Yields of fission products from 19, 32 and 44 MeV proton induced fission of ^{232}Th

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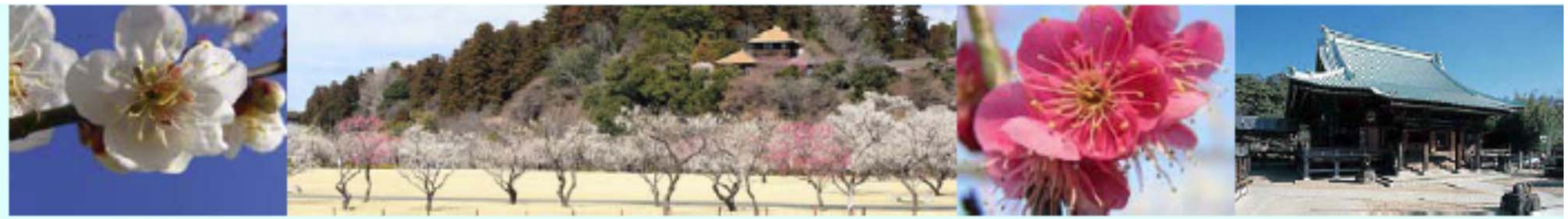


ABSTRACT

The yields of various fission products in the 19-, 32- and 44-MeV proton-induced fission of ^{232}Th have been experimentally determined by recoil catcher and an off-line γ-ray spectrometric technique using the BARC-TIFR Pelletron facility at India and medical cyclotron (MC-50) at (KIRAMS) Seoul, Korea. The mass-yield distributions were obtained from the fission-product yield data using charge-distribution corrections. From the mass yield distribution of present work and the literature data at various energies, the peak-to-valley (P/V) ratio was obtained in ^{232}Th(p,f) and are compared with the similar data in ^{238}U(p,f). From these data following observations were made. (i) The mass-yield distributions in the ^{232}Th(p,f) reaction are triple humped unlike in the ^{238}U(p,f), where it is double humped. (ii) The yields of fission products for A=133-134, A=138-139 and A=143-144 and their complementary products in the ^{232}Th(p,f) reaction in the three energies of present work are higher than those of other fission products. This is due to the nuclear structure effect, which has been observed for the first time in the energy range of present work but not at other energies from the literature data of earlier work due to missing mass yield data for relatively short-lived fission products. (iii)The yields of symmetric products increase with excitation energy, which causes the decreasing trend of the peak-to-valley (P/V) ratio. (iv) At all excitation energies the P/V ratio in ^{232}Th(p,f) reaction is lower than the ^{238}U(p,f) reaction. However, the decrease trend of P/V ratio with excitation energy is more faster in ^{232}Th(p,f) reaction than in ^{238}U(p,f) reaction. This is due to the different type of potential energy surface in in the fissioning system ^{233}Pa* (^{232}Th(p,f)) compared to ^{239}Np*(^{238}U(p,f)).

INTRODUCTION

- Mass and charge distributions of photon-, neutron-, and proton-induced fission of pre-actinides and actinides are important for the understanding of the fission processes related to effect of nuclear structure and dynamics of descent from saddle to scission [1, 2].
- Mass distribution in the photon-, neutron-, and proton-induced fission of pre-actinides (e.g. W, Au, Pb, Bi) and heavy-Z actinides (e.g. Es to Lr) are symmetric in nature, whereas for medium-Z actinides (e.g. U to Cf) are asymmetric in nature.
- Mass distribution in the photon-, neutron-, and proton-induced fission of light-Z actinides (e.g. Ac, Th, Pa) are asymmetric with triple humped.
- With increase of excitation energy and Z of the actinides, mass distribution changes from asymmetric to symmetric and the effect of nuclear structure decreases.
- Photon-, neutron-, and proton-induced fission of Th and U are of more interest due to their applications in accelerated driven sub-critical system (ADSs), advanced heavy water reactor (AHWR), conventional light and heavy water reactor, and fast reactor.
- Photon-, neutron-, and proton-induced fission of ^{232}Th is more interesting compared to ^{238}U due to its different type of behavior expected from the systematic and theory, which is called as Th anomaly.

- The yields of various fission products in the proton-induced fission of ^{232}Th have been experimentally determined by recoil catcher and an off-line γ-ray spectrometric technique.

