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OPTIMUM TIMING PARAMETERS IN THERMAL NEUTRON ACTIVATION ANALYSIS OF URANIUM USING DELAYED NEUTRON COUNTING

F. ÖZEK - A.Z. ORTAOVALI

Ankara Nuclear Research and Training Center,
Beşevler - Ankara

ABSTRACT

A theoretical treatment of cyclic activation analysis of uranium employing a thermal neutron source and delayed neutron counting is given. Optimum timing parameters for the maximum detector response are obtained. Cycle optimization for 95 % and 90 % of the maximum detector response is also examined. Estimated detection limits based on the realistic values of the thermal neutron flux and detection efficiency are presented.

ÖZET

Uranyumun gecikmeli nötron sayımına dayanan tekrarlı termal nötron aktivasyon yöntemi ile analizi teorik olarak incelendi. Maksimum dedektör sayımını veren optimum zamanlama parametreleri elde edildi. Bu parametreler, maksimum dedektör sayımının % 95 ve % 90'ı için de araştırıldı. Son olarak, uranyumun adı geçen yöntem ile dedeksiyon limiti üzerinde hesaplamalar yapıldı.

INTRODUCTION

The detection of delayed neutrons from fission products following neutron activation has served as an attractive method for nondestructive assay of fissile materials, since interference does not occur, except oxygen. In the particular case of thermal neutron irradiated uranium, the neutron emission from oxygen does not arise as the threshold neutron energy for the $^{17}O(n,p)^{17}N$ reaction is about 8 MeV.

In neutron activation analysis (NAA) of the fissile materials, improved detection sensitivities can be obtained by cyclic activation (CA) of the sample and cumulatively counting the induced delayed neutrons /1-4/. In the present work, a theoretical study on optimum timing parameters in cyclic thermal NAA of uranium using delayed neutron counting is carried out.

CYCLIC ACTIVATION ANALYSIS OF FISSILE MATERIAL

Cyclic activation equation

In CA, a technique which is best suited for the study of short-lived activities /5/, the procedure is to irradiate the sample for a time (t_i), transfer it to the counting position in a time (t_w), count the induced activitiy (t_c), then return the sample to the irradiation position (t_r) for the next cycle. The cycle period is thus defined as

$$T = t_i + t_w + t_c + t_r \tag{1}$$

The detector response accumulated during the first cycle is:

$$D_1 = N \sigma_f (dn/F) \phi \varepsilon \cdot \sum_{j=1}^{k} \frac{a_j}{\lambda_j} (1-e^{-\lambda_j t_i})(e^{-\lambda_j t_w})(1-e^{-\lambda_j t_c}) \tag{2}$$

where N : number of fissionable nuclei in sample
σ_f : fission cross-section
dn/F : total yield of delayed neutrons per fission
ϕ : neutron flux
ε : detection efficiency
k : number of delayed neutron groups
a_j : relative yield of $_j$th group per fission
λ_j : decay constant of $_j$th group

The cumulative detector response accumulated at the end of the nth cycle is:

$$D_c = D_1 \sum_{j=1}^{k} \left[\frac{n}{1-e^{-\lambda_j T}} - \frac{e^{-\lambda_j T} (1-e^{-n\lambda_j T})}{(1-e^{-\lambda_j T})^2} \right] \tag{3}$$

Eq.3, according to our knowledge, was first constructed by Batchelor and Hyder /6/ in their study on delayed neutron energy spectra. The variation of D_c with n, for a total experiment time t_t = 5 min and t_w=t_r=0.5 s is plotted in Fig.1, using the data given in Table-1. In this particular case, the maximum detector response D_o occurs at the optimum number of cycles n_o=28 which corresponds to the optimum cycle period T_o≈11 s.

For the greater detector response, the longer experiment time, the greater number of cycles and the shorter cycle periods are required, as the total experiment time, t_t, increases. In this study, the values of n_o (or T_o) for a wide range (in practical terms) of t_t and t_w were obtained. The findings indicate that the optimum cycle period T is independent of t_t for t_t<30 min and increases linearly with the increasing sample transfer time (t_w, t_r). The results

are shown in Fig.2, with $t_w=t_r$.

TABLE-1. Delayed neutron properties for thermal fission of U-235 /7/.

