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MASS DISTRIBUTION IN FISSION SPECTRUM NEUTRON INDUCED FISSION OF U-235

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Abstract: The mass yield distribution for the fission spectrum neutron induced fission of U-235 was mearsured for 40 nuclides. 10 nuclides'yields are reported for the first time. The irradiation was performed at the HWRR of IAE. The fission rate was counted by fission chamber. The fission product activity was mearsured by using radiochemical method as well as direct Ge(Li) γ-spectrometry. The mass yield curve is given and its features are described. And besides, fission yields of Mo-99 are discussed.

(Mass distribution, fission spectrum neutron, radiochemical method, Ge(Li) γ-spectrometry, Mo-99 yield)

Introduction

The mass distribution of fission products from thermal neutron induced fission of U-235 has been extensively investigated over the past many years. However, little work /1-4/ has been done on the fission spectrum neutron induced fission and in those studies the disagreement of the fission yield values for some nuclides is considerable. Hence it was felt to be desirable to carry out a more comprehensive and detailed study on the mass distribution of fission spectrum neutron induced fission of U-235.

On the other hand in 1980 we mearsured radiochemically the fission yields /5/ of 4 important nuclides, Zr-95, Mo-99, Nd-147 and Ce-144. In the case of Mo-99 yield, our value of 6.38±0.18% appears to be obviously higher than its thermal fission yield (6.11%). This makes someone doubt the validity about the value.

Therfore an attempt was made to measure as many as possible the fission yields of nuclides in the fission spectrum neutron induced fission of U-235 using radiochemical and Ge(Li) γ-spectrometric method. The aim of the present study was to obtain additional information for the mass distribution and to test the validity of our Mo-99 fission yield value.

Experimental

Irradiation and Fission Rate Measurement

The targets for neutron irradiation were 1.6cm diameter by 0.02cm thick disks of uranium metal with the weights from 0.5 to 1.0g and isotopic composition of 1.1% U-234, 90.2% U-235, 0.3% U-236 and 8.4% U-238. Irradiations were made in a concrete and paraffin shielded room of 3.5m ×3.0m ×2.2m at the heavy water reactor of IAE. The source of fission spectrum neutrons was a uranium plate enriched in U-235 and put in the thermal neutron beam from the reactor.

A sandwich of standard sample-target-standard sample was put in a double fission chamber. A diagrammatic sketch for neutron irradiation was presented in Fig.1. The fission rate of standard sample was directly counted in the double fission chamber during the entire irradiation. The standard samples were prepared by electrodepositing uranium on platinum disks quantitatively. The quantity was checked accurately by α-counting, 'thermal weighing' and volumetric analysis.

The fission rate in the target was calculated by multiplying the fission rate in standard sample by the ratio of the weights of the target and of the standard sample. Since the irradiation was carried out with fission neutrons obtained from the slow neutrons induced fission of U-235, it was necessary to surround the fission chamber with a thick layer of boron and cadmium so as to absorb all the slow neutrons. The thicknesses of the layers were 1.0cm for the boron carbide and 0.1cm for cadmium.

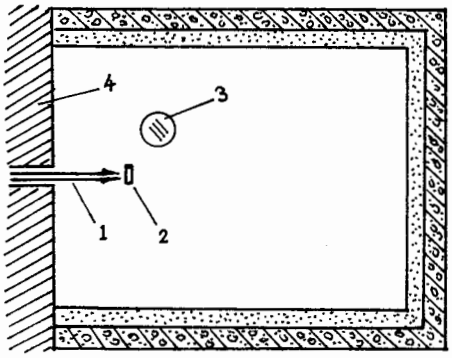

Fig. 1. Experimental arrangement for the irradiation of the target.
1 - reactor neutron beam, 2 - U-235 converter
3 - double fission chamber, 4 - reactor wall

Fission Products Measurements

γ-spectrometry method Some of fission product yields were directly determined by high-resolution γ-spectrometry of an irradiated uranium target. The γ-ray spectrometer system used in this work was based on lithium-drifted germanium detector of 130cm with the resolution of 1.85 keV (FWHM) for the 1.33 MeV γ-ray of Co-60. Pulses from the detector were amplified and fed into SCORPIO-3000 multichannel-computer system via livetime corrector/pileup rejector unit. These complex γ-ray spectra were analyzed with the computer program SAPL /6/ to obtain the intensities of photopeaks.

