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

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ABSTRACT

We determined the yields of fission products in the photon-induced fission for ^{232}Th and ^{238}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 ^{nat}Pb, ^{209}Bi, and ^{232}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 $^{232}Th(\gamma, f)$ and $^{238}U(\gamma, f)$ are compared with the similar data in $^{232}Th(n, f)$, and $^{238}U(n, f)$ to examine the role of excitation energy on nuclear structure effects. Similarly, the present data in the $^{nat}Pb(\gamma, f)$ and ^{209}Bi (γ, 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.

INTRODUCTION

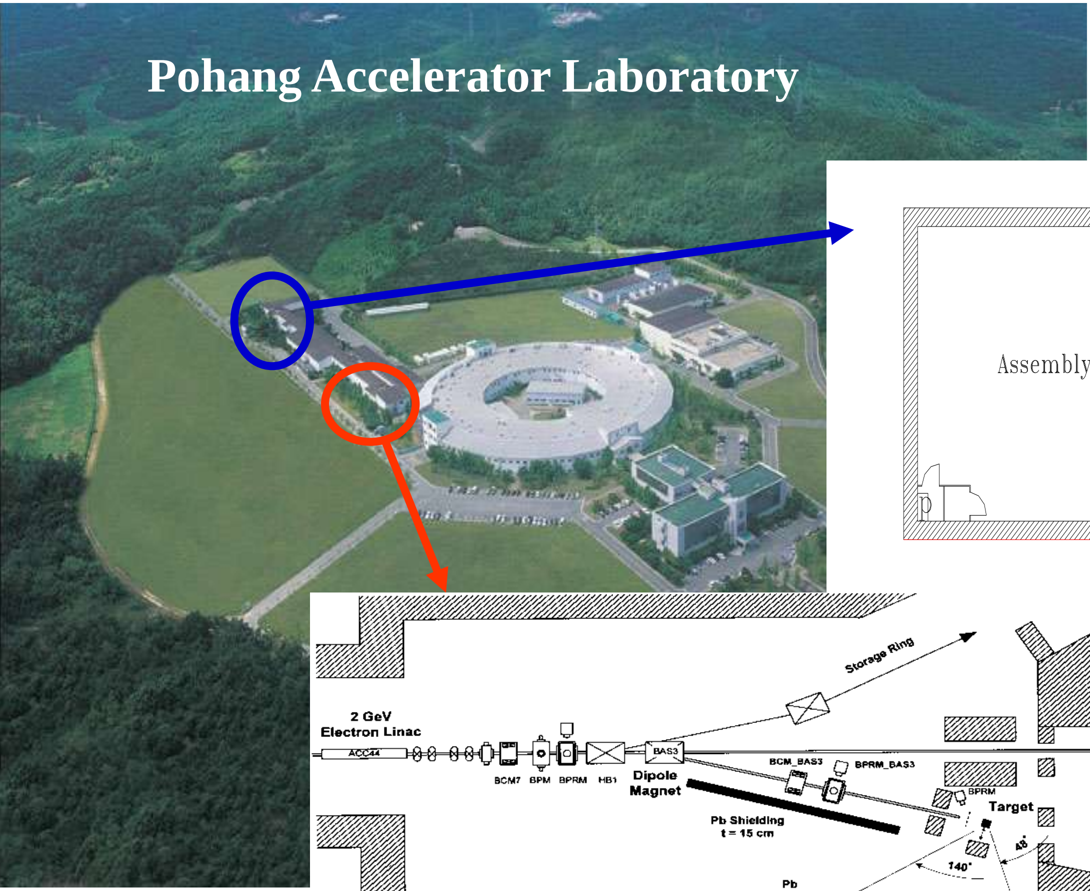
- Studies on fission product yields (mass yield distribution) in the neutron- and photon-induced fission of pre-actinides (^{nat}Pb, ^{209}Bi) and actinides (^{232}Th, ^{238}U) are important for their application in ADSs [1, 2] fast reactor [3] and AHWR [4].

- In ADSs, the heavy elements such as ^{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.

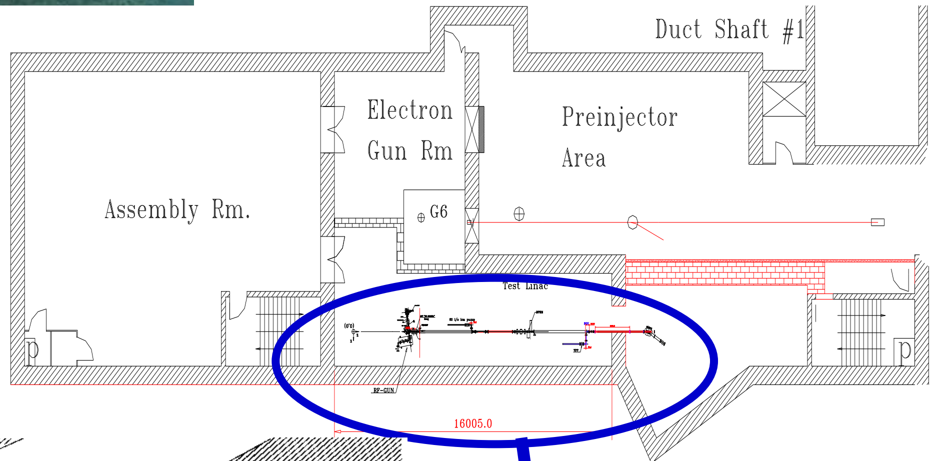
-It is necessary to determine the yields of products in the high energy neutron/bremsstrahlung induced fission of the spallation targets and long-lived minor actinides, which are need for the design of the ADSs.

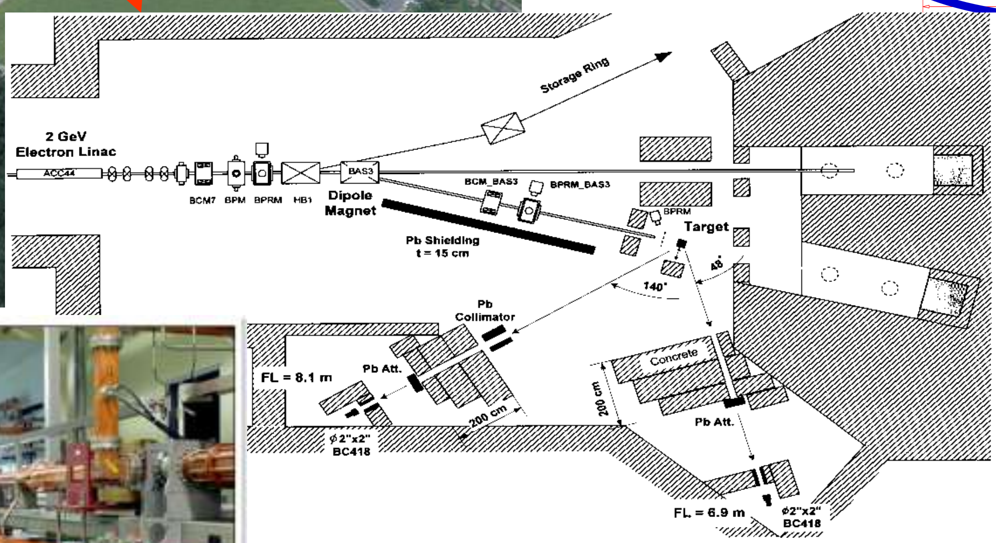
- During the spallation process high energy neutrons and photon (bremsstrahlung) produces, which can cause photo-nuclear reaction and fission of the spallation source and long-lived minor actinides.