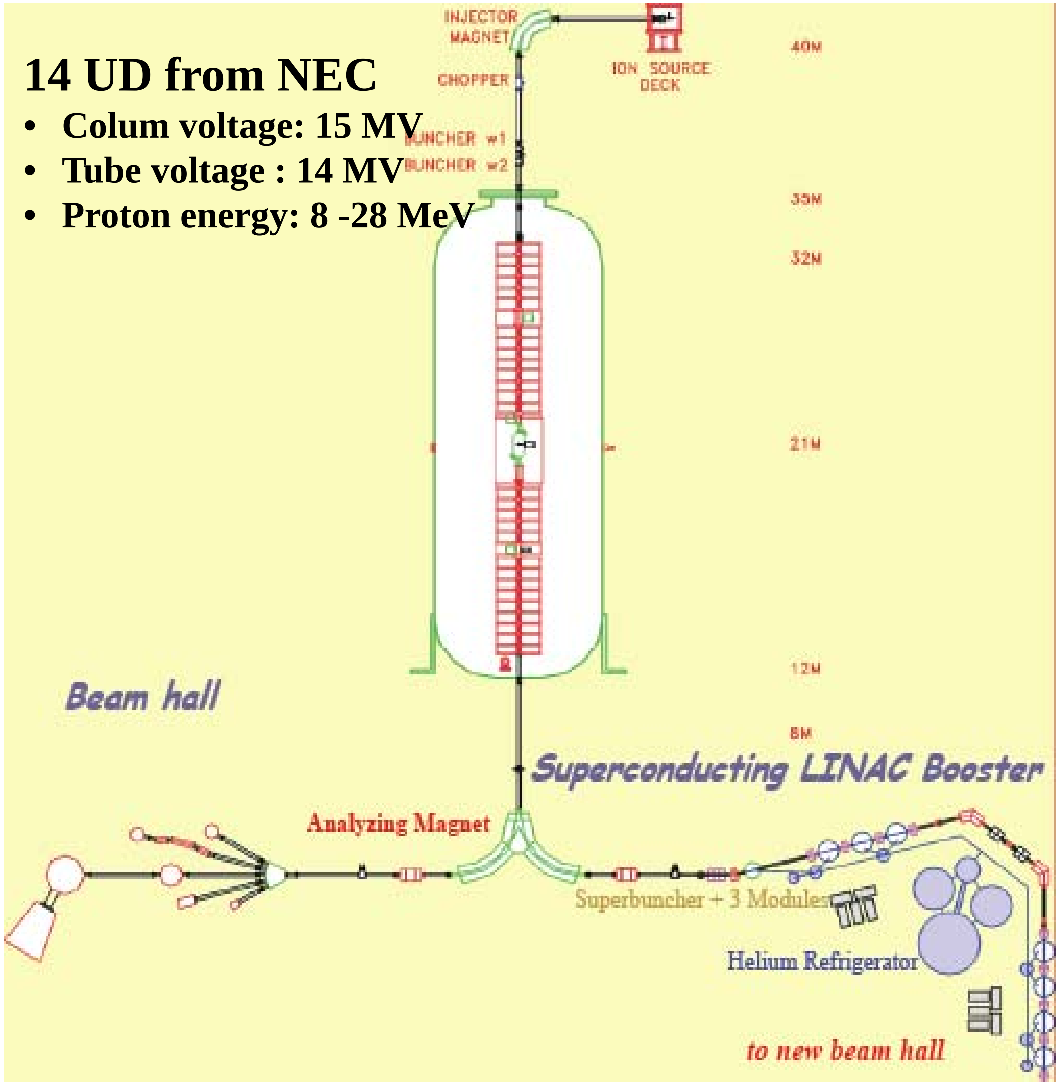
1. 19.55-MeV proton using the 14UD BARC-TIFR Pelletron facility, India.
2. 32- and 44-MeV proton using the medical cyclotron (MC-50) in KIRAMS, Seoul, Korea.

BARC-TIFR Pelletron Accelerator Facility

- The 19.55 MeV proton induced fission of ^{232}Th was carried out by using 14UD BARC-TIFR Pelletron at Mumbai, India.

14 UD from NEC

- **Colum voltage: 15 MV**
- **Tube voltage : 14 MV**
- **Proton energy: 8 -28 MeV**