The full-energy photopeak counting efficiency of the detector as the function of γ-ray energy, Eγ, was determined from a set of standard sources --- Am-241, Nd-147, Ce-144, Ce-141, Mo-99, Ag-111, Na-22, Cs-137, Zr-95, Sc-46, Co-60 and Ta-182. These γ-ray sources were standardized by 4π β-γ coincidence technique, while Am-241 was standardized by α-counting. Important corrections were made as required for γ-cascade coincidence loss and self-absorbtion of γ-rays in the uranium target disk.

Radiochemical Method After irradiation the metal uranium target was dissolved in the 6-10M HCl solution containing some conc. HNO_3 and carrier of

Table 1. Yields of products in the fission spectrum neutron induced fission of U-235

Mass number	Nuclide	Fission yields, %					
		This work		Ferrieu et al. / 3 /	Debertin et al. / 4 /	Petrzhak et al. / 2 /	Bonyushkin et al. / 1 /
		GS	RC				
84	Br-84	1.10±0.07					
85	Kr-85m	1.35±0.09*		1.52±0.05			
87	Kr-87	2.47±0.09		2.59±0.07			
88	Kr-88	3.68±0.09		3.35±0.04			
89	Rb-89	4.07±0.19				5.6±0.4	4.4±0.4
91	Y -91m	5.39±0.18*		6.01±0.05			
92	Y -92	5.92±0.28		5.32±0.10			
93	Y -93	5.79±0.18		5.95±0.08			
94	Y -94	5.95±0.17					
95	Zr-95	6.42±0.20	6.46±0.30	6.52±0.07	6.40±0.07	7.7 ±0.6	5.85±0.55
97	Zr-97	6.12±0.17	6.04±0.46	6.33±0.09	6.09±0.04		6.55±0.70
99	Mo-99	6.37±0.20	6.36±0.17	6.08±0.08	6.22±0.15	6.4±0.4	5.9 ±0.4
103	Ru-103	3.46±0.13		3.25±0.04	3.46±0.03		3.75±0.55
104	Tc-104	1.77±0.05					
105	Rh-105	1.25±0.07		1.33±0.06			1.45±0.15
109	Pd-109		0.125±0.016				
111	Ag-111		0.025±0.006	0.03±0.01		0.031±0.002	0.035±0.007
113	Ag-113g		0.019±0.009*				
115	Cd-115m		0.019±0.004*	0.06±0.01		0.022±0.002	0.0326
125	Sn-125g		0.048±0.015*	0.11±0.04			
127	Sb-127		0.297±0.028	0.19±0.03			
128	Sn-128	0.392±0.013					
129	Sb-129	0.940±0.035		0.89±0.05			
131	I-131	3.06±0.10		3.23±0.06	3.58±0.04		
132	Te-132	4.89±0.16		4.35±0.07	4.82±0.04		5.35±0.50
133	I-133	6.67±0.23		6.96±0.10			
134	Te-134	7.40±0.27		6.53±0.05			
135	I-135	6.59±0.22		6.03±0.04			
138	Xe-138	6.71±0.31					
140	Ba-140	6.00±0.22		6.22	6.09±0.07	6.0±0.5	5.0±0.4
141	Ce-141		5.67±0.32	6.40±0.05			6.1±0.4
143	Ce-143	5.60±0.18		5.89±0.07	5.37±0.04		
144	Ce-144		5.12±0.16	5.91±0.06			
146	Pr-146	3.51±0.18					
147	Nd-147	2.32±0.08	2.33±0.08	1.95±0.06			
149	Nd-149	1.01±0.07		1.23±0.09			
151	Pm-151	0.381±0.02		0.37±0.05			
153	Sm-153		0.154±0.02	0.16±0.05			
156	Eu-156		0.0186± 0.002				
161	Tb-161		0.000687± 0.00006				

GS: γ--Spectrometric method
RC: Radiochemical method
* : Total chain yield caculated by using the isomer ratio from Ref.8,9.

elements to be determined. Under slightly boiling the solution was reflexed for half an hour to reach the isotope exchange completely. In order to save the targets and irradiation time a procedure mainly based on anion exchange was developed to sequentially separate Ag, RE, Sr, Ba, Cd, Zr, U, Te, Mo, Sn and Sb. Ru and Pd were not included in the sequential procedure owing to their complex behavior.

