Mass Yield Distribution in the Photon-induced Fission of $^{232}$Th, $^{238}$U, $^{nat}$Pb, and $^{209}$Bi


$^1$Radiochemistry Division, Bhabha Atomic Research Centre, Mumbai-400085, India.
$^2$Department of Physics, Kyungpook National University, Daegu 702-701 Republic of Korea
$^3$Applied and Pulse Power Division, Bhabha Atomic Research Centre, Mumbai-400085, India.
$^4$Division of Advanced Nuclear Engineering, Pohang University of Science and Technology, Pohang 790-784, Republic of Korea

22nd International Seminar on Interaction of Neutrons with Nuclei
Dubna, Russia, May 27-30, 2014
We determined the yields of fission products in the photon-induced fission for $^{232}\text{Th}$ and $^{238}\text{U}$ with the end-point energy of 10 MeV at the electron linac of Electron Beam Center in Kharghar, Navi Mumbai, India and those for $^{\text{nat}}\text{Pb}$, $^{209}\text{Bi}$, and $^{232}\text{Th}$ with 45-80 MeV and 2.5 GeV at Pohang Accelerator Laboratory in Pohang, Korea. The peak-to-valley (P/V) ratio, average light mass ($<A_L>$), and heavy mass ($<A_H>$) at different excitation energy were obtained from the mass yield data of this work and literature data. The value of P/V, $<A_L>$, and $<A_H>$ in the $^{232}\text{Th}(\gamma, f)$ and $^{238}\text{U}(\gamma, f)$ are compared with the similar data in $^{232}\text{Th}(n, f)$, and $^{238}\text{U}(n, f)$ to examine the role of excitation energy on nuclear structure effects. Similarly, the present data in the $^{\text{nat}}\text{Pb}(\gamma, f)$ and $^{209}\text{Bi} (\gamma, f)$ were compared with the literature data at other bremsstrahlung energies to examine the role of excitation energy on average mass ($<A>$) and P/V ratio.
Studies on fission product yields (mass yield distribution) in the neutron- and photon-induced fission of pre-actinides ($^{\text{nat}}$Pb, $^{209}$Bi) and actinides ($^{232}$Th, $^{238}$U) are important for their application in ADSs [1, 2] fast reactor [3] and AHWR [4].

In ADSs, the heavy elements such as $^{\text{nat}}$Pb, $^{209}$Bi, $^{232}$Th, and $^{238}$U can be used as the spallation source to produce high energy neutrons by the bombardment of high (GeV) energy proton from the accelerator.

It is necessary to determine the yields of products in the high energy neutron/bremsstrahlung induced fission of the spallation targets and long-lived minor actinides, which are need for the design of the ADSs.

During the spallation process high energy neutrons and photon (bremsstrahlung) produces, which can cause photo-nuclear reaction and fission of the spallation source and long-lived minor actinides.
1. Pohang Neutron Facility based on electron linac

Pohang Accelerator Laboratory

Pohang Neutron Facility based on 100-MeV e-linac

Pohang High Energy Radiation Facility with 4.0 GeV e-linac
Experiment with Bremsstrahlung at PNF

-The yields of fission products in the 45-80 MeV and 2.5 GeV (γ, f) of natPb, 209Bi and 232Th by off-line γ-ray spectrometric technique and using the electron linac at PAL, Pohang, South Korea [9-15].

Operation Condition of electron linac:
• Electron energy: 45, 50, 60, 70, 80 MeV, 2.5 GeV
• Beam current: ~ 15 mA
• Beam width: 1.5 μs
• Repetition rate: 3.75 Hz

Bremsstrahlung Target:
• thin W foil (100 mm × 100 mm × 0.1 mm)
2. Facility in Electron Beam Center, Kharghar, Navi-Mumbai

The yields of fission products in the 10 MeV (γ, f) of $^{232}$Th and $^{238}$U using the electron linac at EBC, Kharghar, Navi-Mumbai, India [16].

<table>
<thead>
<tr>
<th>Facility</th>
<th>EBC, India</th>
</tr>
</thead>
<tbody>
<tr>
<td>W (Ta)foil size</td>
<td>100 cm²</td>
</tr>
<tr>
<td>W (Ta) thickness</td>
<td>1 mm</td>
</tr>
<tr>
<td>W(Ta) distance</td>
<td>3 cm</td>
</tr>
<tr>
<td>from exit window</td>
<td></td>
</tr>
<tr>
<td>Sample distance</td>
<td>10 cm</td>
</tr>
<tr>
<td>From W(Ta)</td>
<td></td>
</tr>
<tr>
<td>Electron energy</td>
<td>10 MeV</td>
</tr>
<tr>
<td>Beam current</td>
<td>50 mA</td>
</tr>
<tr>
<td>Pulse width</td>
<td>10 µs</td>
</tr>
<tr>
<td>Repetition rate</td>
<td>400 Hz</td>
</tr>
</tbody>
</table>
- The metal targets were wrapped with 0.025 mm thick Al foil and additionally with 0.018 mm Al foil.
- The sample was irradiated for 0.5-5 h for 45-80 MeV and 2.5 GeV. After each irradiation, the samples were cooled for 30 m to 2 h.

