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NUCLEAR DATA EVALUATION OF ACTINIDES RELEVANT TO THORIUM FUEL CYCLE

Takaaki Ohsawa

Atomic Energy Research Institute, Kinki University
Kowakae, Higashi-osaka 577, Japan

Masaharu Inoue

Department of Nuclear Engineering, Kyushu University
Hakozaki, Higashi-ku, Fukuoka 812, Japan

Abstract: Neutron nuclear data evaluation has been completed for Th-232, Pa-231 and Pa-233. Evaluation was made relying on the experimental data for the fission and capture cross sections and resonance parameters of Th-232. Theoretical models were employed if there were no or sparse data available. The neutron inelastic scattering leading to the excitation of strongly coupled states were calculated combining the coupled-channel and Hauser-Feshbach methods by means of the generalized transmission coefficients. The radiative capture cross sections for Pa-231 and Pa-233 were calculated with the statistical model. The (n,2n) and (n,3n) reaction cross sections were obtained using the Segev-Fahima model which allowed the preequilibrium effects in a simplified manner. The average prompt and delayed neutron multiplicities for Pa-isotopes were calculated using semi-empirical relations.

(evaluation, Th-232, Pa-231, Pa-233, various cross sections)

Introduction

The development of thorium fuel cycle gives rise to increasing requirements for the nuclear data of nuclides relevant to the main fissile and fertile nuclides, U-233 and Th-232, as well as of those nuclides produced from these materials by neutron reactions. Protactinium-233, for example, is a long-lived isotope with the half-life of 27.0 days and with large capture cross section. Neutron capture by Pa-233 is harmful in two senses, because it results in a loss of a neutron and a potential fissile nuclide at the same time.

Another protactinium isotope Pa-231 is also important since it is produced by way of (n,2n) reaction of Th-232 and leads to production of U-232 by neutron capture and subsequent beta-decay. (Note that the (n,2n) reaction cross section for Th-232 is greater than that for U-238 by several tens of percents, meaning that the reaction is more important in thorium fuel cycle.) Uranium-232 is known as the starting point of a decay chain of nuclides, among which are highly radioactive nuclides such as Bi-212 and Tl-208.

A complete nuclear data base is essential in estimating the buildup of the rare isotopes in a reactor as well as in analyzing the problems pertinent to transportation, reprocessing and refabrication of the fuel materials. This report describes the methods and results of evaluation of nuclear data for Th-232, Pa-231 and Pa-233.

Thorium-232

The previous evaluation made by the present author/1/ and incorporated into JENDL-2/2/ has been used and tested in reference to recent differential data and integral analyses. The results were reviewed and necessary modifications were made. We discuss below only those data that have been revised in the present work, so the reader is referred to our previous publication/1/ for information not described in this report.

Thermal Region

Newer measurements of the thermal neutron capture cross section yielded values not greatly deviated from our previous(also ENDF/B-V) evaluation 7.4 b(7.35±0.21 b by Kobayashi/3/, 7.41±0.08 b by Chrien/4/, 7.33±0.06 b by Jones/5/, 7.33 b by Poenitz/6/), so the value was not altered. The total cross section was adjusted so as to reproduce the Rayburn's value 13.28 b at 1.44eV/7/.

Resonance Region

New measurement by Kobayashi/8/ is in excellent agreement with another measurement by Olsen/9/ and has revealed that the Rahn's values/10/ of the resolved resonance parameters, adopted in our previous evaluation, were underestimated. Thus the data were revised according to the following principles: (a) For the 22 resonances Kobayashi measured, we adopted his values; (b) We assigned Kobayashi's average value 24.7 meV in place of the Rahn's value 21.2 meV to those resonances whose gamma widths were not known; (c) Keeping the negative energy resonances unchanged from the JENDL-2 evaluation, only the coefficient of a 1/v-shaped correction term to the capture cross section was adjusted.

Fast Region

Fission Cross Section New measurement by K.Kanda et al./11/ yielded values a bit higher than our previous evaluation, but there were no significant differences in shape and magnitude. The result of integral test by Ganesan/12/ revealed that calculation using the JENDL-2 data, among other evaluations, gave the best agreement for the capture-to-fission ratio in fast reactor assembly. Therefore, our previous evaluation was adopted without change.

Capture Cross Section There seemed to be no need for modification in the capture cross section in the fast region.

(n,2n) and (n,3n) Cross Sections The benchmark analysis/12/ showed that our previous evaluation gave the best agreement to the measured (n,2n)-to-fission ratio in fast reactor assembly. This confirms that, at least, the spectrum-averaged (n,2n) cross section of JENDL-2 is sufficiently good without further modifications.