Standard radiochemical procedures were used to separate and purify the samples for measurement. Chemical yields for separated samples were obtained, with one exception, by gravimetrically measuring the carrier quantities in the final samples. The exception, RE(Pr,Nd,Tb Sm,Eu), was yielded by complexometric titration. Most of final samples were mounted as precipitates on the teflon counting dishes. For nuclides of rare earth elements the final samples were eletrodeposited on gold disks.

The β or γ activity of the samples purified was counted. Two low background beta counters used in the present work were a 2π-gas-flow-proportional counter and a 2π-plastic-scintillation counter. The background rate was about 2~4 cpm. The gamma activity was measured by a well type NaI(Tl) (3"×3") anticoincidence spectrometer. Except for the nuclides with a longer life time, counting lasted over two to four half lives.

The absolute efficiencies for detected fission products were determined as a function of precipitate thickness using standardized high specific activity solution with $4\pi\beta$-γ coincidence counting. The efficiencies of nuclides unavailable as standardized sources (for instance, Br-84 and Tb-161) were estimated from a general curve relating efficiency to mean β-particle energy or to γ-ray energy.

Calculations

The observed fission rate and fission product activities were corrected by various factors and then the fission yields were obtained. The calculation details were described in the earlier paper /7/ and omitted here.

Results and Discussion

In order to obtain better statistical results, the determination of all the fission yields were repeated several times. Six targets were irradiated for radiochemical measurements and seven targets for direct gamma spectrometric measurements. Over 300 spectra were obtained. The cumulative fission yields for 40 mass chains in the region mass A= 84 - 161 are presented in Table 1. These 40 measured mass chains make up about 128% of the total of 200% expected from fission. The

