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ATOMIC ENERGY COMMISSION

THERMAL AND SIXTEEN GROUP CROSS SECTION DATA FOR Pa^{233}, U^{234} and U^{236}

by

R. Shankar Singh
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Abstract

Latest available nuclear data have been collected for Pa^{233}, U^{234} and U^{236} and recommended values of thermal cross sections and resonance integrals have been given for these isotopes for use in thermal reactor calculations in the analysis of thorium fuel cycles. Sixteen-group cross sections for studying fast systems based on the same fuel cycle, have been prepared. Accuracies in the present data and the need for further measurements and improvements have been specified.

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1. INTRODUCTION

Nuclear data for reactor calculations are required not only for the materials which are used in a reactor to start with, but also for such elements or isotopes which are produced in a reactor during its operation. Such data are essential when one studies the long term behaviour of a reactor like the fuel burn-up estimates or the reactivity changes during operation etc. Availability of cross section data for the heavier isotopes of fuel has been limited due to several reasons and attemps are being made to make measurements in the existing gaps to reduce the errors involved in making rough estimates or guesses in certain energy regions. The users of nuclear data have to keep in touch with the latest information on them and also have to 'prepare' the data in the manner required by them as and when such information becomes available.

The present work is part of such an attempt to prepare the cross-section data for the isotopes Pa^{233}, U^{234} & U^{236} which are needed in the study of thorium fuel cycles in different types of reactors. Latest information available on thermal cross sections and resonance integrals has been presented for these isotopes and sixteen-group cross sections have been prepared for studying fast reactor systems, using the material collected from various sources such as CINDA[1], EANDC (European American Nuclear Data committee), UKAEA Nuclear Data Library, BNL compilations etc.

2. DATA FOR THERMAL REACTOR CALCULATIONS

The usual parameters required are the cross sections for capture (σc) and fission (σf), the resonance parameters and the resonance integrals for absorption and fission. These are discussed for the isotopes Pa^{233}, U^{234} and U^{236}, and tabulated in Table I.

2.1. Protactinium - 233

Most of the data for thermal neutrons (2200 m/sec) are from the latest BNL-325[2] compilation. Values selected here are those either recommended in this compilation or chosen from the other various reported values.

The recommended value of capture cross section, σ_c = 43 ± 5 barns, is from the different measurements which have been reported. The resonance capture integral measured at ORNL is = 920 ± 90 barns.

The resonances are measured upto 16 ev (16 resonances) but have been analysed for the resonance parameters only upto 5.152 ev.

The calculated resonance absorption integral comes out to be only ~ 500 barns[3]. Obviously more data on resonance parameters are required to overcome this large disparity.

There is no fission reaction for this isotope at thermal energies.

2.2. Uranium - 233

This isotope is also included here in thermal data to give the latest information available on it.

The cross sections recommended [2] here are those obtained from a least square fitting of the various measured values.

σ_c = 49 ± 6 barns ; σ_f = 524.5 ± 1.9 barns ;

σ_a = 573.1 ± 2.1 barns ; η =2.292 ± 0.006

The resonance integral values are:

RI (fission) = 746 ± 15 barns (0.45 ev cut off) RI(Capture) = 147 barns (sub-cadmium spectrum).

Resonances have been measured [2] upto 40 ev, but the resonance parameters have not been accurately determined because of the multi-level analysis required for them.

The least square value of α (= σ_c / σ_f) at En = .0253 ev, determined indirectly is given as α = 0.0926.

It would be of interest to note the epithermal (0.5 ev cut-off) values of α measured by different people here.

ORNL value [4] α = 0.17 ± .017

GE - KAPL value [5] α = 0.165 ± .012 (This source gives

RI (fission) = 820 $\pm$ 65 barns and

RI (capture) = 138 $\pm$ 10 barns.)

The latter value of α seems to be preferable after the scrutiny of the two sources.

2.3. Uranium - 234

The absorption cross section has been determined from the measured values of the total cross section (6). The recommended value (2) is σ_a = 95 $\pm$ 7 barns.

Leonard and Odegarden (7) have shown that the fission cross section at the 5.2 ev resonance is very small compared to the total cross section and the 2200 m/s value of σ_f is less than 60 mb.

Resonance parameters have been measured upto 369 ev (20 resonances) and Halperin (8) recommends for the resonance capture integral a value of 700 $\pm$ 70 barns. More measurements in support of this figure are perhaps necessary.

2.4. Uranium - 236

A number of measurements of the capture cross section in thermal energy region have been reported.
The recommended value is σ_c = 6 $\pm$ 1 barns.
Leonard and Odegarden (7) have shown that at the 5.49 ev resonance, the fission cross section is less than 100 mb while the thermal cross section is less than 0.2 mb.

Resonance parameters have been measured upto 384 ev (14 resonances) and the recommended values for the resonance capture integral based on activation measurements at ORNL by Halperin (8) is 400 $\pm$ 50 barns. The calculated value of resonance integral from level parameters is smaller and measurements for resonance parameters as well as resonance integral are needed to resolve the differences.

