RECENT MEASUREMENTS OF FISSION NEUTRON YIELD DATA OF MINOR ACTINIDES

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ABSTRACT

An experimental study of the fission neutron yield data for Np-237, Am-241, and Am-243 is under way for both prompt and delayed neutrons. Preliminary results of this experimental program are discussed in this paper.

The fission neutron yields, ν, of Np-237 and Am-243, as well as U-235, were measured for 144 keV silicon-filtered neutron beam at University of Missouri Research Reactor. The measured values were as follows:

ν : Np-237 : 3.13±0.12 Am-243 : 4.00±0.35 U-235 : 2.54±0.06.

These values for minor actinides were about 20 % larger than those of ENDF/B-VI and JENDL-3.2, while that for U-235 was 4 % larger than the data files.

The delayed neutron data for Np-237, Am-241, and Am-243 were measured using fast pneumatic transfer system in Texas A&M University TRIGA reactor. The measured delayed neutron yields, ν_d, per 100 fissions in the thermal neutron spectrum were as follows:

ν_d (per 100 fissions) : Np-237 : 1.29±0.04 Am-241 :0.49±0.02 Am-243 : 0.84±0.04.

The ratios of the present values to those of the evaluated data files were 1.19, 1.14, and 1.05 for ENDF/B-VI, and 1.06, 1.09, and 0.88 for JENDL-3.2.

The 6-group data for the delayed neutron, λ_i and β_i, were also obtained from the experiment and compared with ENDF/B-VI. The measured decay constant, λ_i, agreed well with the data file. Large discrepancies were, however, found in the group fraction, β_i, especially for Am-243.

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1. INTRODUCTION

Fission neutron yield data of Np-237, Am-241, and Am-243 play important roles in a minor actinide (MA) burning reactor in which MA's are the major part of the nuclear fuel. The reliability of these data, however, seems insufficient for the design and the safety analysis of such a reactor. At present, new measurements[1),2)] are under way for both prompt and delayed neutron yield data of these nuclei. Preliminary results of this experimental program are to be discussed in this paper.

The accuracy of fission neutron yield, ν, greatly influences the criticality of the nuclear reactor. For Am isotopes, however, there was no experimental work on ν, while some experimental values are available for Np-237. In the present work, the neutron yields of Np-237 and Am-243 were measured for 144 keV silicon-filtered neutron beam at University of Missouri Research Reactor. The measurement for Am-241 was also attempted, but the reliable result can not be obtained because of the strong γ-ray emission from the sample.

The accuracy of delayed neutron yield and 6-group data, ν_d, β_i, and λ_i, greatly influences the reactor dynamics. For Np-237 and Am-241, Waldo et al.[3)] and Benedetti et al.[4)] measured these data in thermal and fast neutron spectra, respectively, while there was no experiment for Am-243. Waldo et al. tried to make measurements for Am-243, but the Am-241 impurity of their sample prevented them. In the present work, the delayed neutron data for Np-237, Am-241, and Am-243 were measured by using fast pneumatic transfer system in Texas A&M University TRIGA reactor. Measurement in the fast neutron spectrum is in progress and is to be reported elsewhere.[5)]

In the next section, the experimental methods are described for both the fission neutron yield measurement and the delayed neutron measurement. Then, the measured values are compared with those form the evaluated nuclear data files, ENDF/B-VI and JENDL-3.2.

2. EXPERIMENT

2.1 Fission neutron yield measurements

Experimental technique adopted here was newly developed to overcome the restriction of the amount of actinide samples. Proton recoil counters were used as the neutron detector, which can discriminate high energy fission neutrons from numerous scattered ones of 144 keV. The absolute fission rate was estimated by solid state track detectors. Both the proton recoil detectors and the track detectors were calibrated by Cf-252 fission source. The experimental arrangement is shown in Fig. 1. Details of the measurement were as follows.

Samples

Two kinds of samples were used in the measurement. One is the neutron emission sample for the fission neutron measurement, which had the dimension of $12.7mm^{\phi} \times 1.02mm^{t}$ and 100~300 mg of the oxide of the target actinide covered by titanium metal of $0.05mm^{t}$. The other is the fission rate sample, where the actinide was electroplated on platinum backing disk and covered by a gold or nickel layer. The samples contain the actinide of 0.08 ~ 2.3 mg.

The isotopic purity of both samples were 97.7 % for U-235, 99.999 % for Np-237 and 99.997 % for Am-243, respectively.

