NUCLEAR DATA MEASUREMENTS FOR P&T AND FUTURE PLANS IN JNC

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Abstract

Measurements of thermal neutron capture cross-sections (σ_0) and resonance integrals (I_0) of some important fission product (FP) nuclides, performed at JNC for partitioning and transmutation (P&T) studies, are presented. Method of the measurements and the results are reviewed, and possible reasons for discrepancies between the present data and that obtained by other researchers are discussed. Future plans on nuclear data measurements for P&T studies are presented.

1. Introduction

The reduction of the environmental loads is one of the important issues of the countries all over the world. In the field of nuclear energy production, the amount of radioactive nuclear wastes should be reduced. To reduce the amount, some methods have to be designed to transform these radioactive nuclides into stable ones.

One of the ways to transform these radioactive nuclides is the transmutation using the reactor neutrons. In order to study schemes of nuclear transmutation using the reactor neutrons, it is essential to know precise values of neutron cross-sections of these radioactive nuclides. Looking at the nuclear data of neutron reactions for these radioactive FP nuclides, the data are rather scarce, and the existing data are old and sometimes poor in accuracy. In this point of view, we have performed measurements of thermal (2 200 m/s) neutron capture cross-sections (σ_0) and resonance integrals (I_0) of some important radioactive FP nuclides and some the surrounding stable nuclides, using an activation method. These nuclides include ^{133,134,135,137}Cs, ^{90}Sr, ^{99}Tc and ^{127,129}I.

In this paper, our experimental method to determine σ_0 and I_0 of these radioactive FP nuclides, and the obtained results are reviewed. Then, our future plans on nuclear data measurements are presented.

2. Nuclear data measurements for P&T by JNC from 1990 to 1999

JNC has organised some researches on nuclear data measurements by several universities in Japan. These researches include:

- Measurements of fast neutron induced fission cross-section of americium isotopes (Department of Quantum Science and Energy Engineering, Tohoku University).
- Neutron capture cross-section measurement of ^{237}Np with lead slowing-down spectrometer (Research Reactor Institute, Kyoto University).
- Preliminary experiment of neutron capture cross-section of ^{99}Tc with lead slowing-down spectrometer (Research Reactor Institute, Kyoto University).
- Measurement of neutron capture cross-sections of ^{99}Tc (Research Laboratory for Nuclear Reactors, Tokyo Institute of Technology).
- Measurement of fission cross-section and fission neutron spectrum of ^{237}Np by an advanced technique (Department of Quantum Science and Energy Engineering, Tohoku University).

At the same time, JNC has continued their own effort on measurements of neutron capture cross-sections of long-lived FP nuclides (LLFP) using some research reactors in Japan, from 1990 until now [1-9]. This paper concentrates on the latter topic, and describes our experimental methods and results.

The experimental procedure to determine σ_0 and I_0 is based on an activation method, in which samples are irradiated with neutrons and then γ rays are measured which are emitted during de-excitations of the daughter of the capture products, to determine reaction rate R of the capture reaction. For the γ-ray measurements, a high purity Ge detector with a large volume is used. This enables determination of γ-ray yield in an efficient and reliable manner and thus the reaction rate R is determined precisely in case the precise values of emission probabilities of the γ-rays are available. In

order to determine σ_0 and I_0, at the same time, irradiations and measurements are also done for samples with a Cd shield.

The procedure to determine σ_0 and I_0 from the obtained R is based on Westcott's convention [10]. It was already described elsewhere [1], and only a brief summary is given here:

In the Westcott's convention, the reaction rate R in well-moderated neutron fields is expressed as:

$$R = n\,\upsilon_0\,\sigma_{\mathrm{eff}}$$

where in the convention $n\upsilon_0$ is the "neutron flux" with neutron density n including thermal and epithermal neutrons and with velocity υ_0=2 200 m/s, and σ_{eff} an effective cross-section. The σ_{eff} is written as:

$$\sigma_{\mathrm{eff}} = \sigma_0\,[g\,G_{\mathrm{th}} + r(T/T_0)^{1/2}\,s_0\,G_{\mathrm{epi}}]$$

where σ_0 is the reaction cross-section for 2 200 m/s neutrons and g the measure of deviation of the cross-section from the $1/\upsilon$ law in the thermal energy region. In the analysis the g is assumed to be unity. The quantity $r(T/T_0)^{1/2}$ gives the fraction of epithermal neutrons in the neutron spectrum, and s_0 is defined as:

$$s_0 = 2I'_0/((\pi)^{1/2}\sigma_0)),$$

with I'_0 the reduced resonance integral, i.e. the resonance integral after subtracting the $1/\upsilon$ component. The G_{th} and G_{epi} are self-shielding factors for thermal and epithermal neutrons, respectively. The above equations are combined to read:

$$R/\sigma_0 = G_{\mathrm{th}}\,\phi_1 + s_0\,G_{\mathrm{epi}}\,\phi_2,$$

where ϕ_1 and ϕ_2 represent simplified flux factors. The ϕ_1 and ϕ_2 can be determined by using flux monitors whose cross-sections and resonance integrals are already determined precisely. As flux monitors, we use Co and Au, which differ in sensitivities to thermal and epithermal neutrons. By using two flux monitors with different sensitivities to thermal and epithermal neutrons, ϕ_1 and ϕ_2 can be determined unambiguously. The self-shielding factors G_{th} and G_{epi} are usually almost unity and can be calculated by considering geometries of irradiations.

