Cumulative mass yields in the neutron-induced fission of ^{239}Pu at the resonance energy of 0.3 eV†

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Radiochemical techniques were used to measure the mass distribution of products in the fission of ^{239}Pu induced by neutrons at the 0.3 eV resonance. Cd and Gd filters produced neutron spectra suitable for these measurements. The valley-to-peak ratio thus obtained is 0.65 of that in fission induced by thermal neutrons.

[NUCLEAR REACTIONS, FISSION ^{239}Pu(n,f), $E=0.3$ eV; measured yields of Sr, Ag, and Cd isotopes.]

The investigation of fission phenomena in the 0.3 eV resonance of ^{239}Pu is of importance for scientific as well as for technical reasons: (i) In the energy range of slow neutrons, the fission of ^{239}Pu is strongly influenced by the resonance at 0.3 eV. Thus, at a neutron temperature of 300 K, fission is mainly affected by the low-energy tail of this resonance, whereas at higher temperatures (>1000 K), ^{239}Pu fissions to a considerable part in the maximum of the resonance. Therefore, the resonance at 0.3 eV plays a different role in high temperature reactors. (ii) The valley-to-peak ratio (VPR) of the mass distribution of ^{239}Pu varies greatly from resonance to resonance in the experimentally investigated range of neutron energy (<200 eV).[1] The VPR for the 0.3 eV resonance has already been studied several times (Refs. 2–4). However, the results of these experiments are either not consistent or are difficult to compare since they were obtained under different experimental conditions. Therefore, chain yields for masses 91 and 111–118 were redetermined in this work.

Targets of 0.5–1 mgPuO_2 (90.83% ^{239}Pu, 8.19% ^{240}Pu, 0.91% ^{241}Pu, and 0.07% ^{242}Pu) were irradiated for 1–2 hours in the core of the swimming pool reactor SAPHIR at the Swiss Reactor Institute. The targets were thin enough to prevent self-absorption effects. Two irradiation positions (1 and 2) with identical flux conditions were used. The cadium ratio and the neutron temperature were 2.5 and 295 K, respectively. A ^{239}Pu target was irradiated in position 1 in a cylindrical Gd-Al container with a thickness of 5.2×10^{20} atoms Gd cm^{-2}. The thermal neutron flux ($E_n<\sim0.1$ eV) was almost completely suppressed with this filter. However, neutrons with energies $>\sim0.2$ eV could pass through the filter without serious attenuation. Thus, fission was induced in the target mainly in the 0.3 eV resonance but to a considerable extent also in the resonances at higher energy. In order to determine the contribution to the fission rate by the more energetic neutrons, a second ^{239}Pu target was simultaneously irradiated in position 2 in a cylindrical Cd container having a thickness of 4.6×10^{21} atoms Cd cm^{-2}. This thickness was sufficient to suppress strongly fission in the 0.3 eV resonance as well as at lower neutron energies.

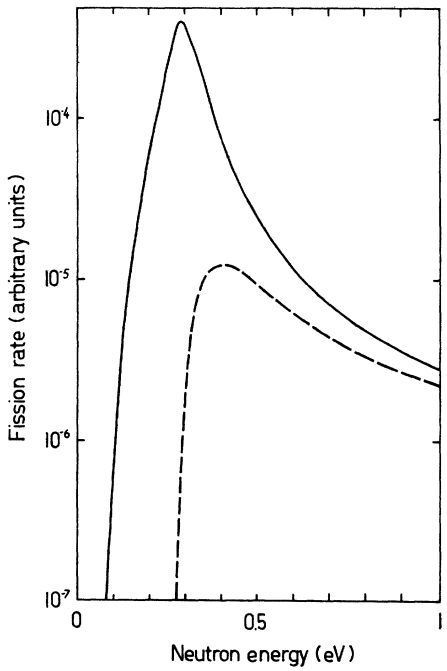

FIG. 1. Calculated fission rates are plotted as a function of the neutron energy. The solid and the dashed lines represent fission rates in Gd and Cd filters, respectively.

TABLE I. Nuclides and chain yields in the fission of ^{239}Pu for different neutron energies.

Nuclide	Fraction of chain yield (%)	Precursors considered	γ rays used Energy (keV)	γ rays used Absolute intensity (%)	Chain yield (this work) 0.3 eV[a] (%)[b]	Chain yields (Ref. 5) Thermal neutrons (%)	Chain yields (Ref. 5) Fission neutrons (%)
^{91}Sr	99.7 (Ref. 5)		556	58 ± 4 (Ref. 9)	2.4 ±0.2	2.49 ±0.03	2.43 ±0.07
^{111}Ag^{g}	99.7 (Ref. 5)	^{111}Pd^{g}	342	6 (Ref. 10)	0.28 ±0.04	0.27 ±0.03	0.35 ±0.01
^{112}Ag	100.0 (Ref. 5)	^{112}Pd^{g}	617	43 (Ref. 11)	0.13 ±0.02	0.107±0.004	0.193±0.005
^{113}Ag^{g}	81.05 (Ref. 8)		298	10.8 (Ref. 8)	0.076±0.01	0.078±0.005	0.127±0.003
^{115}Cd^{g}	86.0 (Ref. 5)	^{115}Ag^{g}	528	30±3 (Ref. 12)	0.024±0.006	0.037±0.001	0.080±0.005
^{117}Cd^{g}	100.0 (Ref. 5)		273	27 (Ref. 13)	0.018±0.004	0.034±0.004	0.065±0.007
^{117}Cd^{m}			564	4.2 (Ref. 14)			
^{118}Cd	99.8 (Ref. 5)		1230[c]	5.0±2.0 (Ref. 15)	0.030±0.018	0.034±0.005	0.065±0.007
^{140}Ba	99.8 (Ref. 5)		537	23.8±1.2 (Ref. 16)	...	5.58 ±0.06	5.25 ±0.07