1. Pohang Neutron Facility based on electron linac



Pohang Neutron Facility based on 100-MeV e-linac





Pohang High Energy Radiation Facility with 4.0 GeV e-linac

Experiment with Bremsstrahlung at PNF

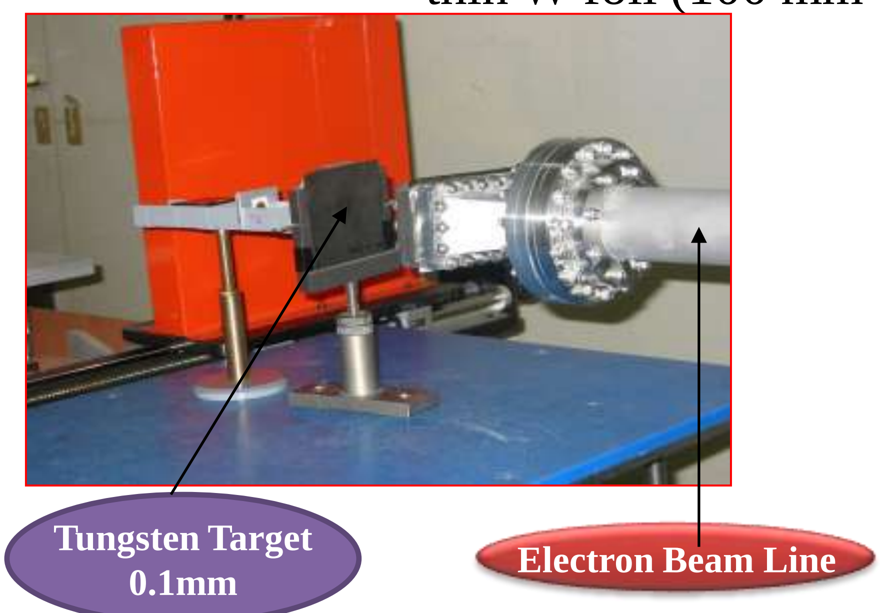
-The yields of fission products in the 45-80 MeV and 2.5 GeV (γ, f) of ^{nat}Pb, ^{209}Bi and ^{232}Th by off-line γ-ray spectrometric technique and using the electron linac at PAL, Pohang, South Korea [9-15].

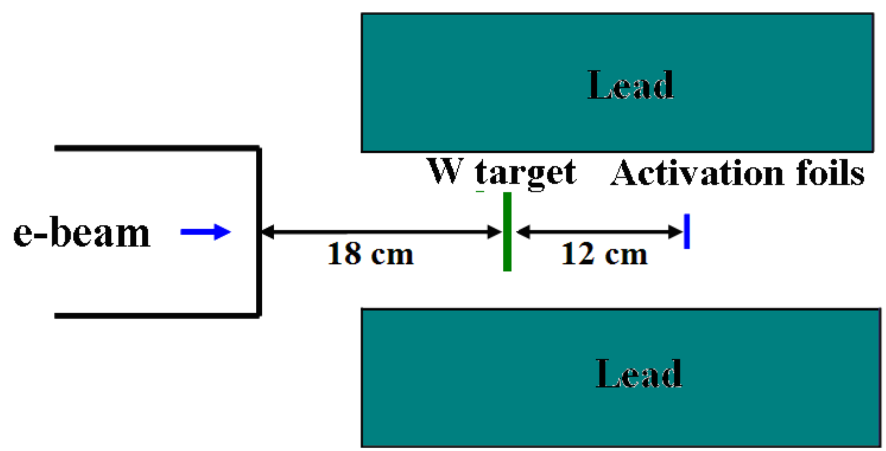
Operation Condition of electron linac:

- 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

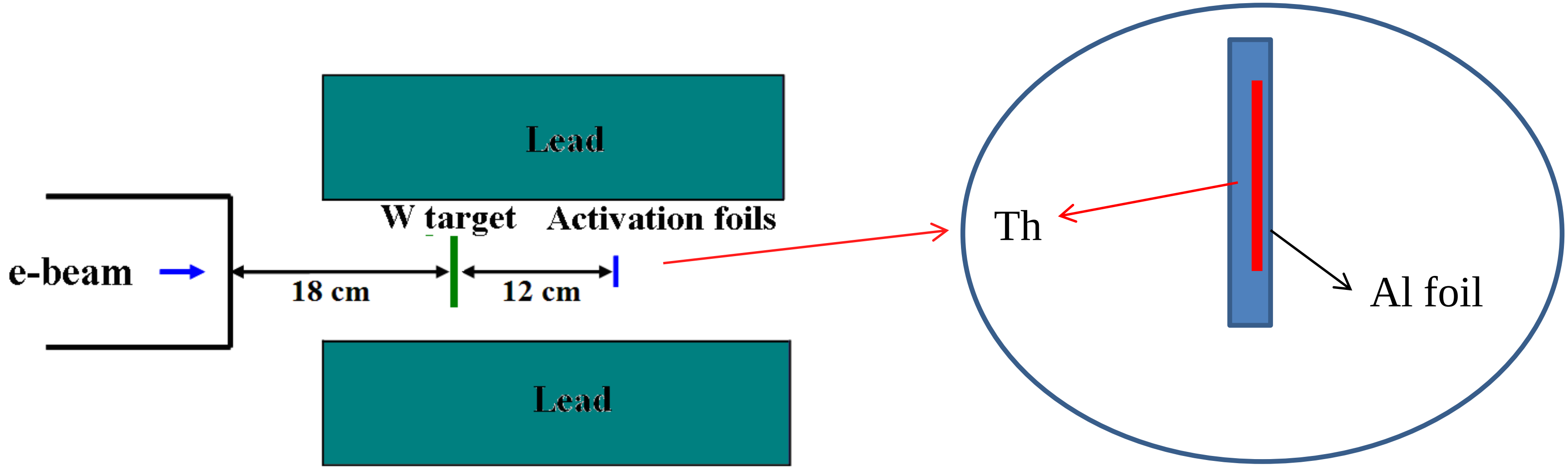
- The yields of fission products in the 10 MeV (γ, f) of ^{232}Th and ^{238}U using the electron linac at EBC, Kharghar, Navi-Mumbai, India [16].