Inelastic Scattering Cross Section We applied the method of unified calculation of direct and compound inelastic processes (CC/HF method) developed by the present authors/13,14/. This method consists of two steps: (i) the direct inelastic scattering component and the generalized transmission coefficients $\tilde{T}$ was calculated using the coupled-channel(CC) model code JUPITOR-1/15/ together with the CC optical potential parameters tuned to Th-232/16/; (ii) the Hauser-Feshbach(HF) calculation was made to obtain the compound-nuclear(CN) component of inelastic scattering using the $\tilde{T}$'s calculated with CC model for the entrance channel and the transmission coefficients T calculated with the spherical optical model(SOM) for the exit channels. The code ELIESE-3/17/ was used in the second step calculations with some modifications. The ground-state rotational levels 2+ and 4+ were assumed to be strongly coupled to the ground state 0+. One of the important findings of this method was that the behavior of the generalized transmission coefficients was significantly different from that of the SOM-transmission coefficients, and this resulted in higer CN formation cross section.

As for the inelastic scattering to the vibrational levels lying at several hundred keV, the direct component was calculated, when necessary, applying the asymmetric distorted-wave approximation(ADWA)/18/ in which a SOM potential representing the scattering cross sections was used for the entrance channel and a 'bare' optical potential was used for the exit channels.

The total inelastic scattering cross section obtained in this evaluation is shown in Fig. 1.

Elastic Scattering Cross Section The elastic scattering cross section was obtained by subtracting partial reaction cross sections from the total cross section. The differential elastic scattering was calculated with the CC/HF method.

Fission Neutron Multiplicity

One of the problems concerning the energy-dependence of prompt neutron multiplicity ν_p for Th-232 is wether the negative slope of $\nu_p(E)$ below 1.6 MeV observed in some experiments is actual or not. New measurement by Howe/19/ in the energy range from 1.1 to 49 MeV confirmed this trend down to 1.1 MeV. Thus we considered that the peculiar behavior is the real one and adopted the JENDL-2 value/1/ without change:

$$\begin{aligned}\nu_p(E) &= 3.653 - 1.000E_n \quad \text{for } E_n \lesssim 1.57 \text{ MeV}\\ &= 1.847 + 0.1515E_n \quad \text{for } 1.57 < E_n < 20 \text{ MeV}\end{aligned} \tag{1}$$

The delayed neutron multiplicity was estimated applying the systematics of Tuttle/20/.

Energy Spectrum of Secondary Neutrons

Use has been made of the code PEGASUS/21/ to calculate the secondary neutron spectrum taking into account the effect of preequilibrium emission of neutrons in inelastic scattering.

Protactinium-231

This nuclide was not included in the earlier versions of JENDL, so new evaluation was made.

Fast Region

Total Cross Section No measurements have been done of the total cross section in the fast region. Hence optical model calculation was made using the potential parameters for Th-232/1,16/.

Fission Cross Section Below 3 MeV, there were three measurements by Muir/22/, Williams/23/ and Dubrovina/24/. Above this energy, measurements were made by Kobayashi/25/, Plattard et al./26/, Fursov/27/ and Iyer/28/. These data were grouped into two families: the higher one (Kobayashi, Muir, Williams) and the lower one(Plattard, Fursov). Among these we adopted the data of Plattard et al. up to 12 MeV, since the recent measurement by Fursov et al. relative to Pu-239(n,f) reproduced well the structure in the region below 8 MeV. Above 12 MeV we extrapolated the cross section using the curve of Mann/29/. This resulted in the fission cross section curve different from Drake's evaluation /30/, the latter being based on the systematics observed between the fission cross section value at 3 MeV and the parameter $Z^{4/3}/A$, and lacking of the step-like increase around 7 MeV caused by second-chance fission. The fission-neutron spectrum averaged cross section calculated using the present evaluation and the Watt formula/31/ was 844 mb, which was in agreement with the measured value of Williams/23/ but lower than that of Kobayashi/25/ by 20%. This suggests that there still remains some uncertainty in the quantity.

Capture Cross Section No experimental data were available. Statistical model calculation including the Moldauer effects was made using the code CASTHY/32/. The average resonance parameters $<\Gamma_\gamma>$ =40 meV and $\langle D\rangle$=0.47 eV/33/ were used to normalize the cross section.

(n,2n) and (n,3n) Cross Sections We calculated the cross sections employing Segev-Fahima's prescription/34/. This method, as was originally given, required an experimental data to normalize the excitation function. Since no measured data were available for Pa-231, we contrived to combine the method with the optical model and other available data. Thus for instance the compound and preequiliblium components of the cross section were calculated as follows:

$$\begin{aligned}\sigma_c &= (\sigma_R - \sigma_f)[\sigma_c / (\sigma_c + \sigma_p)]\\ \sigma_p &= (\sigma_R - \sigma_f)[\sigma_p / (\sigma_c + \sigma_p)]\end{aligned} \tag{2}$$

where the values of the fractions in [ ] were taken from Th-232(n,2n) analysis assuming that the quantities were not so different for neighboring actinides. The reaction cross section σ_R were taken from SOM and CC calculations and the two results were compared in the form of fission-neutron-spectrum averaged cross sections in Table 1. This result showed that the reaction cross section from CC calculation gave better agreement to the recent measurement by Hashimoto/35/, the SOM calculation tending to be smaller. This provides another support to the use of CC model in estimating the reaction cross section for actinides. Figure 2 shows the present evaluation compared with an older one/30/.