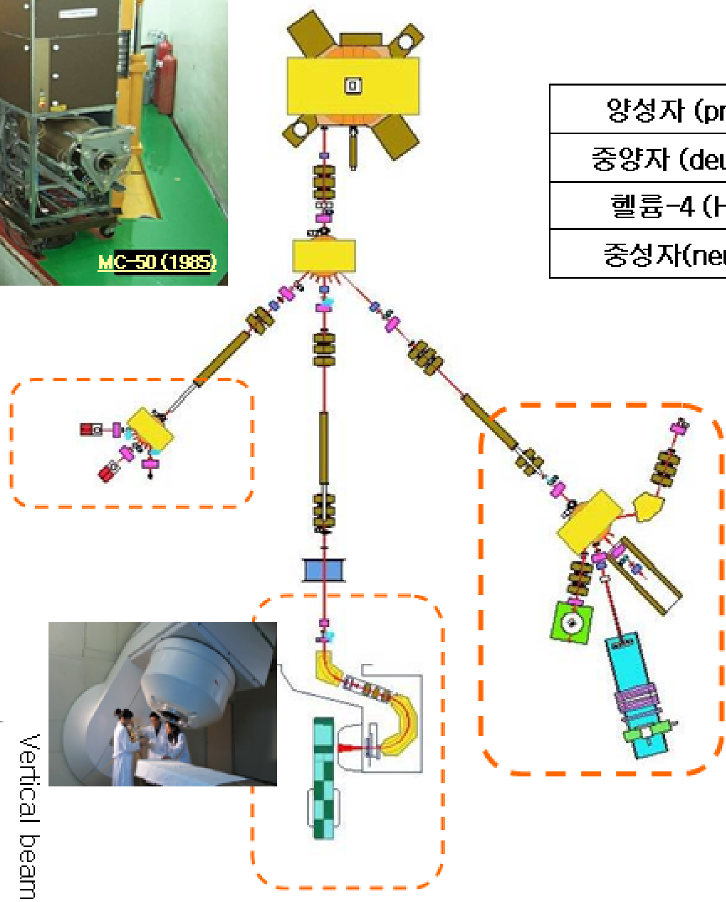


Proton Irradiation Facility

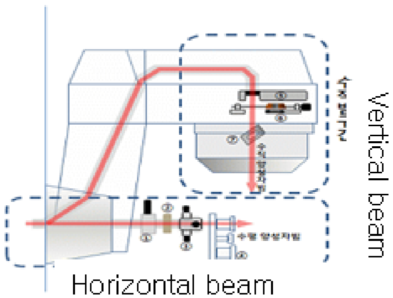
- **6m level above analyzing magnet**

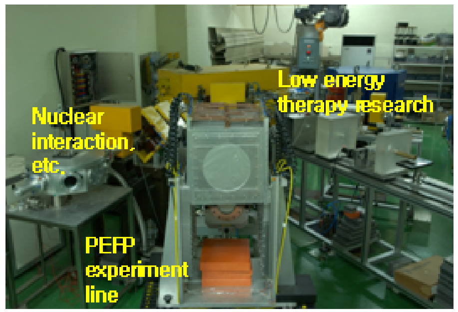
MC 50 Cyclotron Facility, KIRAMS

- The 32.21 and 44.76 MeV proton induced fission of ^{232}Th was carried out using medical cyclotron (MC-50) at the Korea Institute of Radiological and Medical Science (KIRAMS) in Seoul, Korea.

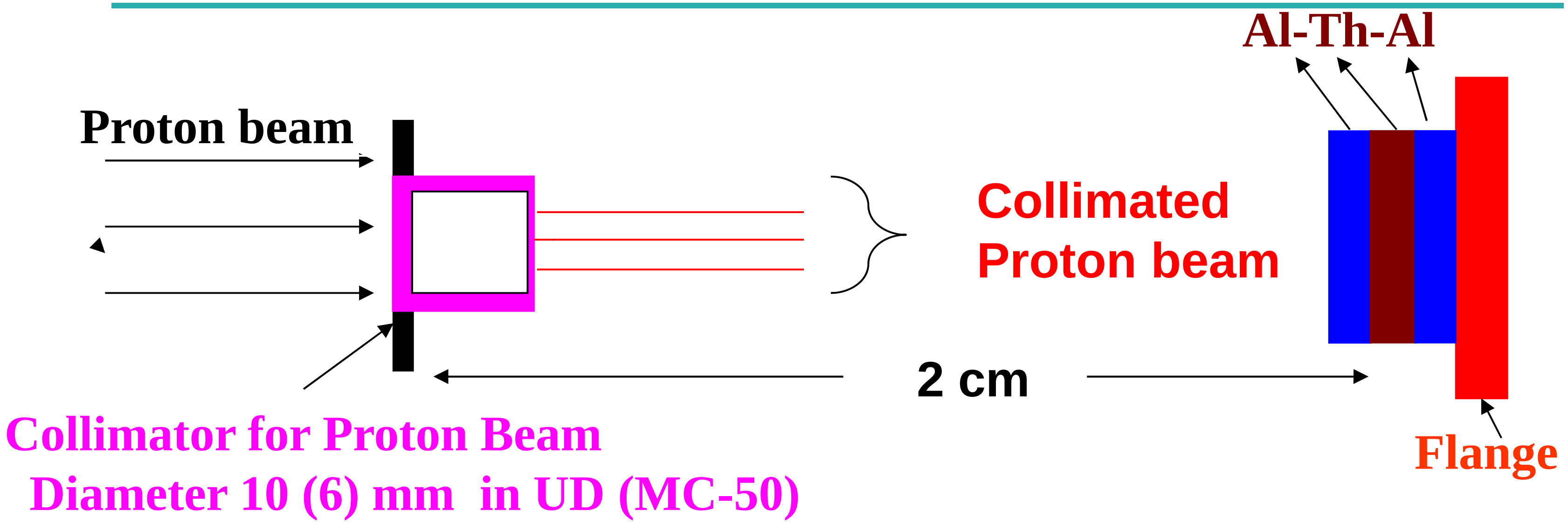


양성자 (proton)	20~51 MeV / 40 uA
중양자 (deuteron)	10~25 MeV / 30 uA
헬륨-4 (He-4)	20~50 MeV / 1 uA
중성자(neutron)	$E_{n,max} < E_{proton} - 2MeV$

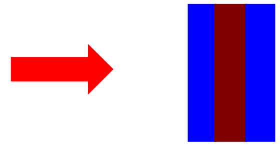




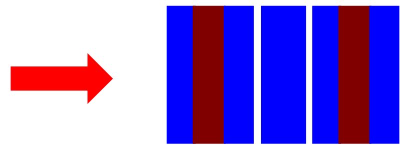
Experimental Arrangement and Sample Preparation