Facility	EBC, India
W (Ta)foil size	100 cm^2
W (Ta) thickness	1 mm
W(Ta) distance from exit window	3 cm
Sample distance From W(Ta)	10 cm
Electron energy	10 MeV
Beam current	50 mA
Pulse width	10 μs
Repetition rate	400 Hz

Specification of Samples for 45-80 MeV and 2.5 GeV

	Chemical Purity (%)	Weight (g)	Thickness (mm)	Size
^{232}Th	99.999	0.2-0.3	0.025	0.25 cm^2
^{nat}Pb	>99	12.417	0.5	25
^{209}Bi	>99	74.417	3.0	25
Al	99.99	Natural	0.025 , 0.018	

- 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.



Specification of Samples for 10 MeV

	Chemical Purity (%)	Weight (g)	Thickness (mm)	Size
^{232}Th	99.999	0.2822	0.025	2.72 cm^2
^{238}U	>99	0.2414		2.5 cm^2
Al	99.99	Natural	0.025 , 0.018	

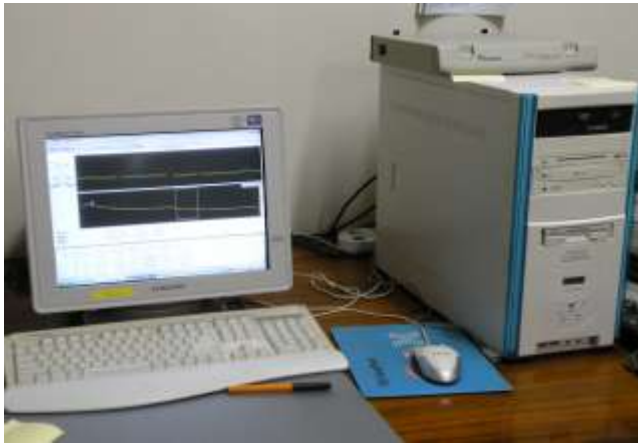
The metal targets were wrapped with 0.025 mm thick Al foil and additionally with 0.018 mm Al foil.

- 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)

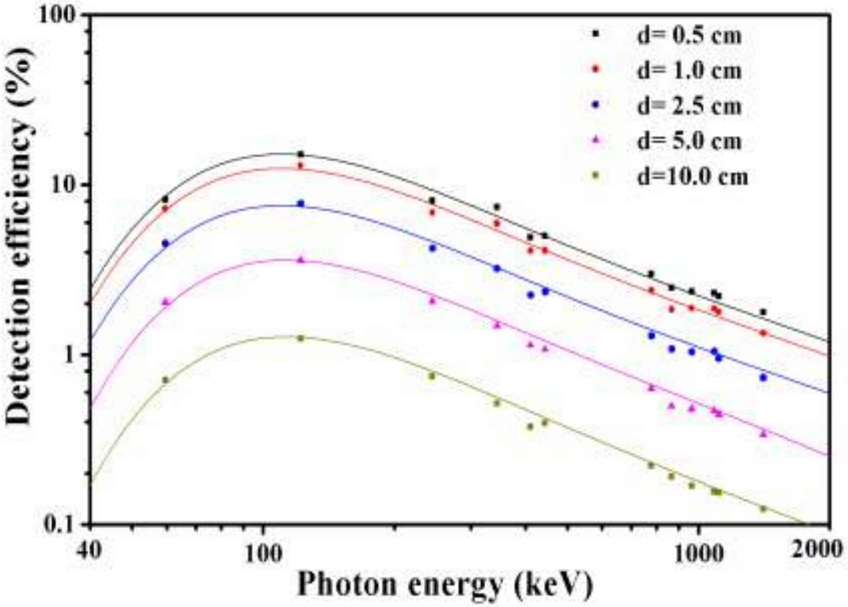


Detection Efficiency:

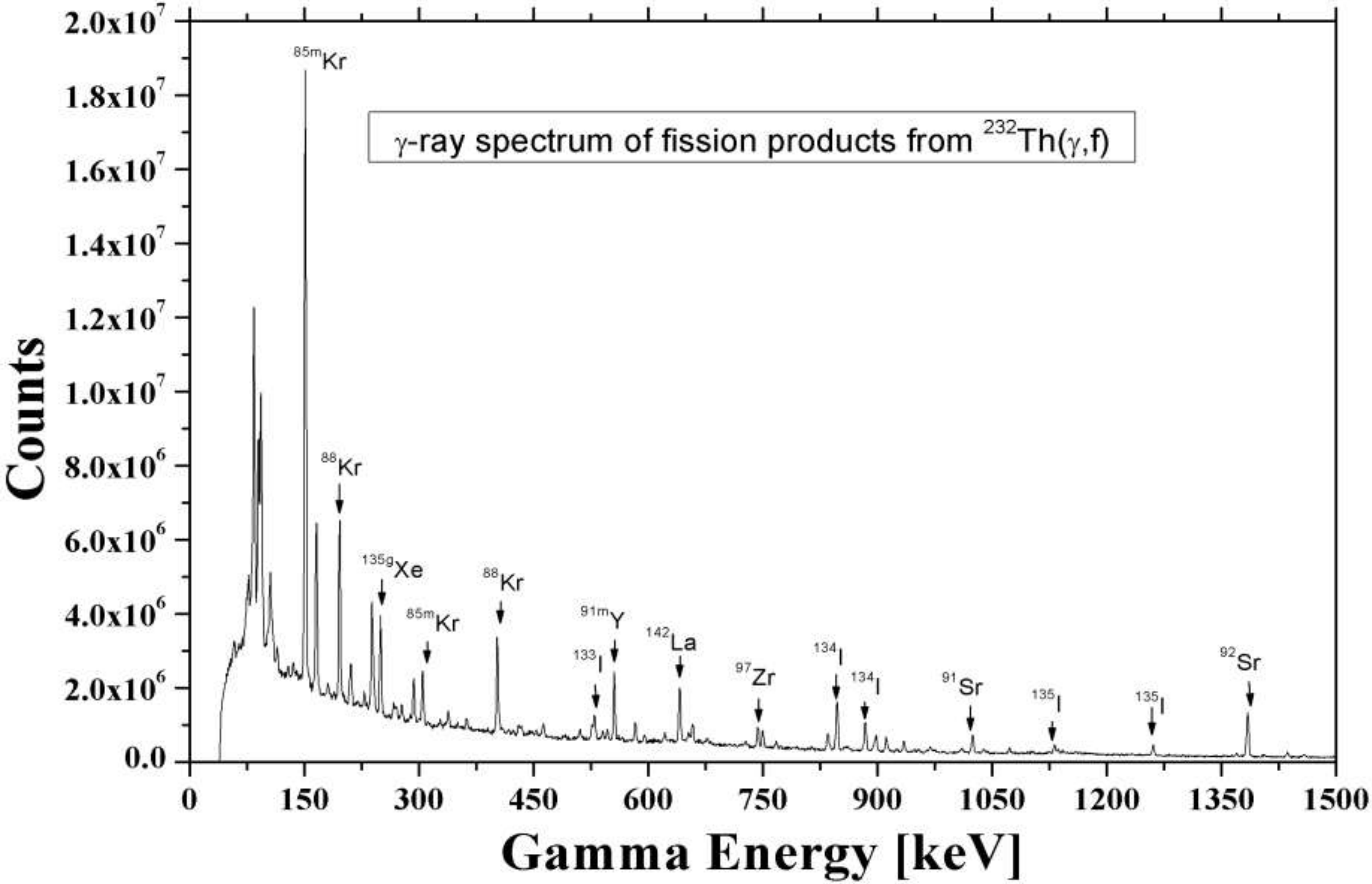
$$\varepsilon(E_\gamma) = \frac{N(E)}{I_\gamma(\%)}\frac{1}{A_{ref}.e^{-\lambda.t_d}}$$