Group index j	Half-life (s)	Relative yield a_j
1	55.72	0.033
2	22.72	0.219
3	6.22	0.196
4	2.30	0.395
5	0.61	0.115
6	0.23	0.042

dn/F = 0.0158 /8/
σ_f = 577 b /9/

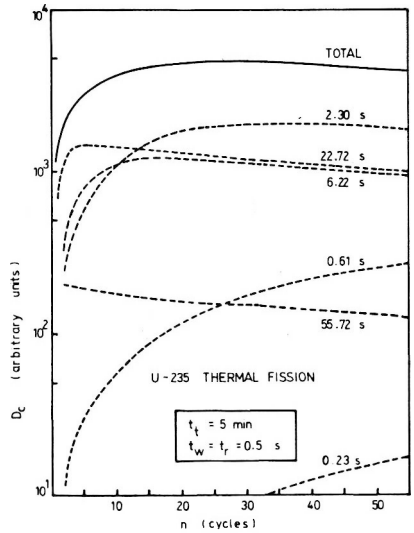


Fig. 1. Variation of D_c with n including individual group responses

In practice, the problems such as the consumption of pressurized gas, the deformation of sample tubes, etc. are often encountered with pneumatic fast transfer systems and it is therefore desirable to employ smaller cycle numbers, i.e. longer cycle periods, as long as the loss in counts is not significant /10/. Fig. 2 also includes T_o values corresponding to 95 % and 90 % of the maximum detector response, D_o.

Comparison of cyclic and conventional activation analysis

Fig. 3 shows the variation of detector response as a

function of total experiment time for cyclic, Eq.3, and single-cycle conventional activation, Eq.2. The superiority of cyclic activation to conventional case is clearly displayed. For D (cyc)>D (con), though, the cyclic t_t must be longer than about 3 min. Using the data given in Fig. 3, D (cyc)/D (con) ratios for various transfer and total experiment times were obtained and presented in Fig. 4. The D (cyc)/D (con) ratio can be regarded as the cyclic gain or advantage factor.

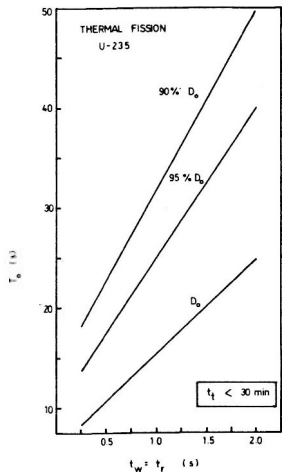


Fig. 2. Variation of optimum cycle period with sample transfer time

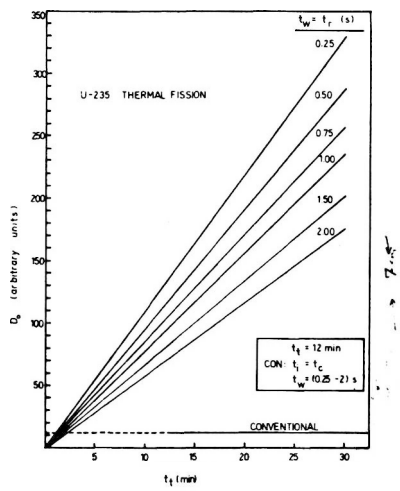


Fig. 3. Comparison of cyclic and conventional detector responses

Detection limits

The computed detection limits for CA analysis are plotted in Fig. 5. The detection limit here is defined as the ^{235}U mass to give a not delayed neutron count which is equal to three times the square root of the background count (BG). The conventional detection limit was estimated as about 1.3×10^{-7}g using the conventional timing conditions given in Fig. 3.

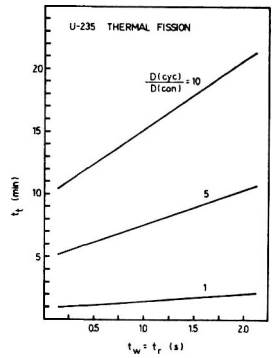


Fig. 4. Dependence of cyclic-to-conventional detector response ratio on total experiment and sample transfer times.

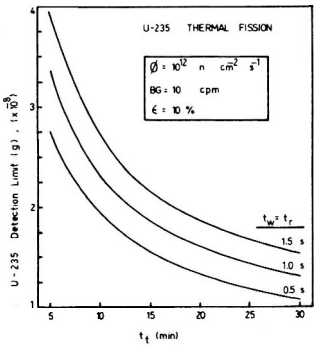


Fig. 5. Calculated detection limits for uranium.

CONCLUSION

Although the $^{238}U/^{235}U$ abundance ratio is about 10^2, the thermal neutron cross-section ratio σ_f ($^{235}U/^{238}U$) is greater than 10^6, /9/. It is for this reason that the delayed neutron emission from thermal neutron irradiated uranium has been assumed confidently to be due to ^{235}U fission only.

The computations indicate clearly that the cyclic detection limit improves with the increasing total experiment time (Fig. 5). Though, it is not expected to decrease

significantly for t_t>30 min. The cyclic-to-conventional detection limit ratio equals ten for t_t=20 min and t_w=t_r=0.5 s.

In practice, t_t can be selected from Fig. 4, after deciding on suitable D (cyc)/D (con) ratio. Then T_o, or n_o=t_t/T_o, can be obtained rapidly, using the data given in Fig. 2.

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