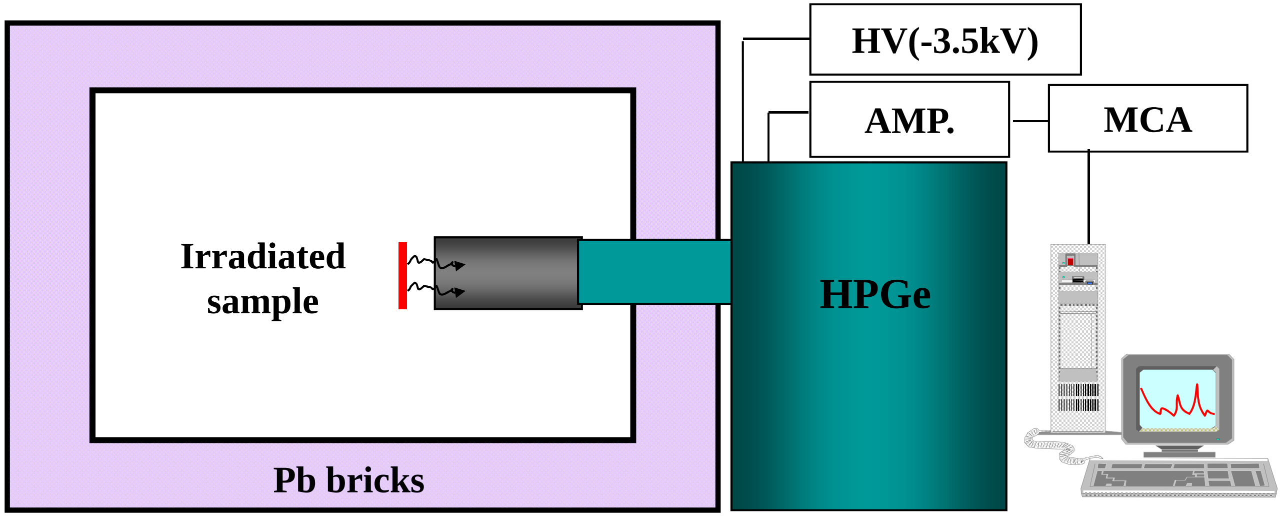
- The natural ^{232}Th metal target having the size of 1 cm^2 with thickness of 0.025 mm was covered with 0.025 mm thick super pure Al foil on both side of the target to make the Al-Th-Al stack.



- In the case of MC-50 cyclotron, in between two sets of Al-Th-Al stacks, a 3.8 mm thick Al was used as energy degrader. The two Al-Th-Al stack targets and thick Al degrader were also additionally wrapped with 0.025 mm thick Al.



Gamma ray Spectrometer



1: High-Purity Coaxial Germanium detector (HPGe),
(ORTEC, Model GEM-20180-p, Serial No. 39-TP21360A);
2: Preamplifier (ORTEC, Model 257 P, Serial No. 501);
3: Amplifier (ORTEC-572);
4: 4-Input Multichannel Buffer, Spectrum Master-919, (ORTEC);
5: Computer (Maestro, GammaVision)
6: Bias supply (High Voltage: +2kV, -3.5 kV) (ORTEC - 659)

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 proton induced fission cross-section of the target nuclei and Φ is the proton flux. 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.

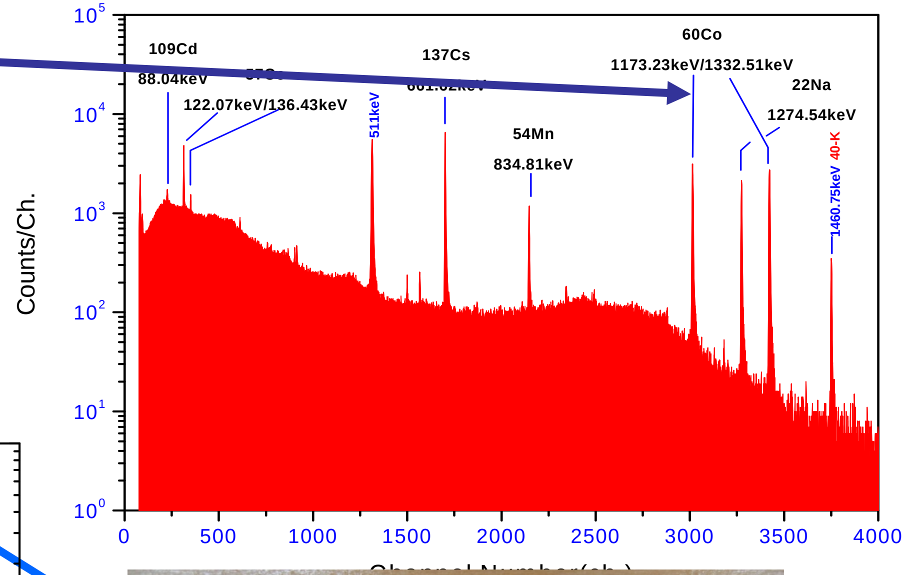
- From the relative **cumulative yields (Y_R)** of the fission products, their relative mass-chain yields (Y_A) were determined by :