Fifth order polynomial fitting: $\ln \varepsilon = \sum_{i=0}^{5} a_i (\ln E)^i$

Nuclide	Half life (Yrs.)	No. of γ-rays	E_γ(keV)	I_γ(%)
^{241}Am	432.2	1	59.5412	35.9
^{152}Eu	13.537	11	121.78 244.70 344.28 411.12 443.97 778.91 867.38 964.08 1085.87 1112.07 1408.00	28.58 7.58 26.50 2.23 3.15 12.94 4.25 14.61 10.21 13.64 21.01



Typical γ-ray spectrum of fission products in the 70-MeV bremsstrahlung-induced reaction from ^{232}Th



3. Data Analysis

➢ **Determination of Average Excitation Energy :**

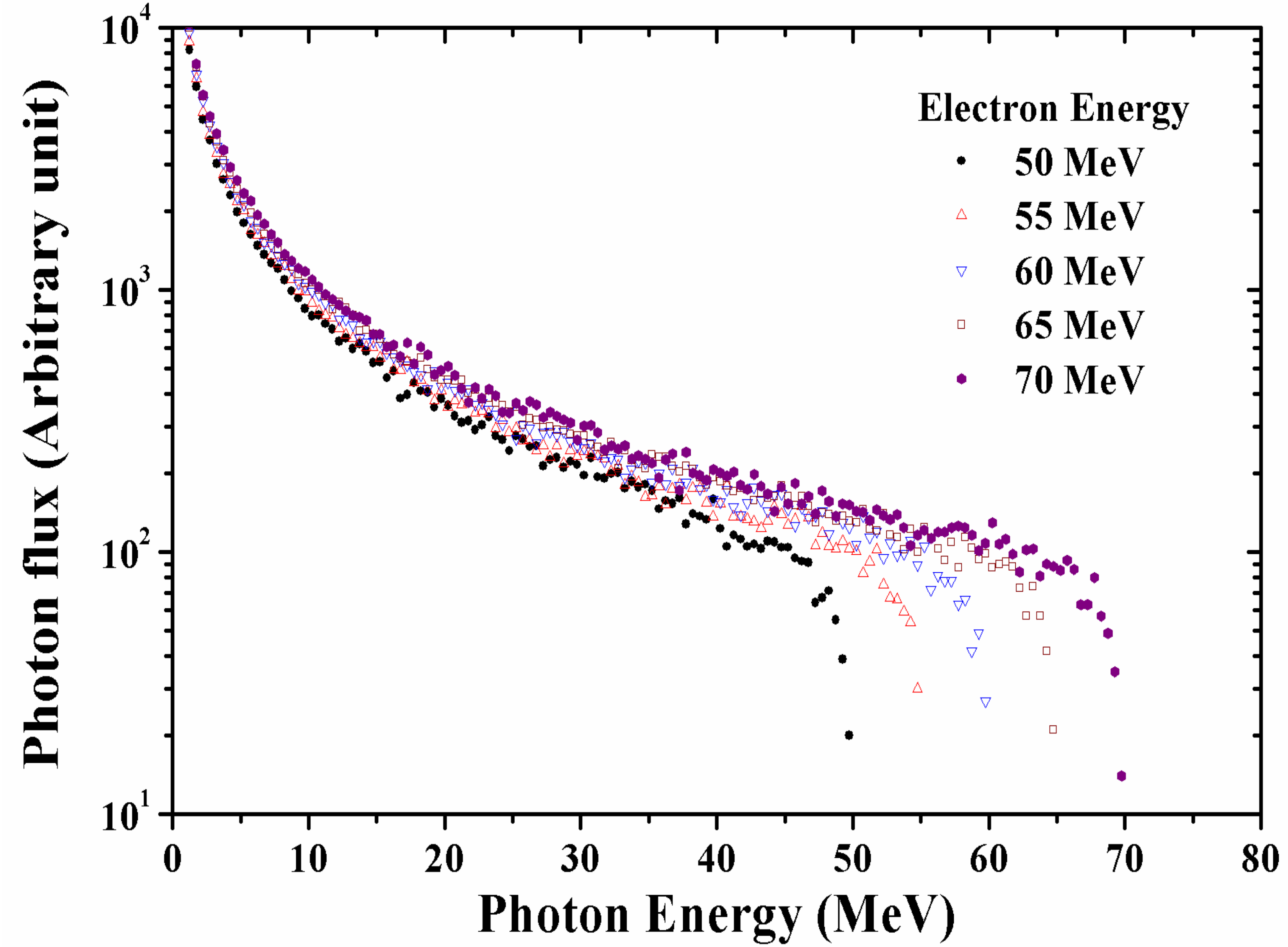
$$\left\langle E^*(E_e) \right\rangle = \frac{\int_0^{E_e} E_e \; N(E_e, E_\gamma)\sigma_F(E_\gamma)dE_\gamma}{\int_0^{E_e} N(E_e, E_\gamma)\sigma_F(E_\gamma)dE_\gamma}$$

$N(E_e, E_\gamma)$ **The number of photons with an energy E_γ 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_γ**
Using the TALYS 1.4

E_γ [MeV]	40	45	50	60	70	80
<E*> [MeV]	15.87	16.95	17.86	19.76	21.25	22.49

$N(E_e, E_\gamma)$ **The number of photons with an energy E_γ 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 (ϕ) 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. λ is the decay constant of the isotope of interest and ε is the detection efficiency of the γ-rays in the detector system. I_γ is the abundance or the branching intensity of the chosen γ-rays of the reaction products.

