Quantification of fissile materials by photon activation method in a highly shielded enclosure


Contact: pmdighe@barc.gov.in
Motivation

• Quantitative evaluation of Special Nuclear Materials
• Non destructive assessment of dense and large nuclear waste drums
• Localization of nuclear material in waste packages
• Development of photonuclear libraries
• Security applications
Non-destructive Passive Methods of quantifications

• X-ray tomography
• Passive gross neutron measurements
• Segmented gamma scanning of waste drums
• Gamma spectroscopy with HPGe detectors
• Hybrid K-edge Densitometer
• High sensitivity calorimeters
Non-destructive Active Methods of quantifications

• Neutron induced activation analysis
  • Pulsed neutron sources
  • LINAC based neutron sources
• Photon induced activation analysis
  • Bremsstrahlung generation from LINAC
  • Gamma sources
Method of photo-fission

Photofission principle

$e^{-}$

Bremsstrahlung

Actinide

$\gamma$

Excited Nucleus (GDR)

Actinide*

Fission

$\gamma$

$n\uparrow$

FP

FP
Method of photo-fission

The fragments attained 90% K.E.

Excitation of nucleus

Emission of prompt neutrons

Emission of prompt Gammas

Stopping of fragments

Contact: pmdighe@barc.gov.in
Bremsstrahlung spectrum > 4 MeV photon energy generated by 10 MeV electron beam

Photo fission cross sections of U-238
Aim of experiments

• Explore the possibility of use of 10 MeV electron LINAC for non-destructive quantitative identification of fissile materials
• Establish the mass v/s count rate relationship for delayed neutron & gammas for quantification of unknown mass
• Estimate the decay parameters of various actinides for 10 MeV end point energy
Experimental set-up at 10 MeV LINAC
Radiation detectors used

- A set of six numbers of High sensitivity Helium-3 proportional counters housed in a 25mm thick HDPE box for delayed neutron measurements
- 2 Nos. of 76 mm diameter BGO scintillators for delayed gamma measurements
- REM meter for neutron dose measurement
- Scintillator based gamma dosimeter
Accelerator Beam parameters

Electron beam parameters
• Energy: 10 MeV
• Current: 50 mA
• Pulse width: 10 µs
• Pulse Frequency: 50 Hz
• Scan frequency 1.1 Hz

Natural Uranium Samples

Four natural uranium plates (100 mm length x 50 mm width) of 35gm, 70gm, 145gm and 160gm weights. To avoid emission of radioactive gas after photo fission the samples are sealed inside high purity aluminium (1S grade) box.

Dose measured on the samples for 60 seconds irradiation time and 600 seconds cooling time
For 160 gm sample: 0.05 mSv/h in contact with the sample.
The experiments

• The natural uranium samples were positioned at the geometric centre of the neutron moderator assembly.

• The BGOs were placed outside the helium-3 detector assembly with axes perpendicular to the accelerator beam.

• The samples were irradiated for 60 seconds duration.

• Delayed neutron and delayed gammas were measured after the accelerator beam was switched OFF.

• Dose on the sample measured after 10 m cooling.
Delayed gamma energy spectrum

Contact: pmdighe@barc.gov.in
Delayed neutron decay curves
Delayed gamma decay curve

Counts Per Second

Time in second

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Mass V/s count rate

Counts Per Second

Delayed gamma

Delayed neutron

Natural Uranium sample (Gram)
Data analysis

The count rate measured at a given time is sum of a number of exponentials contributed by various fission fragments decaying with varying half lives

\[ C(t) = N \varphi \nu \varepsilon \sum_{i=1}^{N} A_i \exp \left( -\lambda_i t \right) T_{\text{const}}^i T_{\text{pulse}}^i T_{\text{cyc}}^i \]

\( C(t) \) is the count rate measured at a given time \( t \), \( N \) is the no. of uranium atoms, \( \varphi \) is the photo fission rate, \( \nu \) is the rate of delayed neutron or delayed gamma emission per fission, \( \varepsilon \) is the efficiency of the detection system, \( A_i \) is the weight factor, \( \lambda_i \) is the decay constant, and \( T_{\text{const}} \), \( T_{\text{pulse}} \) and \( T_{\text{cyc}} \) are the corrections for the finite time irradiation, pulsed beam irradiation and scanned mode irradiation.
Data analysis

• The efficiency of the neutron detection system was calibrated using Cf-252 and Am-Be neutron sources.

• The photo fission delayed neutron decay parameters for U-238 were taken from reference (Giacri-Mauborgne M. L., «Création d’une bibliothèque d’activation photo-nucléaire et mesures de spectres d’émission de neutrons retardés», Ph. D thesis l’université de Caen de Basse-Normandie : Physique, 198 p., 2005).

\[
\begin{array}{cc}
\text{Ai} & \text{T1/2} \\
0.017 & 55.6 \\
0.165 & 22.36 \\
0.183 & 5.43 \\
0.373 & 1.86 \\
0.18 & 0.47 \\
0.085 & 0.19
\end{array}
\]

\( \nu = 3.03 \% \)
Data analysis

• Estimated delayed neutron count rate is 7157 cps for 160 gm sample. Experimentally observed value is 6358 cps.

• 10 MeV LINACs with 250 W beam power, unshielded 100 mg of natural uranium can be detected with good counting statistics

• The mass count rate linearity is within 5 %
Delayed gamma decay group formation for
10 MeV end point energy

\[ f(t) = M \sum_{i=1}^{N} A_i \exp(-\lambda_i t) T_{const}^i T_{pulse}^i T_{cyc}^i \]

| \( A_i \) | 0.31 | 0.28 | 0.14 | 0.16 | 0.10 |
| \( (ln(2)/\lambda_i) = T_{1/2} \text{ (sec)} \) | 0.73 | 4.66 | 25.02 | 96.27 | 1155.3 |

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Effect of shielding for neutron radiation

![Graph showing the transmitted fraction per incident neutron against thickness (cm) for Polyethylene and Water.

Contact: pmdighe@barc.gov.in]
Effect of shielding for gamma radiation

% Transmission

Thickness of Iron shield (cm)

- 10 MeV
- 5 MeV
- 2 MeV
- 1 MeV
- 0.5 MeV
- 0.2 MeV
- 0.1 MeV
Conclusions

Through these experiments we have established that the 10 MeV LINACs, which are conventionally used for industrial applications can be used for non-destructive assay of fissile materials. Even though the photo fission cross sections at 10 MeV end point energy are considerably low, the higher beam intensity compensates the loss and measurable signal is obtained during irradiations.

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Contact: pmdighe@barc.gov.in
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