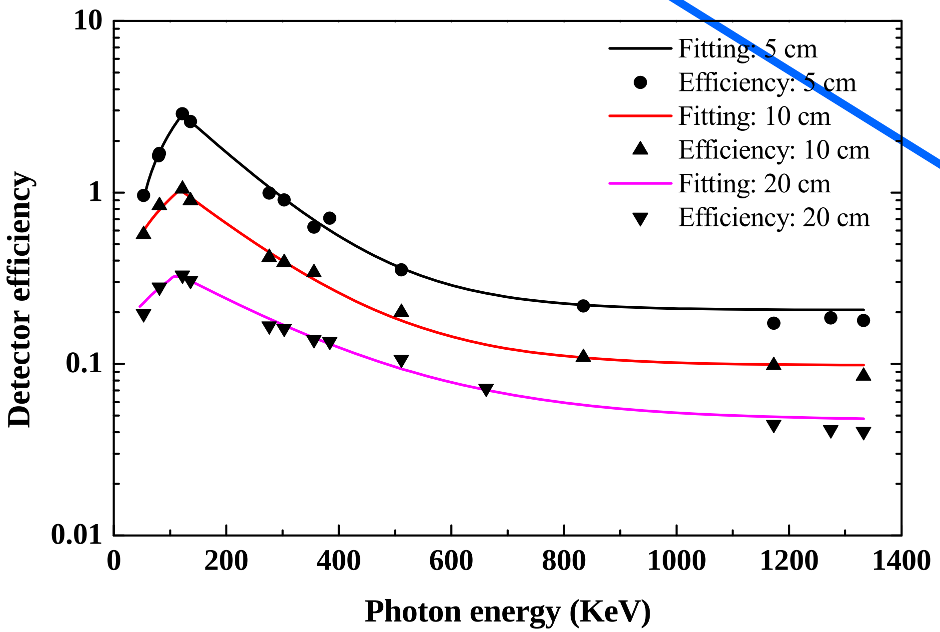
$$Y_A = Y_R / FCY, \qquad FCY = \frac{EOF^{a(Z)}}{\sqrt{2\pi\sigma_z^2}} \int_{-\infty}^{Z+0.5} \exp\left[-(Z-Z_P)^2/2\sigma_z^2\right] dZ$$

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.

Determination of Detector Efficiency

$$\varepsilon = \frac{CPS}{A_0 e^{-\lambda t} \times I_\gamma}$$





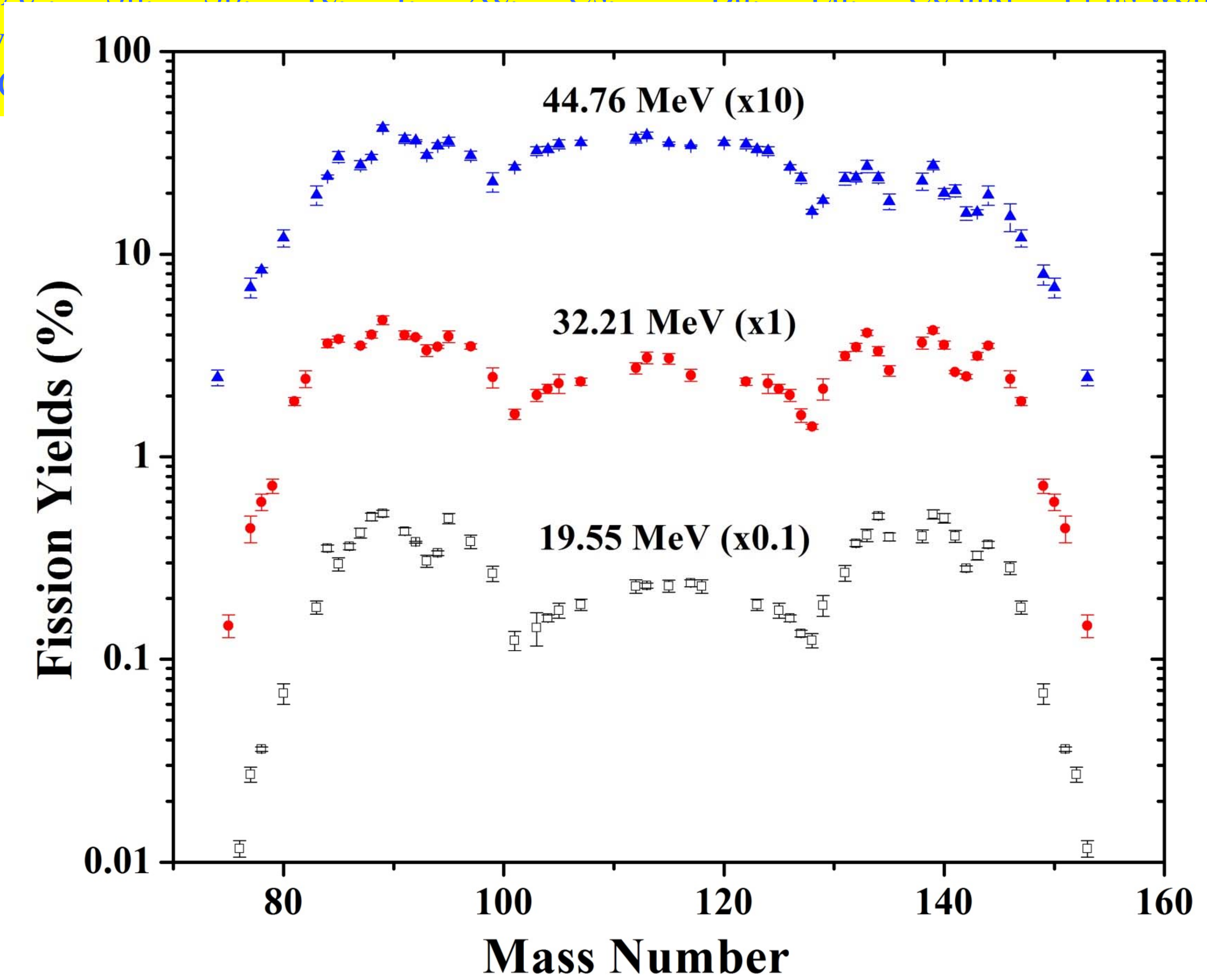
Uncertainties of Measurement

	Source of Uncertainty	%
(a) Random	(I) Counting statistics	3-4
	(ii) Irradiation time	1-1.5
	(iii) Rate of fission ($R=n\sigma\varphi$)	5-7
	(iv) Least sqare analysis)	5-7
	Total (σ_R)	7.8-10.8
(b) Systematic	(i) Half-lives	1
	(ii) Gamma ray abundance	2
	(iii) Branching ratio (abundance)	2-5
	(iv) Detector efficiency	5
	(v) Precursor yields	4-5
	Total (σ_S)	7-9

