". This programme[28] automatically searches for significant peaks from the digital data of the spectra stored on the disk and calculates the net area of the peaks by subtraction from an artificial baseline under the peaks, which represents the natural background and the compton continue from the more energetic peaks on the spectra.

An aliquot of 23.52 mg $U_3O_8$ (22.04 mg U) was analysed by ENAA using the half peak area procedure (PHA) via the 277.6 keV and 106.1 keV lines of the irradiated $U_3O_8$ compound standard sample. Results were 21.88 mg and 18.624 mg.

Results from the analysis of four phosphate samples are given in Table I, data were obtained by HPA and CPA. For comparison, the data for samarium in the same samples are shown. For uranium HPA gives more accurate results, while for samarium the agreement is within statistical errors.

The accuracy of determining each element depends obviously on uncertainties of the nuclear data involved and hence differs from element to element. The values determined during the last ten years are accurate enough to be applied for the monostandard method.[24,25]

The weight of the interested unknown element, $m$, present in the irradiated sample can be calculated from

$$m = \frac{m^* I_0^* \alpha^*}{A^*} \cdot \frac{A}{I_0 \alpha} = K \cdot \frac{A}{I_0 \alpha} \tag{3}$$

where a monostandard nuclide is indicated with *.

Gold has been chosen as a single comparator due to its relatively high resonance integral value ($I_0 = 400$ barn) and to obtain a large epithermal activation in a short irradiation.

## EXPERIMENTAL

For epithermal neutron activation analysis, four phosphate samples of different weights and uranium compound ($U_3O_8$), as a standard

for checking the applied method, were sealed in clean small polyethylene vials and wrapped together with the monostandard of Au (for flux monitor) in aluminium sheet and then put into an aluminium can under Cd-cover for irradiation.

Corresponding blank aluminium foil is also irradiated for background estimation. Irradiation was carried out for one hour in the Triga II reactor of the Institute of Nuclear Medicine of DKFZ, Heidelberg, with a steady state power of 250 kW at an epithermal neutron flux, $\phi_{ep}$, of $1.56 \times 10^{11}$ n.cm$^{-2}$ sec$^{-1}$.

After irradiation the samples and standard were allowed to cool for different times to permit the decay of undesirable short-lived isotopes to the point where it no longer interfered.

Gamma-ray spectra of the cooled samples and standard were measured for 3000 sec with a Ge(Li) detector of volume 34 cc with a 1.9 keV resolution for 1.332 meV $\gamma$-peak of $^{60}$Co, coupled to a 4096 multichannel analyzer. All the spectra measured were recorded on a disk and processed on the PDP-11/34 computer connected to the measuring system. Subsequently the processed data are printed out and the spectra are plotted using Hewlett Packard printer and X-Y recorder.

The efficiency values of the detector for each gamma-ray energy up to 2000 keV for 10 cm and 40 cm sample-to-detector geometries have been previously estimated.[26]

Figure 1(a) shows the gamma-spectra of the standard uranium compound ($U_3O_8$) measured after about 8 d decay time while Figure 2 shows the gamma-spectra of four typical phosphate samples measured at different decay times. The energy range up to 400 keV covering the interested gamma-energy regions for uranium analysis. Figure 3 shows the amplified parts of overlapping gamma-lines 103.2 and 106.1 keV of $Sm^{153}$ and $Np^{239}$ respectively for the phosphate samples. For calculations using Eq. (3) to determine the concentration of uranium present in the irradiated samples $I_0$ values were taken from the literature,[24,27] and the results are tabulated in Table I.

## RESULTS AND DISCUSSION

The constant $K$ for the monostandard Au appears in Eq. (3) is equal to $6.406 \times 10^{-12}$ for the present irradiation condition.