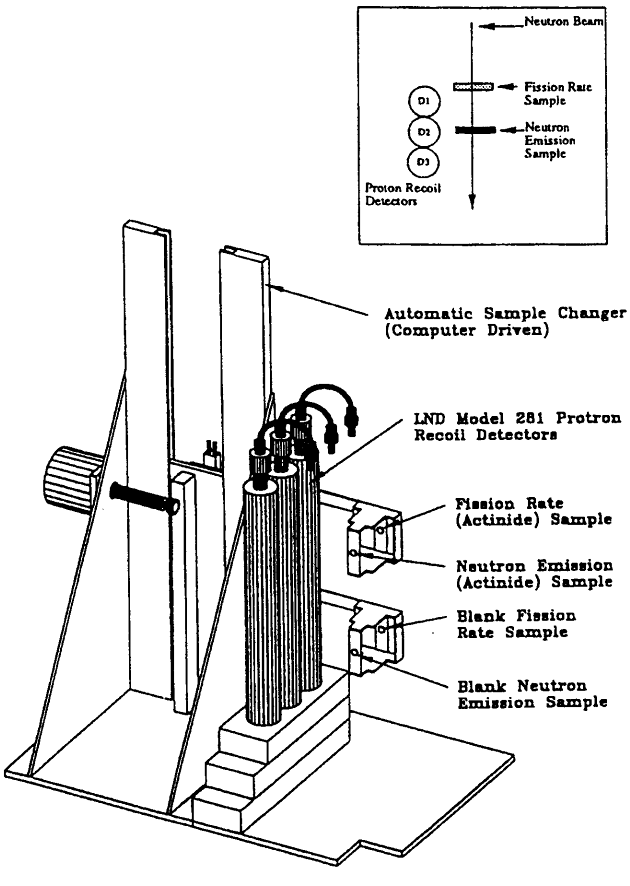

Fig. 1 Samples and detectors for fission neutron yield measurement

In addition to above actinide samples, blank samples and Cf-252 samples were also prepared for the background subtraction and the detection efficiency calibration, respectively.

Fission neutron measurement

A silicon-filtered beam from Missouri University Research Reactor[6] was used as the neutron source. To eliminate the affect of thermal neutrons, a cadmium sheet of 2 mm^t was put between the beam outlet and the sample. The neutron beam was collimated to 16 mm^{ϕ}.

Fission neutrons from the neutron emission sample were counted by three proton recoil detectors. To cut the 144 keV neutrons scattered by the sample, the discrimination level of the recoiled proton energy was set to 200 keV for U-235 and Np-237 and to 440 keV for Am-243. The higher discrimination level for Am-243 aimed to prevent signals due to the pile up of the strong γ -ray from the sample affecting the background counts. For the same reason, a lead sheet was placed between the sample and the detectors for the measurement of Am-243.

The detection efficiency was calibrated by using Cf-252 neutron source under the same experimental condition as the measurement of the actinide samples.

To obtain the number of the fission neutrons from the actinide, the background subtraction and the correction for the scattering of the actinide itself were done by using the count rate from the blank sample.

Fission rate measurement

A solid state track detector was attached to the fission rate sample and irradiated in the silicon-filtered beam simultaneously with the fission neutron measurement. The fission reaction was detected by the tracks of the fission fragments which penetrate the gold or nickel cover of the sample and go into the detector.

To obtain clear image of tracks, various detector material and various chemical etching processes were examined, and consequently, polycarbonate was chosen as the material and the optimum process was determined.

The detection efficiency was calibrated by Cf-252 sample covered by gold or nickel which has the same thickness as the fission rate sample.

Determination of ν

The ratio of the neutron emission rate to the fission rate, which were both calibrated to the absolute values, yields the number of the neutron emission per fission, ν .

2.2 Delayed neutron measurement

Experimental technique adopted here was similar to Keepin's.[7] The actinide samples used in the delayed neutron measurements were identical to those for the measurement of ν . The isotopic purity of Am-241 was essentially 100%.

Delayed neutron measurement

A TRIGA-type research reactor at the Texas A&M University Nuclear Science Center was used as the irradiation field. An existing