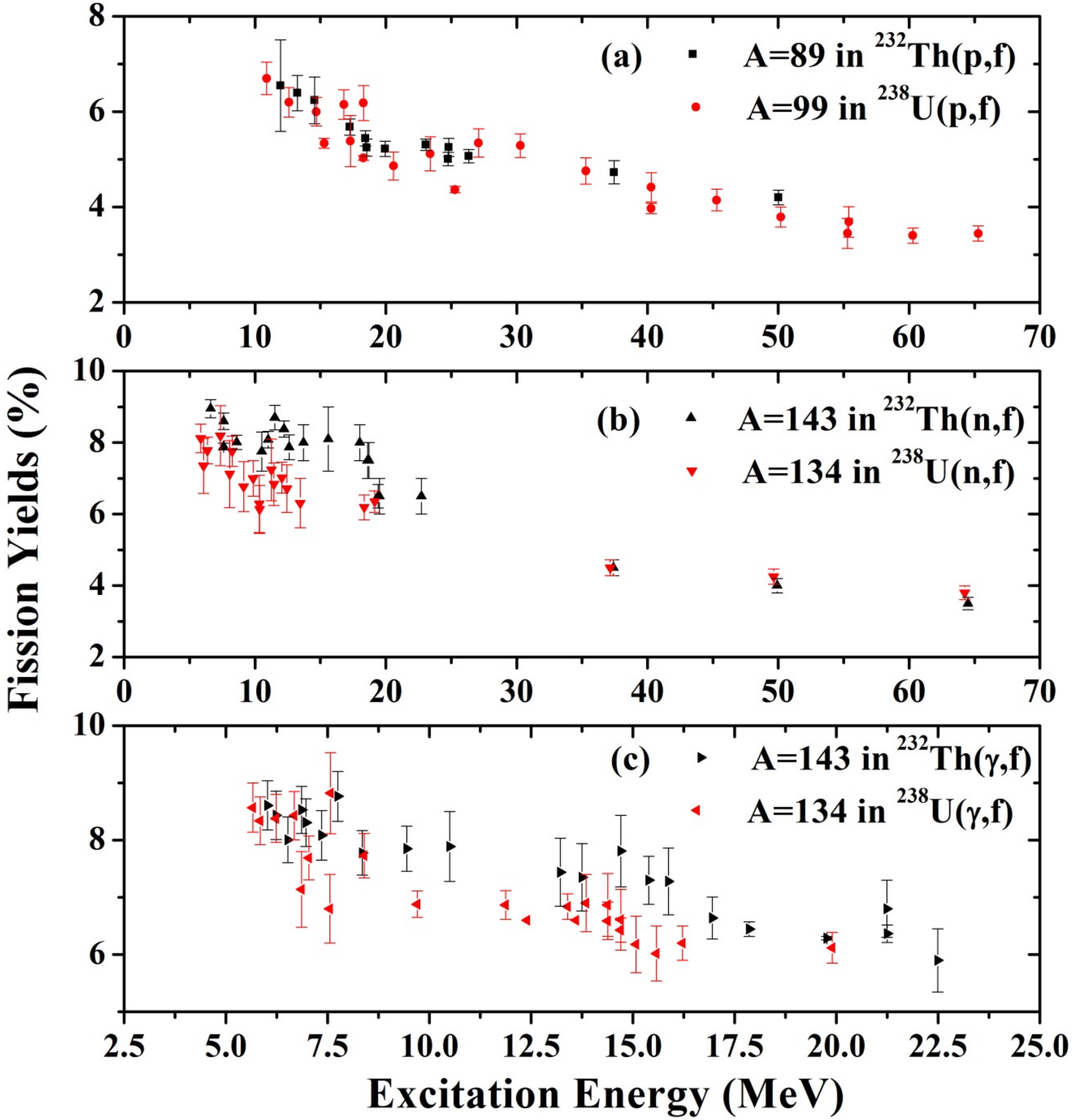
The mass yields distribution of fission products in the 32.21 and 44.76 MeV proton-induced