<table>
<thead>
<tr>
<th>Chemical Purity (%)</th>
<th>Weight (g)</th>
<th>Thickness (mm)</th>
<th>Size</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{232}$Th</td>
<td>99.999</td>
<td>0.2-0.3</td>
<td>0.025</td>
</tr>
<tr>
<td>natPb</td>
<td>&gt;99</td>
<td>12.417</td>
<td>0.5</td>
</tr>
<tr>
<td>$^{209}$Bi</td>
<td>&gt;99</td>
<td>74.417</td>
<td>3.0</td>
</tr>
<tr>
<td>Al</td>
<td>99.99</td>
<td>Natural</td>
<td>0.025 , 0.018</td>
</tr>
</tbody>
</table>
### Specification of Samples for 10 MeV

<table>
<thead>
<tr>
<th></th>
<th>Chemical Purity (%)</th>
<th>Weight (g)</th>
<th>Thickness (mm)</th>
<th>Size</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{232}$Th</td>
<td>99.999</td>
<td>0.2822</td>
<td>0.025</td>
<td>2.72 cm²</td>
</tr>
<tr>
<td>$^{238}$U</td>
<td>$&gt;$99</td>
<td>0.2414</td>
<td></td>
<td>2.5 cm²</td>
</tr>
<tr>
<td>Al</td>
<td>99.99</td>
<td>Natural</td>
<td>0.025, 0.018</td>
<td></td>
</tr>
</tbody>
</table>

The metal targets were wrapped with 0.025 mm thick Al foil and additionally with 0.018 mm Al foil.
- The sample was irradiated for 4-5 h and cooled for 0.5–1.5 h.
Low-background gamma-ray spectrometry

- Coaxial CANBERRA high-purity germanium (HPGe) of diameter 60.5 mm and length of 31 mm.

- The detection efficiency was 20% at 1332.5 keV relative to a 3” diameter × 3” length NaI(Tl)

Detection Efficiency:

$$\varepsilon(E_\gamma) = \frac{N(E)}{I_\gamma(\%) A_{ref} e^{-\lambda \cdot t_d}} \cdot \frac{1}{\varepsilon(1332.5 \text{ keV})}$$

Fifth order polynomial fitting:

$$\ln \varepsilon = \sum_{i=0}^{5} a_i (\ln E)^i$$

| Nuclide | Half life (Yrs.) | No. of $\gamma$-rays | $E_\gamma$(keV) | $I_\gamma$(%)
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{241}$Am</td>
<td>432.2</td>
<td>1</td>
<td>59.5412</td>
<td>35.9</td>
</tr>
<tr>
<td>$^{152}$Eu</td>
<td>13.537</td>
<td>11</td>
<td>121.78, 244.70, 344.28, 411.12, 443.97, 778.91, 867.38, 964.08, 1085.87, 1112.07, 1408.00</td>
<td>28.58, 7.58, 26.50, 2.23, 3.15, 12.94, 4.25, 14.61, 10.21, 13.64, 21.01</td>
</tr>
</tbody>
</table>
Typical $\gamma$-ray spectrum of fission products in the 70-MeV bremsstrahlung-induced reaction from $^{232}$Th
3. Data Analysis

➢ Determination of Average Excitation Energy :

\[
\langle E^* (E_e) \rangle = \frac{\int_0^{E_e} E_e N(E_e, E_\gamma) \sigma_F (E_\gamma) dE_\gamma}{\int_0^{E_e} N(E_e, E_\gamma) \sigma_F (E_\gamma) dE_\gamma}
\]

\[N(E_e, E_\gamma)\] The number of photons with an energy \(E_\gamma\) produced from the incident electron energy \(E_e\) : using GEANT4

\[\sigma_F (E_\gamma)\] The fission cross section as a function of the photon energy \(E_\gamma\)

Using the TALYS 1.4

<table>
<thead>
<tr>
<th>(E_\gamma) [MeV]</th>
<th>40</th>
<th>45</th>
<th>50</th>
<th>60</th>
<th>70</th>
<th>80</th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt;E*&gt; [MeV]</td>
<td>15.87</td>
<td>16.95</td>
<td>17.86</td>
<td>19.76</td>
<td>21.25</td>
<td>22.49</td>
</tr>
</tbody>
</table>
The number of photons with an energy $E_\gamma$ produced from the incident electron energy $E_e$ using GEANT4.
Determination of Yields for Fission Products

- From the observed number of γ-rays ($N_{obs}$) under the photo-peak of each individual fission product, their **cumulative yields** ($Y_R$) relative to $^{135}$I were determined by:

$$N_{obs}(CL/LT) = n \sigma_F(E) \Phi I_\gamma \varepsilon Y_R (1 - e^{-\lambda t_{irr}}) e^{-\lambda t_{cool}} (1 - e^{-\lambda CL}) / \lambda$$

where $n$ is the number of target atoms $\sigma_F(E)$ is the photo-fission cross-section of the target nuclei and $\Phi = \int_{E_b}^{E_e} \phi dE$ is the integrated photon flux from the reaction threshold ($E_b$) to the end-point energy ($E_e$) for the photon flux ($\phi$) at the photon energy $E$. The $t_{irr}$ and $t_{cool}$ are the irradiation and the cooling time, and $CL$ and $LT$ are the real and the live times of counting, respectively. $\lambda$ is the decay constant of the isotope of interest and $\varepsilon$ is the detection efficiency of the γ-rays in the detector system. $I_\gamma$ is the abundance or the branching intensity of the chosen γ-rays of the reaction products.