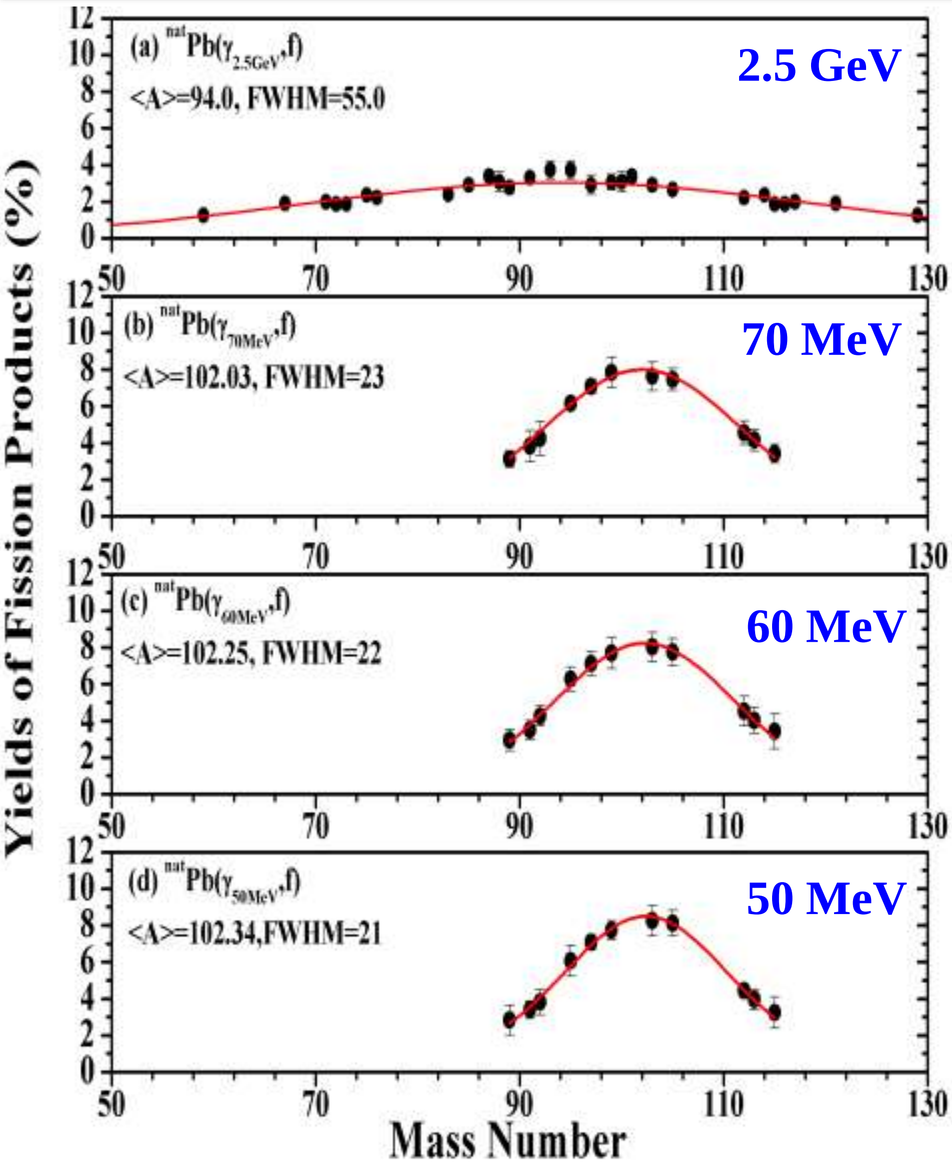
- From the relative **cumulative yields (Y_R)** of the fission products, their relative mass-chain yields (Y_A) were determined by :

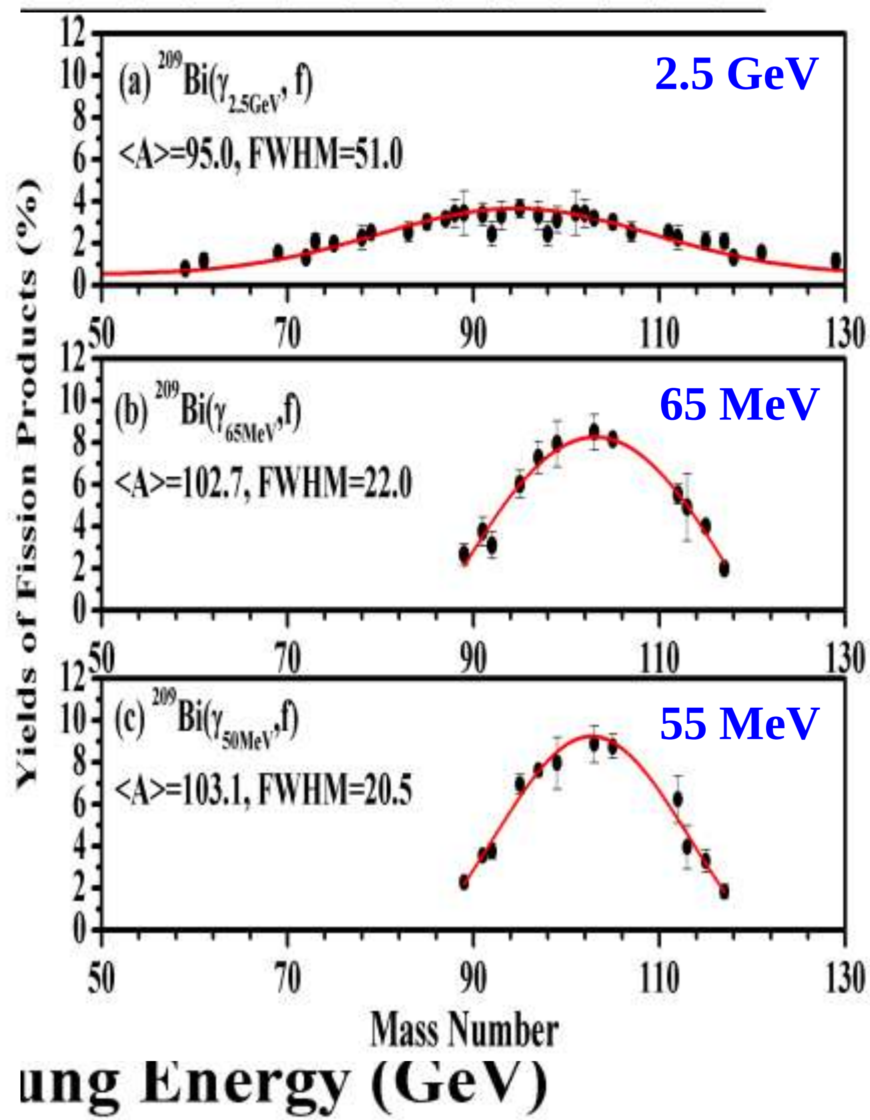
$$Y_A = Y_R / FCY, \qquad FCY = \frac{EOF^{a(Z)}}{\sqrt{2\pi\sigma_z^2}} \int_{-\infty}^{Z+0.5} \exp\left[-(Z-Z_P)^2/2\sigma_z^2\right] dZ$$

where FCY is the fractional cumulative yield, Z_P is the most probable charge and σ_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.

