

A Nuclear Data Project on Neutron Capture Cross Sections of Long-Lived Fission Products and Minor Actinides

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A nuclear data project entitled “Systematic Study on Neutron Capture Reaction Cross Sections for the Technological Development of Nuclear Transmutation of Long-Lived Nuclear Wastes” is being performed. The objective of the project is to improve nuclear data libraries, by making the precise measurements of neutron capture cross sections of Long-Lived Nuclear Wastes (LLNWs), analyzing the measured results theoretically, and supplying reliable calculated capture cross sections for the LLNWs. This contribution presents the outline of the project, and individual results are presented by other contributions.

I. INTRODUCTION

The current Japanese policy for the management and disposal of long-lived nuclear wastes (LLNWs) generated in fission reactors is a sequence of vitrification, interim storage, and then depositing underground together with other nuclear wastes.

Alternatively, if LLNWs are extracted and transmuted into stable nuclides, the environmental loading in the geological repository becomes very small. Moreover, the ethical problem that LLNWs are undesirable legacy for our far descendants will be solved.

From this viewpoint, the nuclear transmutation of LLNWs is a very attractive subject, and neutron capture is the most promising transmutation reaction. Therefore, databases on neutron capture reaction cross sections are indispensable for developing the transmutation technology. However, the accuracy of databases is quite poor both in quality and quantity at the present time. Therefore, the improvement of the database accuracy is an urgent task. Thus, we started a nuclear data project on neutron capture cross sections of Long-Lived Fission Products (LLFPs) and Minor Actinides (MAs).

The project is entitled “Systematic Study on Neutron Capture Reaction Cross Sections for the Technological Development of Nuclear Transmutation of Long-Lived

Nuclear Wastes”, and its objective is to contribute to the improvement of nuclear data libraries, by making the precise measurements of neutron capture cross sections of LLNWs (for example, ⁹³Zr, ⁹⁹Tc, ¹⁰⁷Pd, ¹²⁹I for LLFPs, and ²³⁷Np, ²⁴¹Am, ²⁴³Am, ²⁴⁴Cm, ²⁴⁶Cm for MAs), analyzing the measured results theoretically, and supplying reliable calculated capture cross sections for the LLNWs in the whole neutron energy region. This contribution presents the outline of the project, and individual results are presented in detail by other contributions [1–10].

II. OUTLINE OF PROJECT

A. Overview

The structure of the project is shown in Fig. 1. The project has four participating groups: three groups for the measurements and one group for the theoretical analysis. The measurements are being performed by using the Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI) [11] in the Materials and Life Science Facility (MLF) [12] in the Japan Proton Accelerator Research Complex (J-PARC) as well as two other facilities: the Pelletron facility of the Tokyo Institute of Technology and the electron linear accelerator facility of the Kyoto University. The three groups for the measurements are in charge of the measurements at those three facilities, respectively. The measurement group at

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J-PARC/MLF/ANNRI is composed of three subgroups which are in charge of the neutron beam development, the measurement with a Ge detector array, and the measurement with an NaI(Tl) spectrometer, respectively. The theoretical analysis group analyzes all experimental results and supplies reliable calculated capture cross sections for the LLNWs.

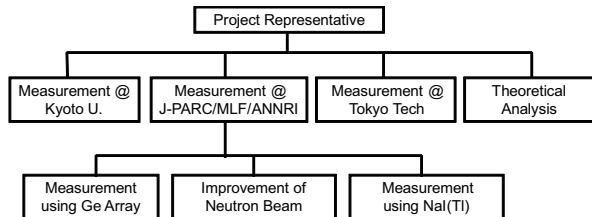


FIG. 1. Structure of the present nuclear data project.