- From the relative **cumulative yields** ($Y_R$) of the fission products, their relative mass-chain yields ($Y_A$) were determined by:

$$Y_A = Y_R / FCY, \quad FCY = \frac{EOF^{a(Z)}}{\sqrt{2\pi}\sigma_z^2} \int_{-\infty}^{Z+0.5} \exp[-(Z - Z_P)^2 / 2\sigma_z^2] dZ$$

where FCY is the fractional cumulative yield, $Z_P$ is the most probable charge and $\sigma_z$ is the width parameter of an isobaric yield distribution. $EOF^{a(Z)}$ is the even-odd effect with $a(Z) = +1$ for even $Z$ nuclides and -1 for odd-$Z$ nuclides.
4. Result and Discussion

- Yields of Fission Products (%) as a function of mass number in the $\gamma$-induced fission of $^{nat}$Pb and $^{209}$Bi.
Yields of Fission Products (%) as a function of mass number in the γ-induced fission of $^{232}\text{Th}$ and $^{238}\text{U}$

The Fission Yield distribution in $^{232}\text{Th}(\gamma, f)$ reaction is triple humped, whereas that in the $^{238}\text{U}(\gamma, f)$ reaction is double humped. This is due to the different type of potential energy surface in $^{232}\text{Th}^*$ compared to $^{238}\text{U}^*$, which is called as the Th anomaly.
Average values of heavy mass \( \langle A_H \rangle \) and light mass \( \langle A_L \rangle \) as a function of excitation energy in the \( ^{232}\text{Th}(\gamma,f) \) and \( ^{232}\text{Th}(n,f) \) reactions as well as in the \( ^{238}\text{U}(\gamma,f) \) and \( ^{238}\text{U}(n,f) \) reactions.

\[
\langle A_L \rangle = \frac{\sum (Y_A A_L)}{\sum Y_A}, \quad \langle A_H \rangle = \frac{\sum (Y_A A_H)}{\sum Y_A},
\]
Fission Yields (%) of symmetric and asymmetric fission products in the $^{232}\text{Th}(\gamma,f)$ and $^{232}\text{Th}(n,f)$ reactions and in the $^{238}\text{U}(\gamma,f)$ and $^{238}\text{U}(n,f)$ reactions.

Fission yields of asymmetric products decrease slightly, whereas those of symmetric products increase significantly with excitation energy.
Peak-to-valley (P/V) ratio as a function of excitation energy in the $^{232}\text{Th}(\gamma,f)$ and $^{232}\text{Th}(n,f)$ reactions and in the $^{238}\text{U}(\gamma,f)$ and $^{238}\text{U}(n,f)$ reactions.

At all excitation energy, the P/V ratio in $^{232}\text{Th}(\gamma, f)$ and $^{232}\text{Th}(n, f)$ reactions are lower than in the $^{238}\text{U}(\gamma, f)$ and $^{238}\text{U}(n, f)$ reactions. This is due to the third peak in the former than later.
Summary for Yield Distribution of Fission Products

(i). The mass yield distribution in the bremsstrahlung induced fission of pre-actinides ($^{nat}$Pb&$^{209}$Bi) is symmetric and for pre-actinide ($^{238}$U) is asymmetric with double humped, whereas for $^{232}$Th, it is asymmetric with triple humped.

(ii). The higher yields of fission products for $A=133-135$, $138-140$, $143-145$ and their complementary products even at the bremsstrahlung energies of 10-80 MeV indicate the effect of nuclear structure. There is no such effect for pre-actinides.

(iii). For $^{nat}$Pb($\gamma$, f) and $^{209}$Bi ($\gamma$, f) the FWHM of the mass yield distribution increases and $<A_L>$ decreases with $E^*$. For $^{232}$Th($\gamma$, f), $^{232}$Th(n, f) and $^{238}$U(n, f), the $<A_H>$ decreases with $E^*$, whereas in $^{238}$U($\gamma$, f), it is constant with $E^*$.

(iv). The P/V ratio in the $^{232}$Th($\gamma$, f), $^{232}$Th(n, f), $^{238}$U($\gamma$, f) and $^{238}$U(n, f), decrease with increase of $E^*$. However the decrease trend is very sharp at the lower $E^*$ for $^{232}$Th($\gamma$, f).