Fig. 1. Yields of Fission Products for Proton-induced fission of ^{232}Th

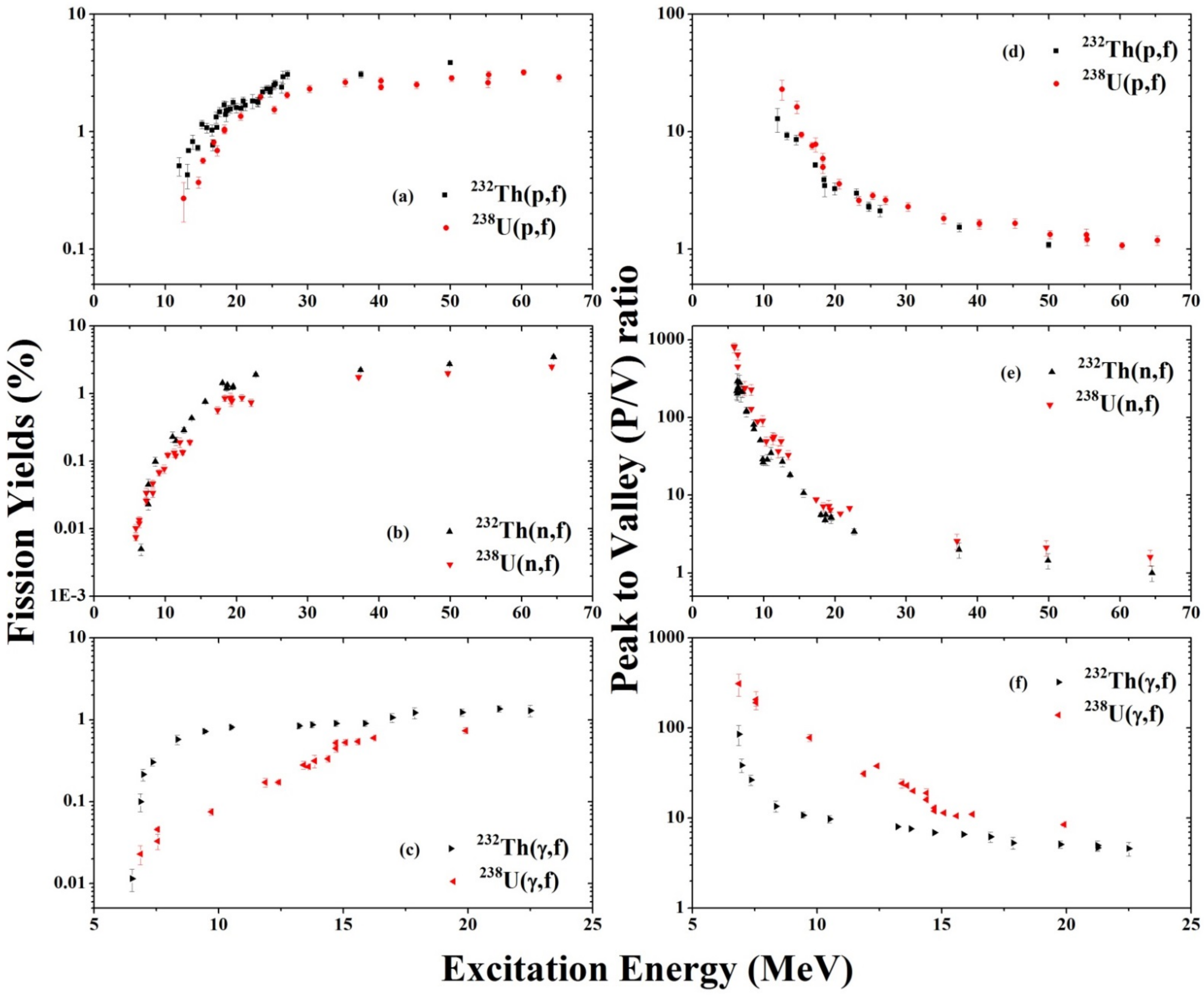
^{107}Rh, ^{113}Ag, ^{128}Sn, ^{131}Sb, ^{134}Te, ^{134}I, ^{138}Xe, ^{138}Cs, ^{141,142}Ba, ^{142}La, ^{146}Ce and ^{146}Pr as well as relatively … ^{140}Ba, ^{142}La, ^{144}…