Fission Spectrum	Calculated Value		Measured Value / Ref.35/
	σ_R(CC)	σ_R(SOM)	
Watt	5.75 mb	3.33 mb	5.38±0.43 mb
Cranberg	5.23	3.03	(older data:
Maxwell	6.00	3.47	4.45 mb)
CSEWG	6.48	3.75	

Table 1. Fission-spectrum averaged (n,2n) cross sections for Pa-231

Inelastic Scattering Cross Section The inelastic scattering cross section was calculated using the CC/HF method. The level scheme was taken from Ref.36 and three low-lying states 3/2-

,5/2- and 7/2- were coupled. The levels were assumed to form a continuum above 0.38 MeV. The level density parameters were taken from Gilbert and Cameron/37/. The total inelastic scattering cross section is shown in Fig. 3. The structure observed in the several hundred keV region is a reflection of that in the fission cross section.

Fission Neutron Multiplicity

No measurements were made. The semiempirical method of Bois and Frehaut/38/ were employed to obtain the ν_p-value for Pa-231:

$$\nu_p\,(E_n) = 2.251 + 0.1341(E_n - 0.6) \qquad (3)$$

where E_n is in MeV. The delayed neutron multiplicity was estimated using Tuttle's method/20/.

Angular Distribution of Secondary Neutrons

The differential elastic scattering cross sections were calculated with the CC/HF method. For inelastic scattering, deviation from 90°-symmetry was obtained for the strongly coupled states. The anisotropy increased with increasing energy. For other levels, the 90°-symmetric distribution as obtained from CASTHY calculation was employed. The emitted neutrons in (n,2n) and (n,3n) reactions were assumed to be isotropic in the center-of-mass system.

Protactinium-233

In the preceding versions of JENDL, the evaluation of Pa-233 by Y.Kanda/2/ mainly relied on the evaluation of Drake/30/ except the (n,n') and some other cross sections. New independent evaluation was attempted by the present authors. Experimental data were even less for Pa-233 compared with Pa-231 so that we resorted to theoretical calculations, of which the tools were common between the two nuclides.

Fast Region

Total Cross Section No measured data were available. Optical model calculation was made to obtain the total cross section using the potential parameter set for Th-232/1,16/.

Fission Cross Section Though no experimental data were available for the fission cross section, measurements were made on the fission probability by means of (^{3}He,df) and (^{3}He,tf) reactions/39/. We thus exploited these data, together with the reaction cross sections calculated from CC, to produce simulated fission cross sections for Pa-233. The method of calculation was described elsewhere/40/. The result is shown in Fig.4. Evidently, the present evaluation is much smaller than that by Drake/30/ and have a remarkable structure near the (n,n') threshold, reflecting the structure in fission probability.

Capture Cross Section No experimental data were available. Use was made of the code CASTHY/32/ to calculate the capture cross section. The average resonance parameter values $\langle\Gamma_\gamma\rangle$ =40 meV and $\langle D\rangle$=0.79 eV/41/ were used to normalize the calculated cross section.

(n,2n) and (n,3n) Cross Section The same method as used for Pa-231 was applied to Pa-233. Figure 2 shows that the present result is much higher than Drake's evaluation/30/ due to the fact that the competing fission cross section was much lower in the present evaluation. The fission-neutron spectrum averaged cross section calculated using this evaluation was 10.75 mb (assuming the Watt spectrum/31/), which was about a fifth of a single measured value $\lesssim$50.2 mb/42/. However, we considered that the present evaluation was more reasonable because of the following two facts: (a) there was a possibility that neutron capture by a trace of Pa-231 contained in the sample (with σ_γ(2200m/s)=190 b) had produced additional amount of Pa-232, as the measurer admitted; (b) the present value agreed with the systematics of Sekine and Baba/43/, while the measured value was off from the general trend.

Inelastic Scattering Cross Section The CC/HF method was applied to estimate the inelastic scattering cross section. The level scheme was taken from Ref.36 with the spin of the 300.4-keV level replaced from 7/2- to 7/2+ as indicated by Gonzalez/44/. A continuum was assumed above 0.5 MeV. The present evaluation was generally much higher than the JENDL-2 value.

