Quantification of Thorium and Uranium by β-delayed neutrons from photofission

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Abstract. Photo fission experiments have been conducted at the 10 MeV LINAC of EBC, Kharghar with Thoria and Natural Uranium samples. The samples were irradiated for 60 seconds duration in pure as well as in mixed form and delayed neutrons and gamma decay data was measured using Helium-3 detectors assembly and BGO scintillator respectively. The experiments conclude that with the 250 W beam power in scanned mode of 1.1 Hz frequency which ultimately deposits 20.2 W beam power on the fissile material samples, unshielded 100 mg of Thorium and natural uranium can be detected with good accuracy. The relative proportions of the Thorium and Uranium was determined within 7 % accuracy by analyzing the delayed neutron decay signature.

Keywords: Photofission, radioactive waste, delayed neutron, delayed gamma, decay curve.
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INTRODUCTION

Advanced instrumentation for non-destructive assay (NDA) of fissile and fertile materials is required in the reprocessing plants for safety, safeguards, accountability and process control. Active interrogation, a measurement technique which uses a radiation source (Photon/Neutron) to probe materials and generate unique signatures is a powerful tool for assaying highly shielded fissile and fertile materials. Photons with energies above 6 MeV can be used to induce photofission in fissile materials. The method involves usage of intense beam of high-energy Bremsstrahlung photons to induce photo fission in actinides and later measure the delayed neutron/gamma decay counts. The measurement of delayed neutrons and delayed gammas together can overcome limitations due to heavy shielding and can bring specific information on localization, quantification of the fissile material and identifying and finding out relative proportions for different types of fissile materials in mixed state [1]. The photon interrogation technique has some added benefits over the neutron interrogation technique. Even though the cross section of photo fission in fissile materials is far lower than the thermal neutron fission cross sections, the lower cross section value is easily compensated because of high intensity of photon beam. The shielding can cause attenuation of the neutron beam, whereas high energy photons have good penetration property. The photon beam can be easily collimated for interrogation applications. The interaction cross-sections for the high energy photons with the benign materials are very small as compared to those with the fissile materials; therefore background activation is negligible. In the present paper the mass of Thorium and natural Uranium have been determined in pure as well as in mixed state by measuring time dependent delayed neutrons and photons after photofission.

EXPERIMENTAL SET-UP

Fig. 1 gives the picture of the experimental set-up. In the experiments the electron beam used was of 10 MeV energy, 50 mA peak current, 10 µs pulse width, 50 Hz pulse frequency and 1.1 Hz scan frequency. The electron beam is made to impinge on 2 mm thick tantalum target and bremsstrahlum is generated. Fig. 2 gives the photon energy spectrum generated and photofission cross section data of U-238 and Th-232 [2,3]. Hermitically sealed natural uranium samples and Thoria samples fabricated by AFD, BARC were used for the experiments. The samples were placed below the converter at the geometric centre of the scan horn and the geometric centre of the neutron detection system. The neutron detection system consists of 6 nos. of Helium-3 proportional counters of 250 cps/nv sensitivity each surrounded by 25 mm thick High Density Polyethylene moderator and placed under the beam in such a way that the beam scans axially the detectors. One no. of BGO scintillator is placed perpendicular to the beam scan as shown in the figure. The total duration of each irradiation was of 60 seconds. After the irradiation was stopped delayed neutron and delayed gamma decay data was acquired.

FIGURE 1. Picture of the photo fission experimental set up at EBC, Kharghar.

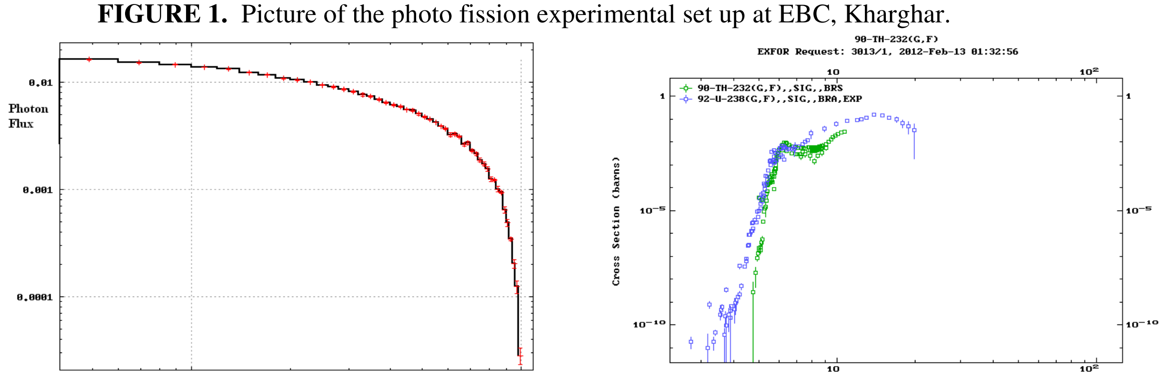

FIGURE 2. Bremsstrahlung photon flux incident on sample surface and photofission cross sections of Th-232 and U-238 samples.