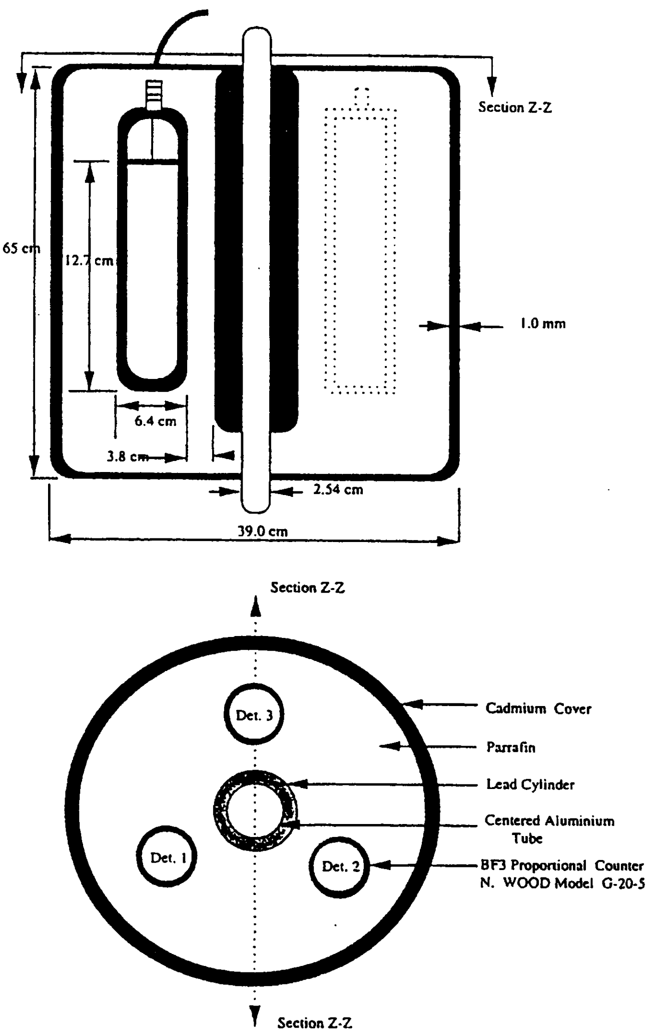

Fig.2 Sample receiver and detectors for delayed neutron measurement

pneumatic transfer system was revised to get enough speed to measure the delayed neutrons belonging to 1 ~ 5th. precursor groups ($T_{1/2}$=0.7 ~ 55sec).

Three BF_3 proportional counters were used for the detection of the delayed neutron emission. The actinide sample contained in a polyethylene vial was irradiated at the core receiver in the reactor and then transmitted to the sample receiver outside the reactor by the pneumatic system. The sample receiver and the BF_3 counters were surrounded by paraffin as shown in Fig. 2 to enhance the neutron detection efficiency. Three irradiation times were chosen to put emphasis on different precursor groups.

The efficiency of the detection system was calibrated by the delayed neutron emission from irradiated U-235 samples whose fission properties were considered to be well known.

Fission rate measurement

After the irradiation of each sample, γ -ray spectroscopy was performed to determine the absolute fission rate. The activities of Ba-140, La-140, Ru-103, I-131, and Mo-99 were measured. The detection efficiency of gamma-rays was calibrated by Eu-152 source depending on the energy of γ -ray. The cumulative fission yields quoted from ENDF/B-VI were used to convert the activities of FP's to absolute fission rates.

Determination of β_i, λ_i, and ν_d

To obtain the relative fraction and the decay constant of each precursor group, a least squares analysis was performed for the measured time attenuation curves of the delayed neutrons. Moreover, by combining the absolute neutron emission rate and the absolute fission rate, the number of delayed neutrons per fission was deduced. Calculated data was used for the 6th. group based upon Tuttle's empirical expression[8] and group-wise parameters given by Benedetti[4].

3. DISCUSSION

3.1 Fission neutron yield

The experimental results of fission neutron yields are shown in Table 1 together with those of the evaluated nuclear data files.

Table 1 Results of fission neutron yields ν (Incident neutron energy : 144 keV)

	Present work	ENDF/B-VI	JENDL-3.2
U-235	2.54±0.06	2.44	2.45
Np-237	3.13±0.12	2.66	2.56
Am-243	4.00±0.35	3.30	3.23

It can be observed that present work gives larger values than both files; 4% for U-235, about 20% for Np-237 and Am-243. The relatively good agreement for U-235 indicates the validity of the present experimental method. For Np-237, however, large discrepancy was observed, though better agreement had been expected since the nuclear data files were evaluated on the basis of previous experimental work. Therefore, some experimental bias may exist in the present measurement.

On the other hand, there has been no experimental data available for Am-243. Hence, if the bias in Np-237 is settled, the present measurement will become invaluable in the evaluation of the minor actinide nuclear data.

3.2 Delayed neutron data

The experimental results of absolute delayed neutron yields are compared with those of the previous measurements and the evaluated nuclear data files in Table 2.