3. SIXTEEN-GROUP CROSS SECTIONS

Multigroup cross sections have been prepared by different laboratories for most of the materials used in fast reactors. For the isotopes Pa^{233}, U^{234} and U^{236}, however, there have been very few attempts. The main reason is the lack of sufficient experimental data for these materials at higher energies to prepare such sets. The situation is improving and measurements are being undertaken in the existing gaps for these isotopes. But it will be a long time before all the required data are obtained through experiments and hence theoretical estimates have to be taken recourse to. In the present compilation, the latest available experimental data, supplemented by calculated values, have been used. The sixteen group structure is the same as YOM[9]. The group cross sections have been prepared by weighting with a neutron energy spectrum corresponding to a Pu-fuelled matallic core of about 500 litres. The sources of different cross sections and their respective accuracies for the three isotopes have been discussed below. Sixteen-group cross sections are given in Table II, III and IV for Pa^{233}, U^{234} and U^{236} respectively.

3.1. Protactinium - 233

There is very little information available on this isotope so that most of the values selected are based on the estimates and guesses made by Goldman[10]. No measurements have been reported above 16 ev.

The fission cross section of Pa^{233} is expected to resemble closely that of U^{238} and therefore σ_f and ν (number of neutrons produced per fission) are taken to be the same as for U^{238} from a latest [11] compilation.

The capture cross sections (σ_c) have been chosen[10] as follows:

2 ev - 10 Kev : Average capture cross sections computed using statistical theory with average resonance parameters. 10 Kev - 100 Kev : From the smooth curve drawn between the adjoining regions.

100 Kev - 500 Kev : Values from Hauser - Feshbach calculations.

Above 500 Kev : Cross section extrapolated to very small values.

Elastic removal (σ_{er}) and transport (σ_{tr}) cross sections have been taken to be the same as for U^{238}.

Inelastic scattering (σ_{in}) cross sections are taken from the estimated values from Hauser - Feshbach calculations.

Inelastic transfer cross sections ($\sigma_{j \xrightarrow{S} j+k}$) have been computed using the new [12] transfer matrix $P_{j \longrightarrow j+k}$ for U^{238}.

3.2. Uranium - 234

A 22-group set was prepared for this isotope in earlier studies[13]. More information has become available since then and the present 16-group set includes the improved values of the fission and capture cross sections, compiled by Parker [14].

Fission cross section (threshold 50 Kev) has been taken from the data measured at ORNL by Lamphere[15] and the $\nu(E)$ values from Parker's [14] data.

No measurements of capture cross sections have been reported, but Hamilton's calculated values reported by Parker [14] in the 1 Kev - 1 Mev range have been used. Above 1 Mev the cross section is extrapolated to 1 mb for energies greater than 4 Mev.

Elastic removal (σ_{er}) and transport (σ_{tr}) cross sections have been taken from earlier compilation[13].

Inelastic scattering cross sections have been taken from Parker's compilation given in the form of a curve.

Inelastic transfer cross sections have been computed from the above values using the transfer matric $P_{j \rightarrow j+k}$ of U^{238}.

3.3. Uranium - 236

Parker [16] has compiled the data for this isotope in 1 Kev - 16 Mev energy range.

Fission cross section has been measured from 700 Kev to 7 Mev. at Los Alamos [17] and ORNL[15].

$\nu(E)$ values are also chosen from those recommended by Parker.

The capture cross section has been measured in the energy range 0.36 - 3.97 Mev by Barry et al [18]. In the energy range 1 Kev - 300 Kev, calculated values by Hamilton, reported by Parker [16] have been used.

Elastic removal (σ_{er}) and transport (σ_{tr}) cross sections have been taken to be the same as for U^{234}.

Inelastic scattering cross section has been taken from Parker's curve based on comparison with U^{238}.

Inelastic transfer cross sections have been computed using the transfer matrix $P_{j \longrightarrow j+k}$ of U^{238}.

Accuracy of the Present Data

Since no measurements have been made for Pa^{233} at higher energies, the value obtained by estimates and comparison with other elements may involve large errors and hence, for this isotope, the data need lot of improvements and is not the ultimate information.

For U^{234} and U^{236}, the fission cross sections should be accurate to $\pm$ 6% upto 6 Mev. The capture cross-sections, wherever the measurements are made, should not be in error to more than 10%, but in other regions (below 300 Kev) the errors may be larger. The scattering and transport data may be considerably in error due to the rough methods in their estimation.

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Table I. Thermal Data for Pa^{233}, U^{233}, U^{234} & U^{236}

Quantity *	Pa^{233}	U^{233}	U^{234}	U^{236}
σ_c	43 ± 5	49 ± 6	95 ± 7	6 ± 1
σ_f		524.5 ± 1.9		
σ_a		573.1 ± 2.1		
Resonance Integral (Capture)	920 ± 90	147	700 ± 70	400 ± 40
Resonance Integral (Capture)		746 ± 15		
α (= σ_c/σ_f)		0.0926 ± .0027(a)		
		0.165 ± .012 (b)		
η		2.292 ± 0.006		

* Cross sections correspond to 2200 m/s. values and all quantities except α and η are in barns.