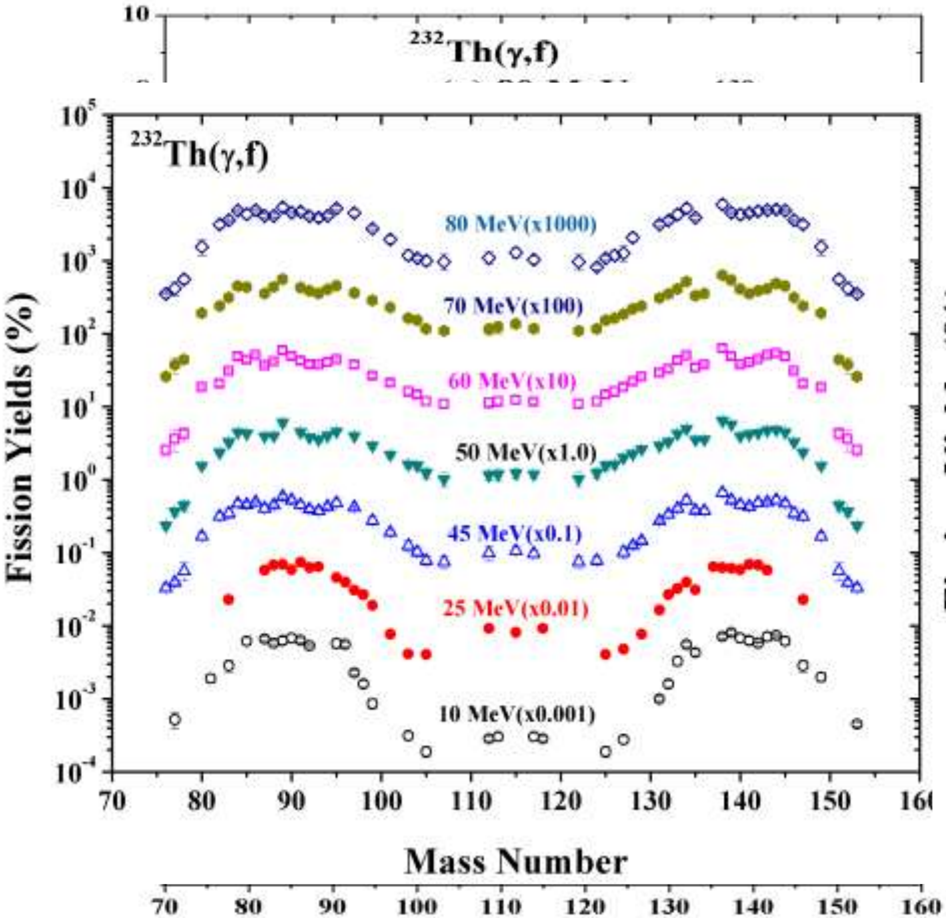
4. Result and Discussion

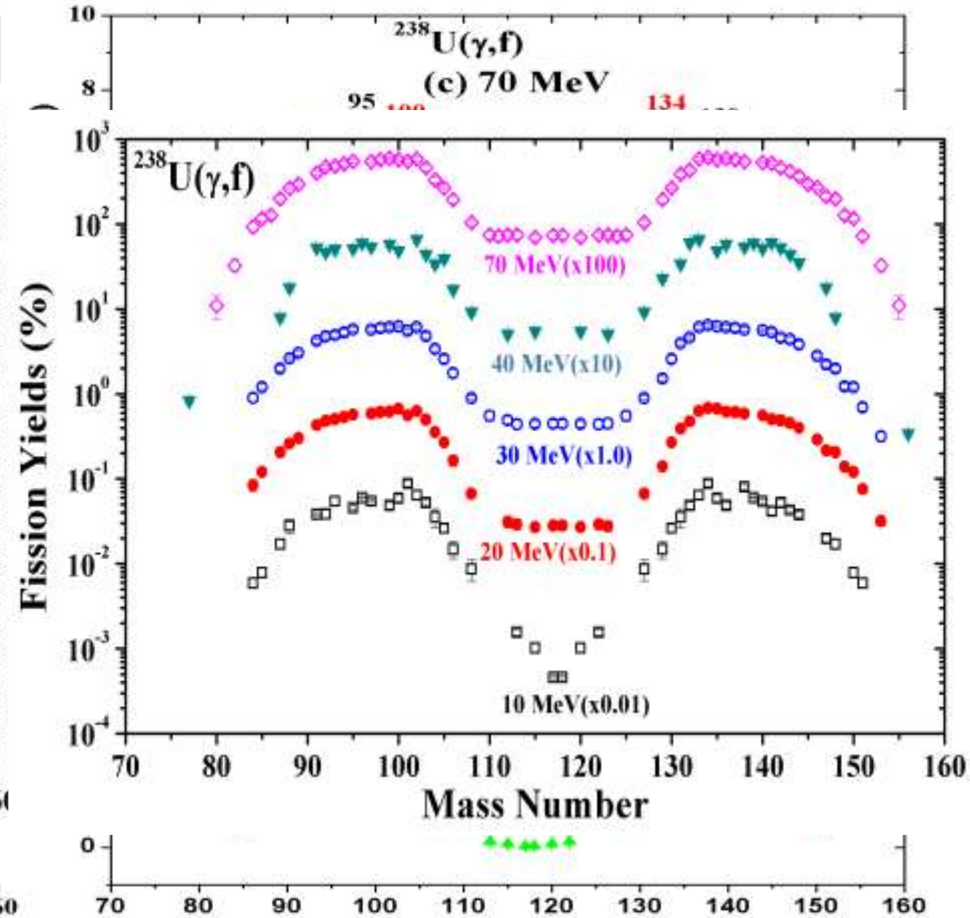
- Yields of Fission Products (%) as a function of mass number in the γ-induced fission of ^{nat}Pb and ^{209}Bi.