Table 2 Results of delayed neutron yields ν_d (per 100 fission)

	Experimental work			ENDF/B-VI	JENDL-3.2
	Present	Waldo et al.[3]	Benedetti et al.[4]		
Energy range	Thermal	Thermal	Fast	Thermal - 4MeV	Thermal - 6MeV
Np-237	1.29 ± 0.04	1.07 ± 0.10	1.22 ± 0.03	1.08	1.22
Am-241	0.49 ± 0.02	0.51 ± 0.07	0.39 ± 0.02	0.43	0.45
Am-243	0.84 ± 0.04	———	———	0.80	0.95

Since ν_d is considered to be constant below several MeV, the experimental values and those of the data files in Table 2 are comparable in spite of the different energy range.

For Np-237, the present work shows better agreement with JENDL-3.2 and Benedetti than ENDF/B-VI and Waldo. For Am-241, however, the present work agrees with Waldo's measurement very well. On the other hand, for Am-243, there has been no experimental work, and JENDL-3.2 gives 13 % larger value than the present work, while ENDF/B-VI agrees with the present value within the experimental error.

Measured 6-group data are compared with those in ENDF/B-VI in Table 3, 4 and 5 for Np-237, Am-241, and Am-243, respectively.

For decay constants, there is no large discrepancy between the present work and ENDF-B-VI, except for 3rd. and 4th. group of Np-237 and 1st. group of Am-241.

On the other hand, large discrepancies are observed in the relative yields especially for 3rd. and 5th. group of Am-243. Therefore, after the completion of the experiment in the fast neutron spectrum, which is in progress, it is recommended that evaluators of the nuclear data files take this experimental work into consideration.

Table 3 Results of 6-group data for delayed neutron from Np-237

	Decay constant λ_i (s^{-1})		Relative yields β_i	
Group	Present work	ENDF/B-VI	Present work	ENDF/B-VI
1	0.0129 ± 0.0006	0.0133	0.040 ± 0.002	0.040
2	0.0324 ± 0.0010	0.0316	0.233 ± 0.017	0.216
3	0.1048 ± 0.0019	0.1168	0.190 ± 0.010	0.156
4	0.341 ± 0.013	0.301	0.322 ± 0.027	0.363
5	0.85 ± 0.06	0.867	0.193 ± 0.007	0.166
6	———	2.76	0.021[a)]	0.059

a) calculated

Table 4 Results of 6-group data for delayed neutron from Am-241

	Decay constant λ_i (s^{-1})		Relative yields β_i	
Group	Present work	ENDF/B-VI	Present work	ENDF/B-VI
1	0.0122 ± 0.0005	0.0133	0.036 ± 0.002	0.036
2	0.0318 ± 0.0016	0.0308	0.309 ± 0.015	0.254
3	0.111 ± 0.007	0.1131	0.195 ± 0.008	0.156
4	0.300 ± 0.017	0.287	0.331 ± 0.039	0.336
5	0.890 ± 0.023	0.865	0.110 ± 0.005	0.172
6	———	2.64	0.018[a)]	0.045

a) calculated

Table 5 Results of 6-group data for delayed neutron from Am-243

Group	Decay constant λ_i (s^{-1})		Relative yields β_i	
	Present work	ENDF/B-VI	Present work	ENDF/B-VI
1	0.0131 ±0.0002	0.0135	0.024 ±0.013	0.023
2	0.0311 ±0.0009	0.0298	0.291 ±0.011	0.294
3	0.107 ±0.007	0.1138	0.221 ±0.008	0.154
4	0.324 ±0.021	0.299	0.384 ±0.016	0.315
5	0.914 ±0.031	0.882	0.051 ±0.002	0.166
6	————	2.81	0.029[a)]	0.048

a) calculated

4. CONCLUSION

Preliminary results of the experimental program on fission neutron data for minor actinides are reported. The fission neutron yields, ν, of Np-237 and Am-243 were measured using a 144 keV silicon-filtered neutron beam at University of Missouri Research Reactor. The measured values were about 20 % larger than ENDF/B-VI and JENDL-3.2. The result for U-235 by the same experimental method shows 4% larger value than the data files.

The delayed neutron data for Np-237, Am-241, and Am-243 were measured by using a fast pneumatic transfer system at the Texas A&M University TRIGA reactor. The measured delayed neutron yields per 100 fissions in the thermal neutron spectrum were 1.29±0.04, 0.49±0.02, and 0.84±0.04. The ratios of the present values to those of the evaluated data files were 1.19, 1.14, and 1.05 for ENDF/B-VI, and 1.06, 1.09, and 0.88 for JENDL-3.2.

The 6-group constants, β_i, and λ_i, were also deduced from the experiment. The measured decay constant, λ_i, agreed well with the data file. Large discrepancies were, however, found in the group fraction β_i especially for Am-243.

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