(a) 2200 m/s. value.

(b) Epithermal (0.5 ev cut off) value.

Table II 16-Group Cross Sections for Pa^{233} (barns)

j	E_L (KeV)	σ_f	σ_c	σ_{er}	σ_{tr}	ν	σ_{in}	$\sigma_{in\,j\to k}$ $k-j=0$	1	2	3	4	5	6	7	8	9	
1.	3668	0.629	0.01	.022	4.05	3.13	2.5	0	.035	.245	.557	.658	.510	.282	.128	.060	.025	
2.	2225	0.573	0.02	.023	4.44	2.77	2.6	.008	.105	.408	.668	.637	.413	.211	.094	.041	.015	
3.	1350	0.425	0.05	.028	4.55	2.62	2.4	.462	.730	.885	.22	.033	.07					
4.	825	0.0272	0.11	.038	4.61	2.52	1.8	1.04	.521	.252	.079	.008						
5.	500		0.25	.062	5.44		1.5	1.05	.45									
6.	300		0.55	.091	5.76		1.05	.664	.386									
7.	180		0.72	.115	8.47		0.8	.398	.402									
8.	110		0.85	.136	10.22		0.6	.220	.221	.169								
9.	67		1.15	.151	11.68		0.32	0	.242	.044	.034							
10.	407		1.3	.155	12.58		0.1	0	0	.073	.027							
11.	25.0		1.5	.158	13.13		0											
12.	15.0		1.7	.166	14.3													
13.	9.1		1.95	.174	15.0													
14.	5.5		2.4	.174	15.1													
15.	2.1		2.8	.063	16.0													
16.	0.5		3.1	0	14.0													

Table III 16-Group Cross Sections for U^{234} (barns)

j	E_L (Kev)	σ_f	σ_c	σ_{er}	σ_{tr}	ν	σ_{in}	$\sigma_{inj \to j+k}$ K=0	1	2	3	4	5	6	7	8	9
1.	3688	1.55	.001	.019	3.98	3.2	1.53	0	.021	.150	0.341	.403	.312	.173	.078	.037	.015
2.	2225	1.52	.003	.029	4.5	2.87	1.64	.005	.067	0.256	0.426	.403	.259	.129	.062	.022	.009
3.	1350	1.45	.025	.03	4.68	2.68	1.62	.314	.494	.602	.146	.019	.045				
4.	825	1.20	.05	.048	5.16	2.38	1.38	.723	.393	.195	.063	.006					
5.	500	0.80	.043	.078	5.81	2.40	1.22	.858	.362								
6.	300	0.25	.042	.101	8.8	2.36	1.4	0.902	.498								
7.	180	0.06	.054	.126	7.9	2.30	1.3	.647	.653								
8.	110	0.04	.076	.147	10.2	2.28	1.0	.364	.350	.286							
9.	67	0.03	.11	.151	10.8	2.25	0.45	0	.329	.072	.049						
10.	40.7	0.02	.16	.158	11.6	2.24		0									
11.	25.0		.23	.171	12.9												
12.	15.0		.36	.171	14.0												
13.	9.1		.55	.180	14.3												
14.	5.5		.86	.175	15.07												
15.	2.1		1.5	.067	15.97												
16.	0.5		4.1	0	17.0												

Table IV 16-Group Cross Sections for U^{236} (barns)

j	E_l (Kev)	σ_f	σ_c	σ_{er}	σ_{tr}	ν	σ_{in}	$\sigma_{in\, j \to j+k}$ K=0	1	2	3	4	5	6	7	8	9
1.	3668	0.00	.015	.019	3.98	3.21	2.15	0	.0301	.211	.479	.566	.439	.242	.110	.052	.021
2.	2225	0.09	.06	.029	4.5	2.75	2.23	[illegible]	.091	.346	.573	.546	.351	.175	.082	.035	.014
3.	1350	0.74	.101	.03	4.68	2.64	2.17	.421	.662	.805	.195	.026	.061				
4.	825	0.4	.35	.048	5.86	2.54	1.86	.973	.530	.263	.086	.008					
5.	500	0.025	.29	.078	5.81	2.45	1.74	1.223	.517								
6.	300		.27	.101	6.8		1.61	1.037	.573								
7.	180		.25	.126	7.9		1.2	.598	.602								
8.	110		.23	.147	10.2		0.9	.328	.315	.257							
9.	67		.22	.151	10.8		0.5										
10.	40.7		.24	.158	11.6												
11.	25.0		.31	.171	12.9												
12.	15.0		.44	.171	14.0												
13.	9.1		.67	.180	14.3												
14.	5.5		1.05	.175	15.07												
15.	2.1		1.8	.067	15.97												
16.	0.5		4.5	0	17.0												