TESTS AND RESULTS

The delayed neutron and delayed gamma measured are the function of the mass of the fissile material. Fig. 3 gives the delayed gamma decay spectra of the samples and fig. 4 gives the delayed neutron decay spectra of the samples. By measuring the count rate at a given time ***t*** the mass of the sample can be estimated from following Eq. 1 [4]. The equation includes the corrections for the pulsed and scanning mode of the irradiation beam.

$$
\begin{aligned}
C(t) &= P\sum_{i=1}^{N} A_i \exp(-\lambda_i t) 1 - \exp(-\lambda_i \tau) \times T_{pulse} \times T_{cyc} \\
T_{pulse} &= \frac{T_{irr}}{n\tau} \frac{1-\exp(-n\lambda_i T_{period})}{1-\exp(-\lambda_i T_{period})} \\
T_{cyc} &= \left(\frac{1}{1-\exp\left(-\lambda_i T_{cycle}\right)}\right)\left(1-\frac{1-\exp(-\lambda_i T_{cycle} N_{cycle})}{N_{cycle}(\exp(-\lambda_i T_{cycle})-1)}\right)
\end{aligned}
\tag{1}
$$

Where, *C(t)* is the number of counts at given time *t*, *P* is production rate equal to $N\varphi\nu\varepsilon$ where *N* is the no. of fissile material atoms, φ is the photofission rate, ν is the rate of delayed neutron or delayed gamma emission per fission, ε is the efficiency of the detection system, A_i is the weight factor of decay group, λ_i is the decay constant of decay group, τ is the pulse width, *n* is the number of pulses falling on the sample, T_{irr} is the total time of irradiation per scan, T_{period} is the pulse period, N_{cycle} is the no. of times the sample is exposed to the beam while scanning, T_{cycle} is the period of scan on the sample. The function of the Eq. 1 is used to fit the delayed neutron decay curves (fig. 4) in ROOT minute and using the published A_i, $T_{½}$ and ν values as listed in Table 1 [5], in the fit the $T_{½}$ was kept fixed and other parameters were used as initializing parameters. As the samples were irradiated for fixed 60 seconds duration and data was acquired with 1 second time bin width, the curves could be fitted for only the first four decay groups and decay half < 1 second could not be extracted. The efficiency of neutron detection system was experimentally determined as using Cf-252 source (0.1) and φ was determined from the intensity of photon flux and the photofission cross sections ($7.2\text{x}10^8$Photons cm^{-1} barns for U-238). *N* of U-238 exposed to photon beam equals to $6.42\ \text{x}10^{22}$. Using these values and ν equals to 0.02 for U-238 the production rate for U-238 is estimated as $9.25\ \text{x}10^4$. Table 2 gives the fitted values of the production rate and weight factors of four decay groups. The fit value of production rate for U-238 is $9.36\text{x}10^4$ which is close to the estimated value. The mass ratio of the uranium and thorium contributing in photofission equals to 1.78 and the ratio of the production rates obtained from the fit equals to 1.83 which is within 3 %. The fitted values from the table 2 were again used to fit the decay curve of mixed sample (fig. 5). The ratio of the two production rates are 1.91 which are again very close to the pure sample ratio (within 7.3%). A decay curve was simulated from the theoretically obtained fit values and was plotted along with the experimental decay curve (fig. 5) and it is observed that both the curves superimpose each other very well. In future experiments will be conducted with improved counting statistics to reduce errors on the parameters determined.

TABLE 1. Weight factor and decay time constant of Th-232 and U-238 nuclei for photo fission delayed neutrons from ref 1.

Group number	Th-232			U-238		
	Ai (%)	$T_{1/2}$	ν/100 fissions	Ai (%)	$T_{1/2}$	ν/100 fissions
1	4.4±0.2	55.6±1.5	0.17	1.98±0.08	56.2±0.8	0.061
2	16.3±1.0	20.3±0.8	0.62	15.7±0.5	21.3±0.3	0.489
3	15.9±1.5	5.45±0.5	0.6	17.5±0.7	5.5±0.2	0.545
4	37.5±3.0	1.98±0.2	1.43	31.1±0.8	2.15±0.1	0.970
5	17.2±2.0	0.43±0.1	0.66	17.7±0.9	0.7±0.06	0.552
6	8.7 +2.0-4.0	0.18±0.03	0.33	16.1+2.0-5.0	0.19±0.02	0 .502

TABLE 2. Experimentally determined parameters of Th-232 and U-238 nuclei for photo fission delayed neutrons.