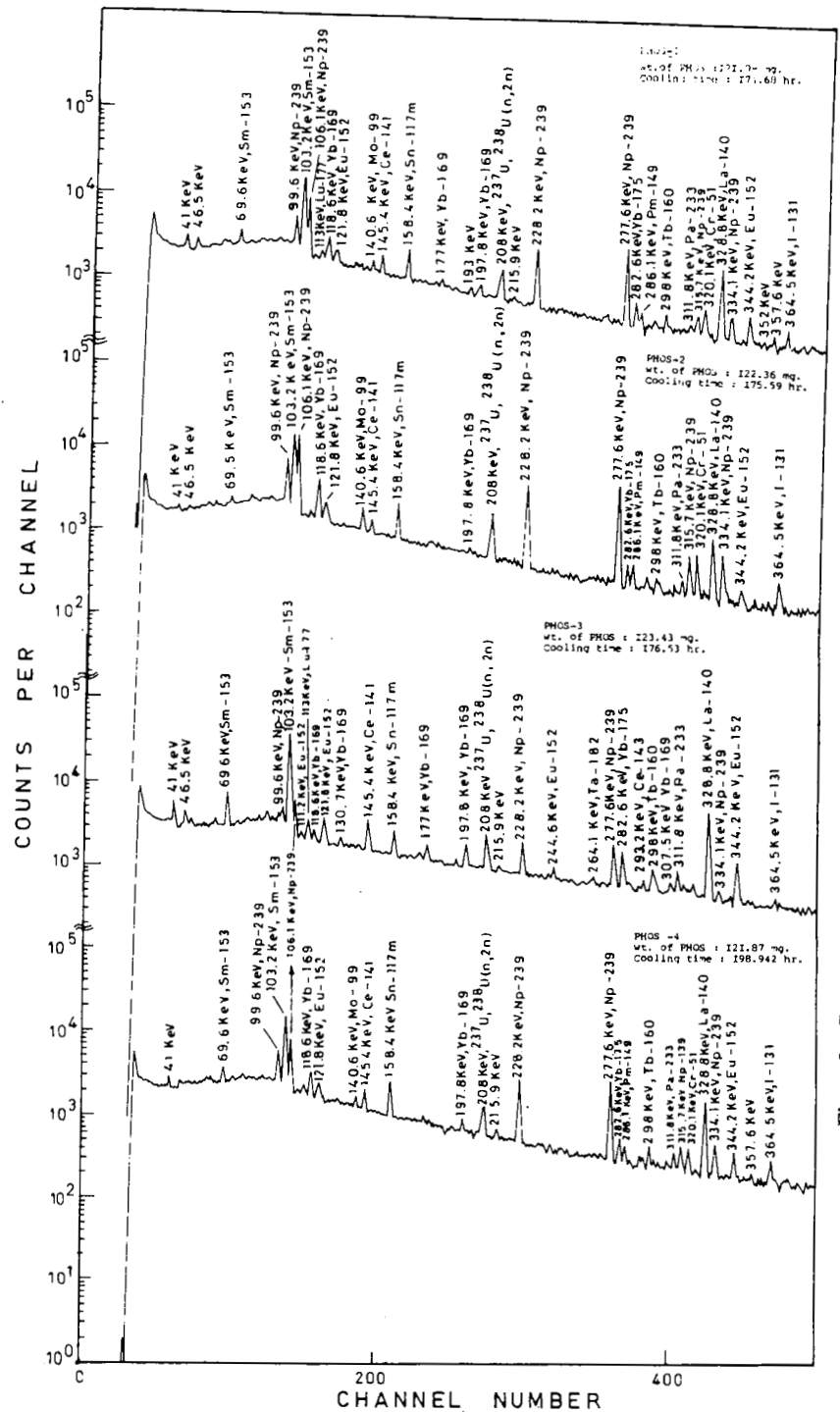



**Figure 2** Gamma-ray spectrum of epithermal neutron irradiation of phosphate samples.

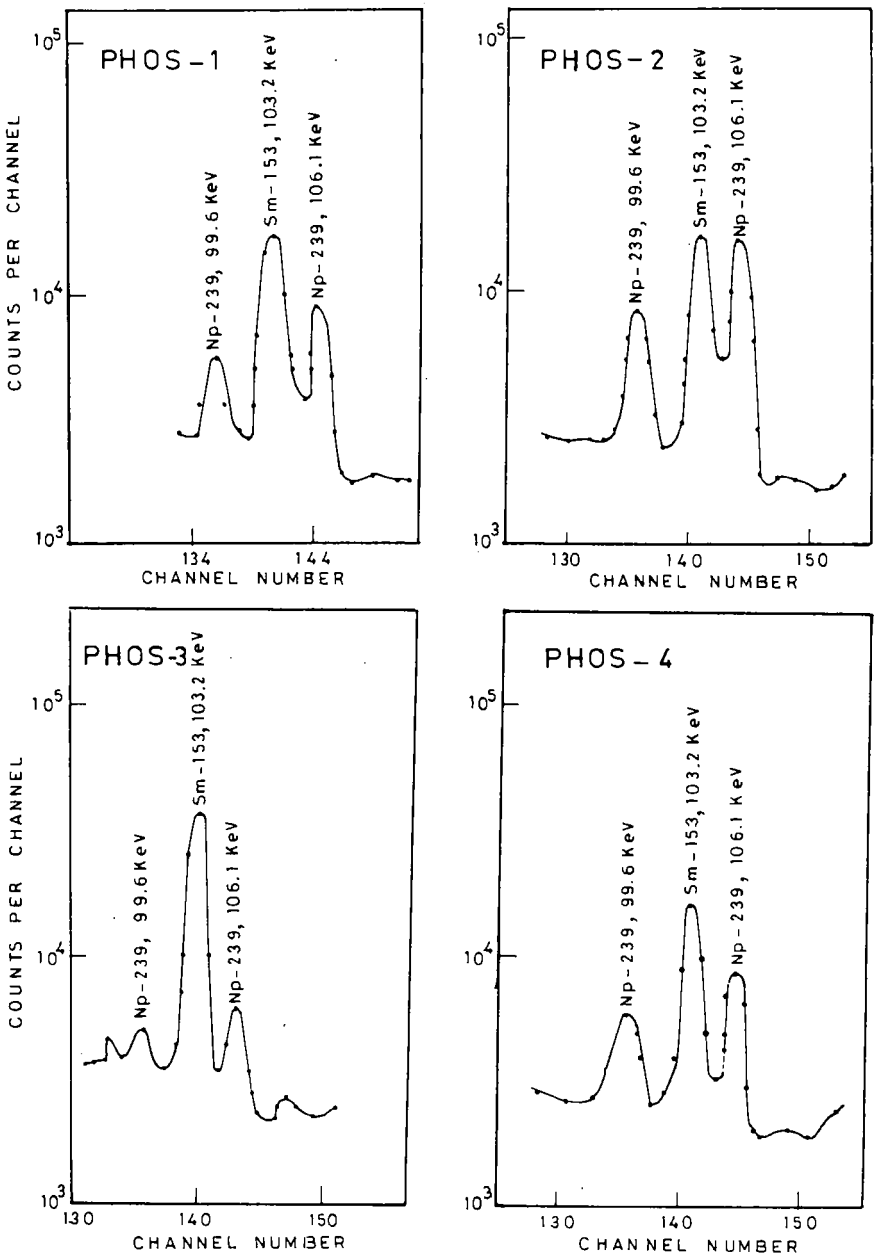



**Figure 3** The amplified parts of gamma spectrum of the samples containing samarium and uranium.

**Table I** Concentration of uranium and samarium in phosphate samples

| Sample | Concentration of uranium (ppm) | | | Concentration of samarium (103.2 keV) ppm | | |
|---|---|---|---|---|---|---|
| | Present work | | Average conc. from ref. (29) of 277.6 keV | Present work | | Average conc. from ref. (29) |
| | HPA 106.1 keV | CPA 277.6 keV | | HPA | CPA | |
| PHOS-1 | 158.7 | 140.5 | 151.3 | 84.2 | 83.0 | 74.4 |
| PHOS-2 | 299.2 | 266.3 | 284.7 | 68.1 | 66.7 | 70.4 |
| PHOS-3 | 79.6 | 77.6 | 79.0 | 228.8 | 228.0 | 154.0 |
| PHOS-4 | 212.9 | 189.9 | 201.0 | 108.2 | 97.3 | 70.1 |



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