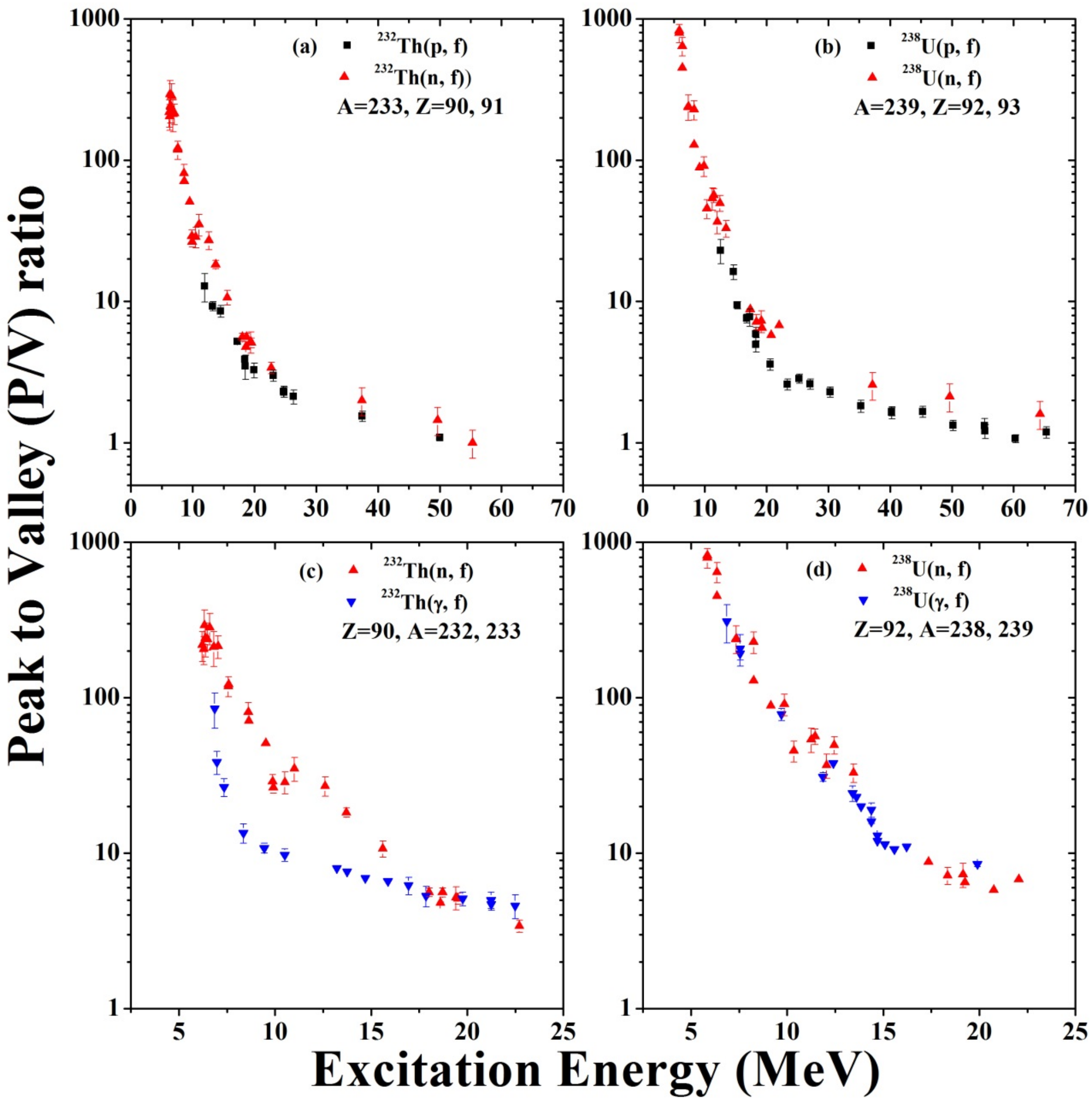
Yields of fission products (%) as a function of excitation energy for (a) A = 89 in the ^{232}Th(p, f) and A=99 in the ^{238}U(p, f), (b) A = 143 in the ^{232}Th(n, f) and A=134 in the ^{238}U(n, f) and (c) A = 143 in the 134 in the ^{232}Th(γ, f) and A=134 in the ^{238}U(γ, f) reactions.



Yields of symmetric fission products (%) and peak-to-valley (P/V) ratio as a function of excitation energy in the (a & d) $^{232}Th(p, f)$ and $^{238}U(p, f)$, (b &e) $^{232}Th(n, f)$ and $^{238}U(n, f)$, (c & f) $^{232}Th(\gamma, f)$ and $^{238}U(\gamma, f)$.



Peak-to-valley (P/V) ratio as a function of excitation energy for (a) A=233 & Z=91,90 i.e. ^{232}Th(*p, f*) and ^{232}Th(*n, f*), (b) A=239 & Z=93, 92 i.e. ^{238}U(*p, f*) and ^{238}U(*n, f*), (c) Z=90 & A=233, 232 i.e. ^{232}Th(*n, f*) and ^{232}Th(*γ, f*) and (d) Z=92 & A=239, 238 i.e. ^{238}U(*n, f*) and ^{238}U(*γ, f*).



Summary for Yields of Fission Products

(i) The yields of fission products in the 19.55-, 32.21 and 44.76-MeV proton-induced fission of ^{232}Th were determined by using an off-line γ-ray spectrometric technique. From the yields of various products mass chain yield were obtained by using charge distribution corrections.

(ii) The mass-yield distributions in the ^{232}Th(p, f) reaction at various energies are triple humped, similar to those of ^{232}Th(γ ,f) and ^{232}Th(n, f) reactions. The approach of symmetric split in the proton- and neutron-induced fission of ^{232}Th is faster than those in ^{238}U.

(iii) The yields of fission products for A = 133–134, A = 138–139, and A = 143–144 and their complementary products in the proton-induced fission of ^{232}Th are higher than those of other fission products. This is due to shell closure proximity based on standard I and II asymmetric mode of fission besides the probable even-odd effect and N/Z effect of the fissioning system on fission products.

Summary for Yields of Fission Products

IV. In the bremsstrahlung, neutron and proton induced fission of ^{232}Th and ^{238}U, the yields of high yield asymmetric products decreased marginally, whereas for symmetric products increased sharply with excitation energies. Accordingly, the P/V ratio in all the cases decreases with excitation energy. This shows the role of excitation energy.

V. At all excitation energies, the P/V ratio in the bremsstrahlung, neutron and proton induced fission of ^{232}Th is lower than those for ^{238}U. This is due to the different type of potential surface in the former than latter.

VI. At same excitation energy, for same-A compound nucleus, the P/V ratio is lower for heavier-Z than lighter-Z system. Conversely, for same-Z compound nucleus, the P/V ratio is lower for lighter-A than heavier-A system. This implies that the P/V ratio systematically decreases with increase of fissility parameters.

ACKNOWLEDGMENTS

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References

[1] R.Vandenbosch and J.R.Huizenga, *Nuclear Fission* (Academic, New York, 1973).

[2] C. Wagemans, *The Nuclear Fission Process* (CRC, London, 1990).

[3] H. A. Tewes and R. A. James, Phys. Rev. **88**, 860 (1952).

[4] H. Kudo, H. Muramatsu, H. Nakahara, K. Miyano and I. Kohno, Phys. Rev. C **25**, 3011 (1982).