Yields of Fission Products (%) as a function of mass number in the γ-induced fission of ^{232}Th and ^{238}U

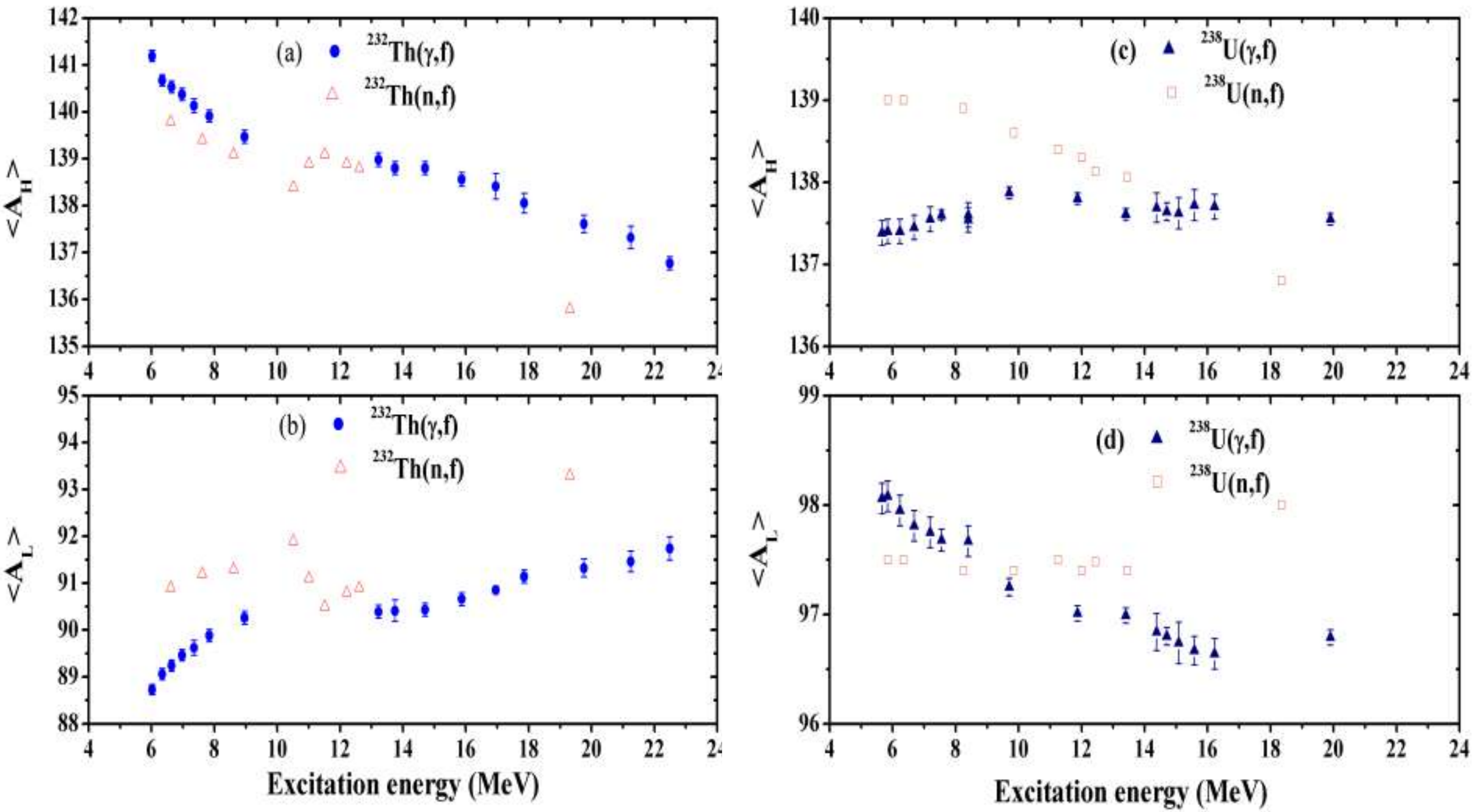




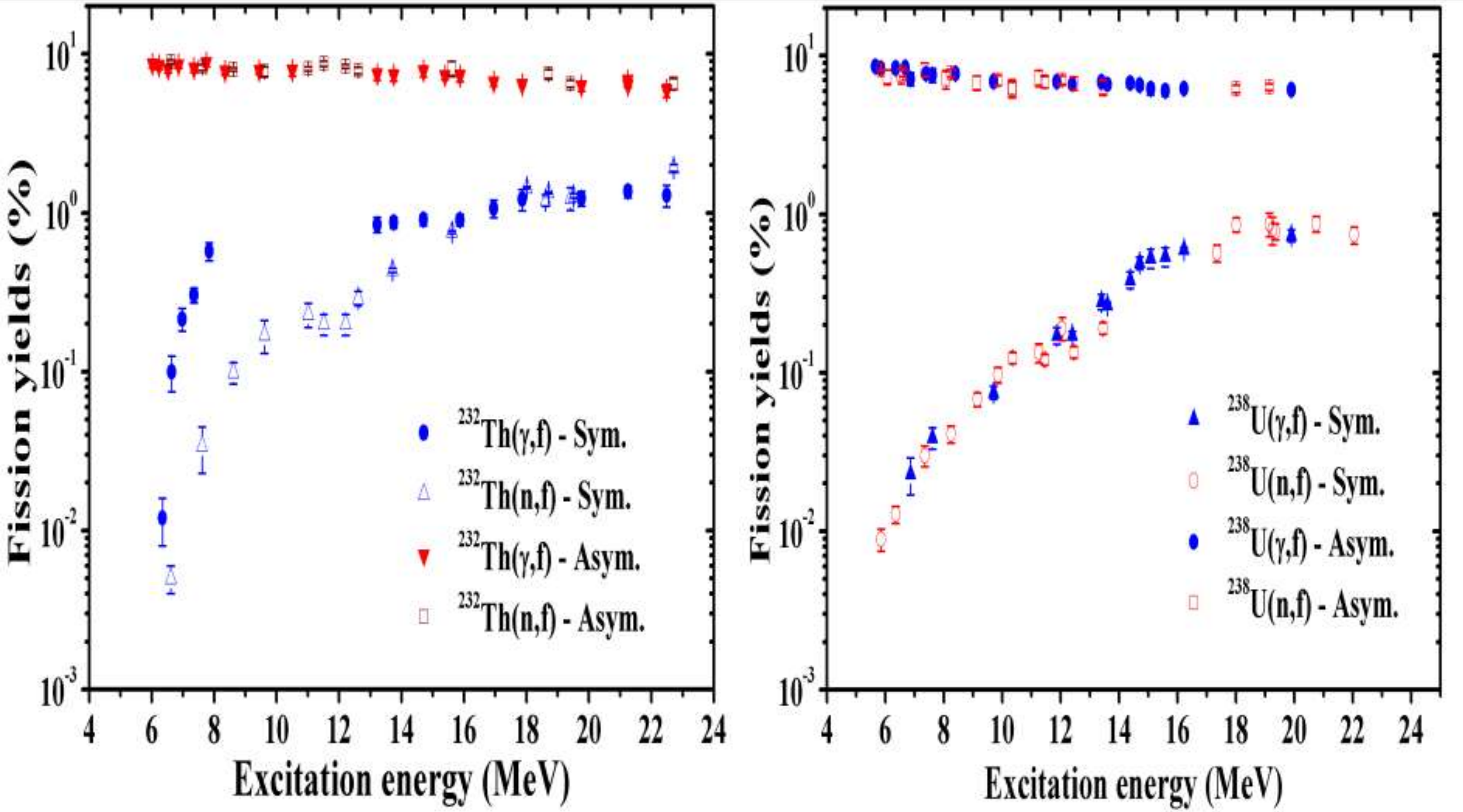
The Fission Yield distribution in ^{232}Th(γ, f) reaction is triple humped, whereas that in the ^{238}U(γ, f) reaction is double humped. This is due to the different type of potential energy surface in ^{232}Th* compared to ^{238}U*, which is called as **the Th anomaly**