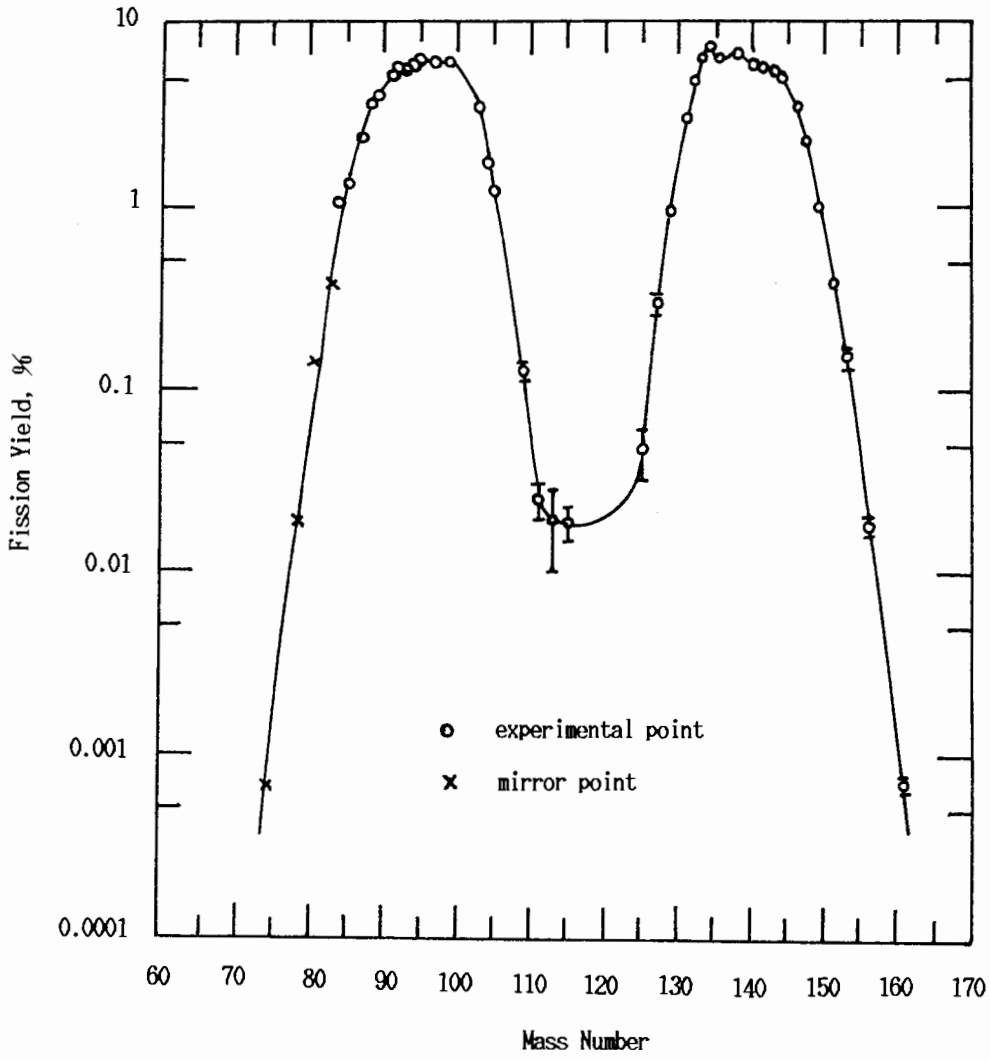

Fig. 2. Mass distribution in U-235 fission by fission sprctrum neutrons

overall error listed in Table 1 is the square root of the sum of the squares of the standard deviation of the average and the errors of fission rate, counting efficiency, chemical yield, γ- ray spectrum analysis with the computer program and correction for γ-ray self-absorption in the uranium target. The errors due to decay schemes are not included.

Prior to measuring fission yields of the fission spectrum neutron induced fission we radiochemically measured the yields of Zr-95, Mo-99 and Ce-144 in the thermal neutron fission of U-235.

The data obtained 6.39±0.21, 6.08±0.16 and 5.34±0.34 percent respectively, are in very good agreement with the compilation values /9,10/. From this agreement, we conclude that in this study the experimental technique is reliable and there is no serious systematic error.

The yields measured in this work are compared in Table 1. with all the known existing literature values reported to date. Experimentally determined fission yields are reported for the first time for mass chains 84, 94, 104, 109, 113, 128, 138, 146, 156 and 161.

From Table 1. it can be seen that most of our values are in reasonable agreement with the literature values with taking into account the errors attached to the data. However, for a few nuclides the differences are beyond the error limits.

The data presented in Table 1. are plotted in Fig.2. Owing to the lack of yields below mass 83 the mirror points of the heavy peak have been used to define the slope of the left wing of the light peak. The area under the mass distribution curve in Fig.2 was determined to be 199.1% which agrees remarkably well with 200% expected. As a result the yield data were not renormalized. The mean masses of the light and heavy groups are located at 95.2 and 138.6 respectively. The sum of these masses (233.8) corresponds to a value of 2.2 for $\bar{\nu}$ (the average number of neutrons per fission). The peak-to-valley ratio is 340. As can be seen clearly from Fig.2, a fine structure peak exists at the mass 134.

A literature servey reveals that for peak nuclides , such as Zr-95, Ba-140 and Ce-144 etc., there is not much to choose between the fission yields of fission spectrum neutrons and fast reactor neutrons induced fission of U-235. But the same is not true for Mo-99. These data are listed in Table 2. A comparison of these data indicates that the difference between the two fission systems really exists. In fast reactor fission the average of the yields is about 5.8 that is obviously lower than thermal fission yield, while in fission spectrum they are near or slightly higher than the thermal yield. Considering this difference it is not suitable that the Mo-99 yield from fission spectrum fission was about 5.7 which was evaluated by some authors /9,10/.

Table.2 Mo-99 fission yields (%)

U-235 fission induced by	
Fission spectrum neutron	Fast reactor neutron
5.9 ± 0.4 /1/	5.9 ± 0.1 /11/
6.4 ± 0.4 /2/	5.80 ± 0.12 /12/
6.08 ± 0.08 /3/	5.43 ± 0.22 /13/
6.22 ± 0.15 /4/	5.46 ± 0.18 /14/
6.36 ± 0.17	5.70 ± 0.17 /15/
/ This work /	6.23 ± 0.13 /16/
	5.41 ± 0.42 /17/
	6.40 ± 0.12 /18/

The reason why the case of Mo-99 is different from that of the other peak nuclides is not known. Further investgation is essential since Mo-99 is generally used as an internal standard in fission yield determinations.

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