Group number	Th-232	U-238
	Ai	Ai (%)
1	11.4±0.7	3.33±0.17
2	36.5±5	22.7±0.37
3	20.3±1.8	11.2±1.5
4	22.9±9	18.6±2.6
Production rate pure sample	5.069E04±523	9.361E04±603
Production rate mix sample	4.9E04±1858	9.4E04±555

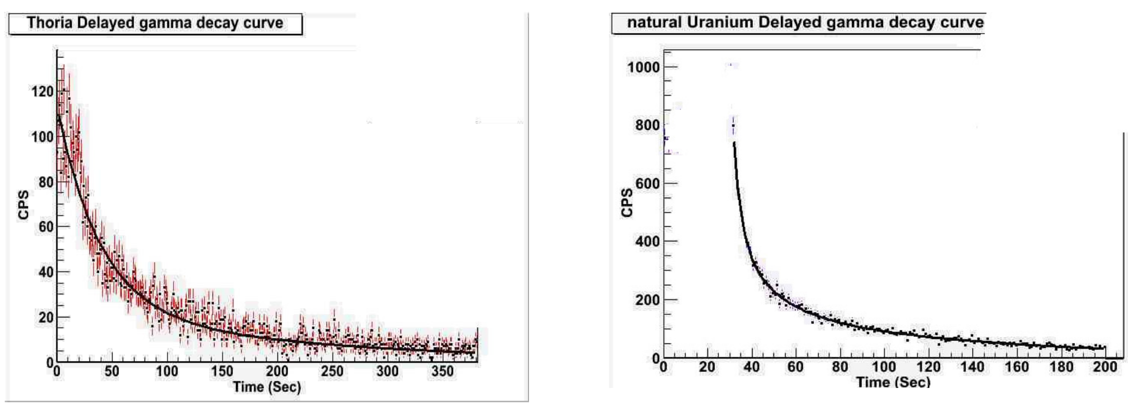

FIGURE 3 Photofission delayed gamma decay curve of Thoria and Nat. U

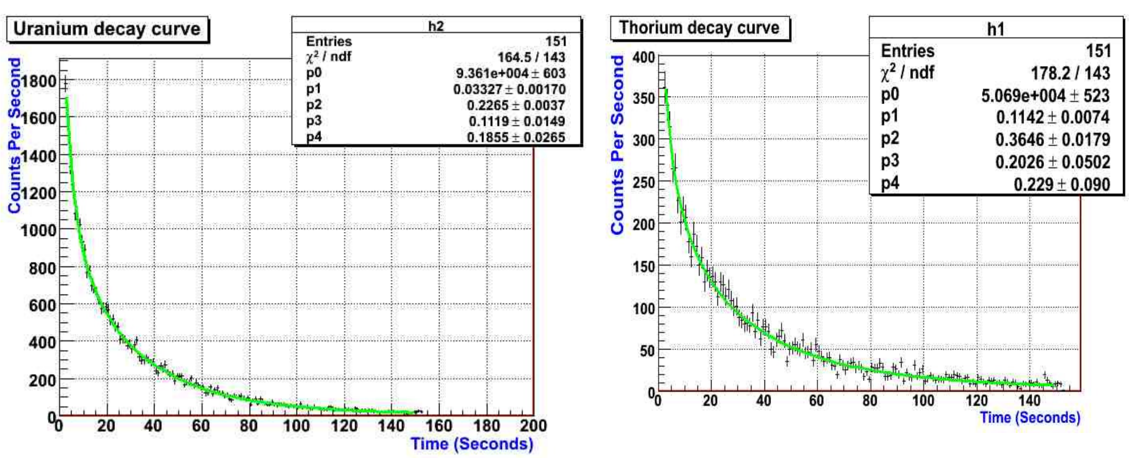

FIGURE 4 Photofission delayed neutron decay curves of Thoria and Nat U with fit

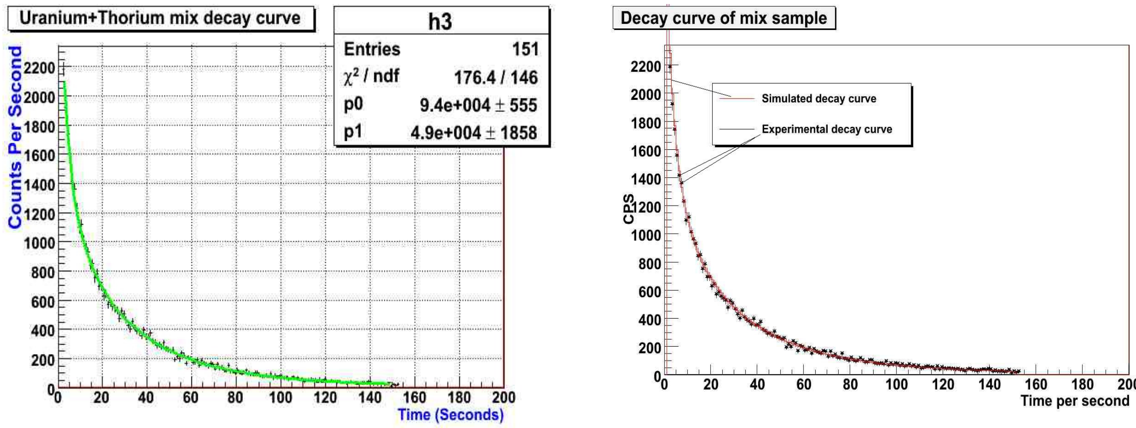

FIGURE 5 Experimental and simulated delayed neutron decay curve of Th-232 and Nat U mixed sample

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