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}Th(γ,f) and ^{232}Th(n,f) reactions as well as in the ^{238}U(γ,f) and ^{238}U(n,f) reactions.

$$\langle A_L \rangle = \sum (Y_A A_L) / \sum Y_A, \quad \langle A_H \rangle = \sum (Y_A A_H) / \sum Y_A,$$

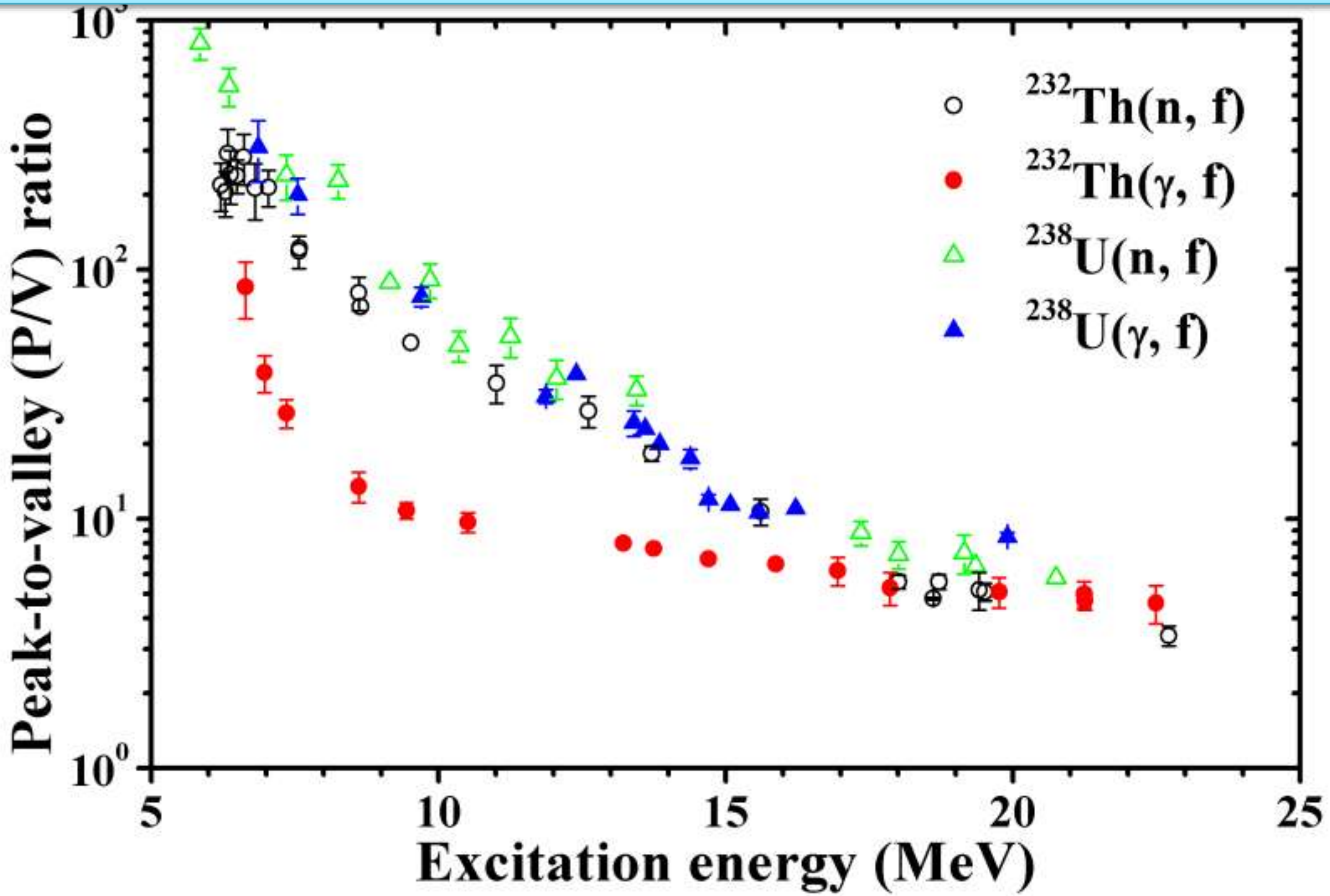


Fission Yields (%) of symmetric and asymmetric fission products in the ^{232}Th(γ,f) and ^{232}Th(n,f) reactions and in the ^{238}U(γ,f) and ^{238}U(n,f) reactions.



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}Th(γ,f) and ^{232}Th(n,f) reactions and in the ^{238}U(γ,f) and ^{238}U(n,f) reactions.



At all excitation energy, the P/V ratio in ^{232}Th(γ, f) and ^{232}Th(n, f) reactions are lower than in the ^{238}U(γ, f) and ^{238}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(γ, f) and ^{209}Bi (γ, f) the FWHM of the mass yield distribution increases and $\langle A_L \rangle$ decreases with E*. For ^{232}Th(γ, f), ^{232}Th(n, f) and ^{238}U(n, f), the $\langle A_H \rangle$ decreases with E*, whereas in ^{238}U(γ, f), it is constant with E*.

(iv).The P/V ratio in the ^{232}Th(γ, f), ^{232}Th(n, f), ^{238}U(γ, 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(γ, f).