B. Sample Preparation

We prepared the LLFP and MA samples shown in Table I. The net weights range from 0.6 mg to 1,000 mg. In the case of ^{237}Np samples, three samples with different thicknesses were prepared. As for ^{244}Cm and ^{246}Cm , six and four samples were prepared, respectively. The radioactivity of the 0.6 mg ^{244}Cm sample reaches 1.8 GBq. All samples were sealed in aluminum or titanium capsules. As for the ^{93}Zr , ^{107}Pd , ^{237}Np , ^{241}Am , ^{243}Am , ^{244}Cm , and ^{246}Cm samples, we prepared a small amount of unsealed samples from the same rod as the sealed samples, and we ourselves analyzed the isotopic compositions of the samples, by using a mass spectrometer and an alpha particle detector.

TABLE I. Prepared LLFP and MA samples for the present nuclear data project.

Sample	Half Life (y)	Decay Mode	Activity (Bq)	Net Weight (mg)
^{93}Zr	1.5×10^6	β^-	47M	472
^{99}Tc	2.1×10^5	β^-	50M	78
^{107}Pd	6.5×10^6	β^-	380k	20
^{129}I	1.6×10^7	β^-	3M	404
^{237}Np	2.1×10^6	α	26M	1000
"	"	"	5.2M	200
"	"	"	1M	38
^{241}Am	4.3×10^2	α	0.95G	7.5
^{243}Am	7.4×10^3	α	0.95G	128
^{244}Cm	1.8×10^1	α	$1.8\text{G} \times 6$	0.6×6
^{246}Cm	4.8×10^3	α	$1.8\text{G} \times 4$	1.1×4

C. Measurement at J-PARC/MLF/ANNRI

1. Neutron Beam

A 3 GeV pulsed proton beam is delivered to the mercury target of MLF. The pulse repetition rate is 25 Hz, and the pulse width is 600 ns. One pulse is composed of two 100 ns bunches. The average proton beam power is 200 kW to 300 kW at the present time.

Three types of liquid hydrogen neutron moderators are used for the MLF neutron source[12]. Decoupled and poisoned moderators are placed on the mercury target, and a coupled moderator is placed under the mercury target. The ANNRI uses the neutrons from the coupled moderator. The temperature of liquid hydrogen is about 19 K.

A layout of ANNRI is shown in Fig. 2. Neutron collimator, filter, and chopper systems are installed to deliver an intense neutron beam with good quality to experimental areas 1 and 2. A Ge detector array is installed in area 1, and the sample position for the Ge array is at 21.5 m from the MLF neutron source. An NaI(Tl) spectrometer is installed in the area 2, and the sample position is at 27.9 m.

The neutron intensities at the 21.5 m sample position of ANNRI are compared with those of DANCE at LANSCE and n_TOF at CERN in Fig. 3 [13]. Figure 3(a) shows the intensity per second and Fig. 3(b) shows the intensity per pulse. As seen from Figs. 3(a) and 3(b), both the intensities per second and per pulse of ANNRI are higher than those of the other facilities by about one order of magnitude.

2. Measurement Using Ge Detector Array

The Ge detector array is composed of two cluster Ge detectors, eight coaxial Ge detectors, BGO shielding detectors, and gamma-ray and neutron shields, as shown in Fig. 4. Since each cluster Ge detector has seven Ge crystals, the array has 22 Ge crystals. Signals from the Ge array are analyzed with digital processors, and recorded in mass storage devices as multi-dimensional data, event by event.

The advantage of the Ge array is its excellent energy resolution, and gamma rays from impurities in the objective sample can be discriminated by analyzing gamma-ray spectra. For example, in Fig. 5, the capture cross sections of ^{107}Pd measured with the Ge array [5] are compared with those contained in JENDL-4.0 [14]. The lowest two resonances at 3.9 eV and 5.2 eV in JENDL-4.0 are attributed to the resonances of ^{105}Pd and ^{109}Ag , respectively, by the analysis of gamma-ray spectra.

The measurements for ^{244}Cm and ^{246}Cm were performed with the Ge array [15]. The present measurements were the first ones with an accelerator neutron source, and the derived capture cross sections below 20 eV for ^{244}Cm and 50 eV for ^{246}Cm were the first measured cross sections.

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