[a] Measured in the 0.3 eV neutron resonance of ^{239}Pu.
[b] Based on the yield of ^{140}Ba (Ref. 5).
[c] γ ray of ^{118}In^{g} measured in the transient equilibrium ^{118}Cd/^{118}In^{g}.

However, neutrons with energies >~0.5 eV were not significantly absorbed. The quantity r is defined as the ratio of the integral fission rate in the Cd-shielded target to that in the Gd-shielded target, normalized to the same reactor neutron flux, and was determined from the ratio of the activity of the fission product ^{140}Ba in both targets. This nuclide was separated from the targets by use of radiochemical techniques (see below). The yield of chain 140 (5.58± 0.06% Ref. 5) can be assumed to be nearly constant in the energy range of interest. The value of r was found to be 0.29.

The quantity r was also calculated using a simple model.[6] The results of this calculation agree within errors with the experimental data. This shows that the influence of the resonances >1 eV of Cd and Gd on the integral fission rate of the ^{239}Pu targets was low. The low-energy part of the calculated fission rates in both filters as a function of the neutron energy is shown in Fig. 1. The burn-out of the filters during the irradiation was calculated to be small.

The temperature of the targets was checked during the irradiation using radiation resistant temperature foils. It did not exceed 500 K. Calculations showed that the influence of the Doppler broadening of the 0.3 eV resonance on our results can be neglected.

After the irradiation the targets were dissolved, Ag, Cd, Sr, and Ba were separated and purified using standard carrier procedures (Ref. 7). The samples were counted on calibrated GeLi γ ray spectrometers. The absolute intensities of the γ ray lines used are given in Table I.

The measured activities were corrected for growth and decay of the fission products during and after the irradiation. The yields Y_{Gd} and Y_{Cd} of a chain c in the Gd- and Cd-shielded targets could be determined relative to chain 140 by use of the yield of chain 140 for thermal neutrons given by Ref. 5 (see above). The chain yield Y_c due to fission in the 0.3 eV resonance was then calculated by the relation

$$Y_c = \frac{Y_{Gd} - rY_{Cd}}{1 - r}\,\%.$$

The chain yields in the 0.3 eV resonance (mean of two experiments) are shown in Table I. The errors in the chain yields (1σ) include uncertainties in the measurements, in the detector calibration, in the decay properties of the nuclides, in the decay and growth corrections, and in the yield of chain 140. The large error of chain 118 is mainly due to the uncertainty in the intensity of the γ ray line used (see Table I).

The yield ratio ^{115}Cd^{g}/^{99}Mo is used to obtain the valley-to-peak ratio (VPR) of the mass distribution. The relation $Q = \mathrm{VPR}_{0.3\ eV}/\mathrm{VPR}_{thermal}$ was calculated assuming that the yield of chain 99 given by Ref. 5 is constant over the energy range of interest. Q is shown in Table II together with values from the literature.[2-4] Our result for Q agrees well with that of Ref. 3 but is inconsistent with the data obtained by Refs. 2,4. The results of Ref. 4 are difficult to compare with our value of Q since

TABLE II. Values of $Q = \mathrm{VPR}_{0.3\ eV}/\mathrm{VPR}_{thermal}$.

Q	References
0.33 ± 0.03	Regier *et al.* (Ref. 2)
0.66 ± 0.05	Van Assche *et al.* (Ref. 3)
0.9 ± 0.1[a]	Toraskar and Melkonian (Ref. 4)
0.65 ± 0.15	This work

[a] Thermal value of VPR taken from Ref. 5.

they were obtained by another experimental technique (counter experiments). Furthermore, uncertainties in the conversion of the VPR value of Ref. 4 to a Q value may contribute to the observed discrepancy. The VPR in the 0.3 eV resonance is significantly lower than the VPR for thermal or fast neutron-induced fission (see Ref. 5). As was pointed out in Ref. 17 our value of the VPR is compatible with a spin assignment of zero for the 0.3 eV resonance as proposed in Ref. 18. At a neutron energy of 0.3 eV fission proceeds mainly (~78%) through the partially open 0^+ channel, which has a very low valley-to-peak ratio (Ref. 17).

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