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Measurements of Delayed Neutron Emission from Np-237, Am-241, and Am-243

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Introduction

Isotopes of transuranic elements are produced as a result of successive radiative capture reactions in the fuel of a nuclear reactor. Typically, these transuranic isotopes decay through long chains, have long half lives and dominate the long term toxicity of the spent reactor fuel. One of the options for waste management is to remove the transuranic from spent fuel by chemical processing, to load them into new special fuel elements, and to transmute them by neutron induced fission into shorter-lived fission fragments. Previous studies have shown the feasibility of actinide transmutation in either Light Water Reactors or Liquid Metal Fast Reactors.[1,2] Due to the anticipated high transuranic loadings in the fuel of actinide burner reactors, the neutronic properties of the transuranic isotopes will have a significant effect on the operational and safety characteristics of such reactors. Experiments to determine delayed neutron group yields and decay constants for Np-237, Am-241, and Am-243 have been designed and carried out. The experiments were conducted at Texas A&M University TRIGA reactor using a very fast pneumatic transfer system.

Experimental Procedure

The sample sizes ranged in mass from 10 mg to 300 mg and had isotopic purities from 97.633% to 99.999%. The samples used in this study consisted of a mixture of aluminum and actinide powders pressed

to form a pellet. A pneumatic system whose irradiation head was in a region with a thermal neutron flux of $9.0 \times 10^{12} \frac{neutrons}{cm^2\ sec.}$ at a power level of 1 MW was utilized for the irradiations.

The detection system consisted of an array of three BF3 proportional counters that were embedded in polyethylene and placed concentrically around a centered tube that received the sample from the pneumatic transfer system. Between the sample and the polyethylene, there was a 0.75 inch thick lead region to shield gamma rays emitted by the Am-241, the Am-243, the fission fragments, and the aluminum contained in the sample after irradiation. The detectors were shielded against background neutrons using an outer cylinder of 1 mm thick cadmium. The gamma sensitivity of the BF3 counters was tested and found to be negligible. The neutron detection efficiency of the BF3 system was determined to be 0.0092 ± 0.0004.

The measuring technique for the delayed neutrons was similar to that adopted by Keepin et al.[3] The samples containing the isotopes of interest were first placed inside polyethylene vials. These vials were then transferred to and from the irradiation position in the reactor by a pneumatic system, controlled by a timer that allowed irradiations from 1 to 999 seconds in duration. The samples were irradiated for different times to emphasize the importance of different delayed neutron groups. The transit time from the irradiation position to the detectors was 0.44 seconds.

The decay of the delayed neutrons was monitored by a computer which had a built-in 8191-channel multiscaler. Different counting times per channel were selected depending on the irradiation time used. Irradiation times of 180.0, 20.0 and 5.0 seconds were used during the course of the measurements. The corresponding counting times per channel were 8.0, 1.0, 0.1 seconds respectively. The dead time of the counting apparatus was measured to be $9.3 \pm 0.2\ \mu s$.

Fission rates were obtained by measuring the activity of certain fission products produced in the samples upon irradiation. For this purpose, the activities of Ba-140, La-140, Ru-103, I-131, Mo-99 were measured. These fission fragments were chosen because their fission yields are among the best known and also because of their relatively long half-lives, which allow the accurate measurement of their activities after the short lived fission products have decayed. The cumulative fission yield of the selected fission

products were taken from the Evaluated Nuclear Data Files (ENDF/B-VI).[4] The gamma spectroscopy was performed using a high purity Germanium detector.

Results and Analysis

In order to determine the delayed neutron group yields and decay constants, the neutron counts per channel were first corrected for dead-time losses. The delayed neutron decay curves were then plotted and approximate yields and decay constants were found by peeling. The peeled values were used as initial guesses in a least square fitting computer program that was developed specifically to determine group yields and decay constants. After fitting the data, five group parameters for the delayed neutrons emitted from Np-237, Am-241, and Am-243 were obtained.

The newly measured values have been compared to the ENDF/B-VI data. In general, very good agreement were obtained between the newly measured values and the ENDF/B-VI data especially for Np-237. For Am-241 and Am-243, good agreement was also obtained between the decay constants of the newly measured data and ENDF/B-VI data. The table below summarizes the delayed neutron yields (Y_i) and decay constants (λ_i) for Np-237, Am-241, and Am-243 from the new measurements and the ENDF/B-VI library.

g_i	This Work		ENDF/B-VI	
	λ_i	Y_i	λ_i	Y_i
	Np-237			
1	0.0129 ± 0.0006	0.00052 ± 0.00003	0.0133	0.000538
2	0.0324 ± 0.0011	0.00302 ± 0.00020	0.0317	0.003120
3	0.1048 ± 0.0019	0.00248 ± 0.0001	0.1190	0.002560
4	0.341 ± 0.013	0.0042 ± 0.0004	0.3060	0.006070
5	0.85 ± 0.06	0.00250 ± 0.00004	0.8710	0.002820
Am-241				
1	0.0122 ± 0.0006	0.000178 ± 0.000006	0.0133	0.000152
2	0.0318 ± 0.0016	0.00151 ± 0.00005	0.0307	0.001092

3	0.111 ± 0.007	0.00096 ± 0.00002	0.1130	0.000672
4	0.300 ± 0.017	0.0016 ± 0.0002	0.2867	0.001440
5	0.890 ± 0.023	0.000538 ± 0.000011	0.8653	0.000195
Am-243				
1	0.0131 ± 0.0015	0.000205 ± 0.000010	0.0135	0.000186
2	0.0311 ± 0.0009	0.00244 ± 0.00008	0.0298	0.002341
3	0.107 ± 0.007	0.0018 ± 0.0006	0.1140	0.001222
4	0.324 ± 0.021	0.00322 ± 0.00012	0.2999	0.002502
5	0.914 ± 0.031	0.00043 ± 0.00004	0.8820	0.001316

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