Fission Neutron Multiplicity

The prompt neutron multiplicity was estimated according to the method of Bois and Frehaut/38/. The result is expressed as follows:

$$\nu_p(E_n) = 2.251 + 0.1403(E_n - 1.0) \qquad (4)$$

The delayed neutron multiplicity was obtained using the systematics of Tuttle/20/. The result was ν_p =0.0228 without any energy dependence.

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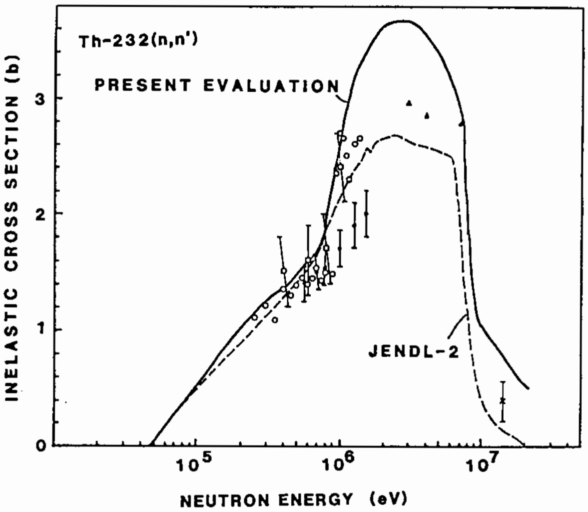

Fig.1 Inelastic scattering cross section for Th-232.

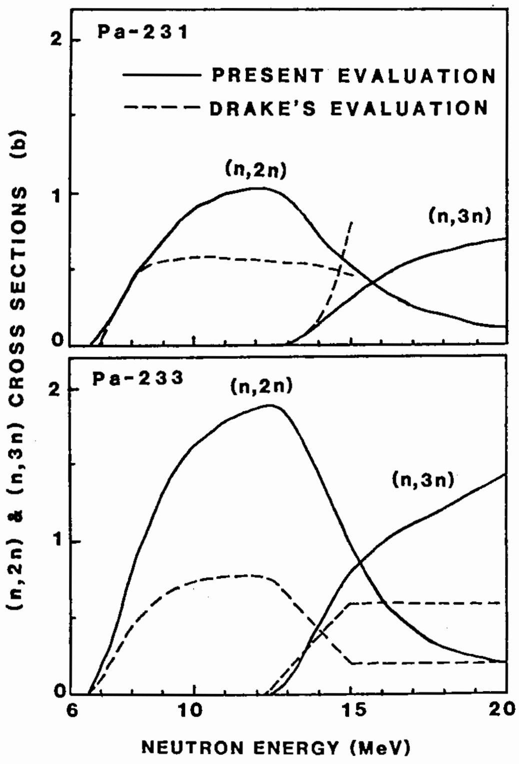

Fig.2 (n,2n) and (n,3n) cross sections for Pa-231 and Pa-233.

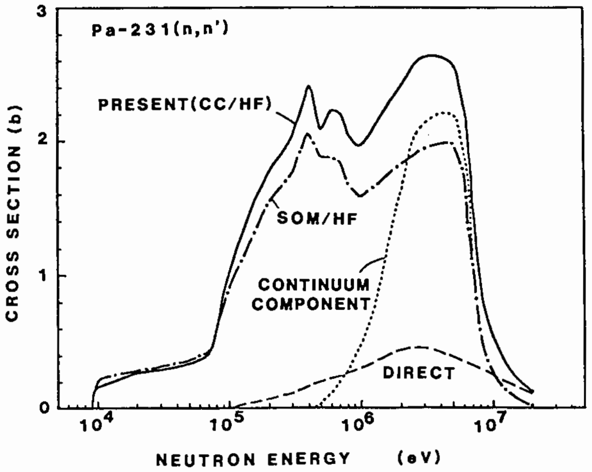

Fig.3 Inelastic scattering cross section for Pa-231.

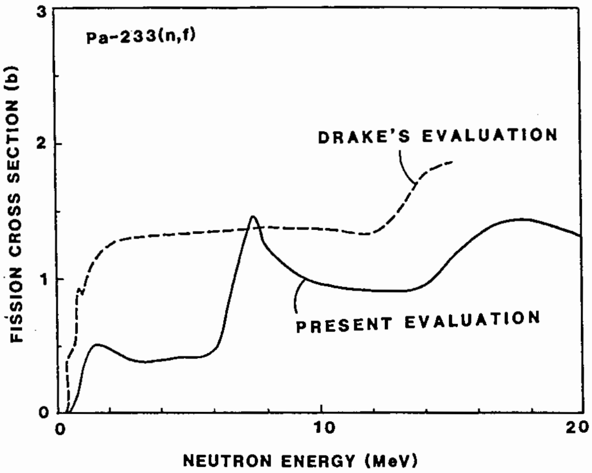

Fig.4 Simulated fission cross section for Pa-233 compared with Drake's evaluation.