From the obtained reaction rates R and flux factors ϕ_1, ϕ_2 for irradiations with and without Cd shield the cross-sections σ_0 and the reduced integrals I'_0 are deduced. The resonance integral, I_0, is deduced from I'_0 using the following relation:

$$I_0 = I'_0 + 2\,\sigma_0\,(E_0/E_{\mathrm{Cd}})^{1/2}$$

where E_0 and E_{Cd} are neutron energy at 2 200 m/s and Cd cut-off energy.

In Table 1, the results obtained by the present authors are summarized [1-9] along with the data previously published by other research groups [11-18]. The table includes results of the neutron capture cross-sections and resonance integrals for long-lived FP nuclides (LLFP) as well as those for their stable isotopes: the latter are also important because these stable nuclides absorb neutrons and affect transmutation rates of LLFP, and also these stable nuclides can be transformed into radioactive ones by absorbing neutrons.

Some of the data obtained by the present author do not differ significantly from those obtained previously. For example, for ^{134}Cs nuclide, the effective cross-section obtained by Bayly *et al.* [17]

agrees ours within limits of errors. Also, the thermal neutron cross-section and the resonance integral of ^{129}I nuclide obtained by the present authors are close to those published by Eastwood *et al.* [14].

On the other hand, for some nuclides, the results obtained by the present authors differ considerably from others. An example of the discrepancy is the result for ^{99}Tc nuclide. Although the thermal neutron cross-section does not differ much, the result of the reduced resonance integral obtained by the present authors [4] are about twice as large as that obtained by Lucas *et al.* [13], as depicted in Figure 1. The origin of this discrepancy may be ascribed to the characteristics of their irradiation: their analysis was based on the same convention as ours which is valid only for well moderated neutron spectrum, but at one of their irradiation positions, the index for the epithermal neutrons, *r*, is as large as 0.15. Even with our data, the number of existing data of resonance integral for ^{99}Tc nuclide is only two. It should be stressed that, in order to be certain that correct values of σ_0 and I_0 are obtained, at least two different types of measurements have to be done. This is also true for other radioactive FP nuclides such as ^{90}Sr and ^{137}Cs.

It should also be noted that the data presented in [4] do not include the error of γ-ray emission probabilities: because of its short life, γ-ray emission probabilities of ^{100}Tc nuclide are determined with an error of as large as 17% [22]. In order to obtain σ_0 and I_0 of ^{99}Tc nuclide more accurately, accurate values of γ-ray emission probabilities of ^{100}Tc nuclide are needed.

3. Future plans on nuclear data measurements for P&T in JNC

Following the decision of the Atomic Energy Commission in Japan that the basic study on P&T should be continued JNC resumes the nuclear data measurement under a basic study scheme for P&T from 2000. Now we are planning to extend our area of nuclear data measurements over capture cross-sections, fission cross-sections and decay data for important LLFP and MA for the energy region from thermal to a few MeV. The plan includes the following researches and developments:

- More precise determination of the capture cross-sections of nuclides such as ^{99}Tc and ^{129}I.
- Development of prompt γ-ray spectroscopic method for the determination of the neutron capture cross-sections of LLFP.
- Development of a new spectroscopic method to measure neutron cross-sections for energy range from thermal to a few MeV.

3.1 More precise determination of the capture cross-sections

As already mentioned above, γ-ray emission probabilities of ^{100}Tc nuclide are not determined with enough accuracy because of its short life. To obtain more precise values for capture cross-sections of the ^{99}Tc nuclide, the γ-ray emission probabilities of ^{100}Tc should be determined more accurately. In order to achieve this, a β-γ coincidence measurement system has been developed for the determination of γ-ray emission probabilities of short-lived nuclides [23]. An experiment has been already performed using the system to precisely determine γ-ray emission probabilities of ^{100}Tc nuclide.

3.2 Prompt γ-ray spectroscopy

For the determination of capture cross-sections of nuclides whose capture products are stable, a conventional activation method can not be applied in which de-excitation γ-rays are observed of daughter nuclides of the capture products. These include some important long-lived FP nuclides such as ^{93}Zr, ^{79}Se and ^{107}Pd. In order to determine capture cross-sections of such nuclides, a prompt γ-ray spectroscopic method is being developed, in which complete level schemes are constructed by in-beam γ-γ coincidence measurements using thermal neutron beam and then γ-ray emission probabilities are determined. By using the obtained emission probabilities, neutron capture cross-sections are determined.

This method is also applicable to nuclides whose capture products are not stable ones: measurements in this method will confirm the results that are already obtained by using other methods such as an activation technique.

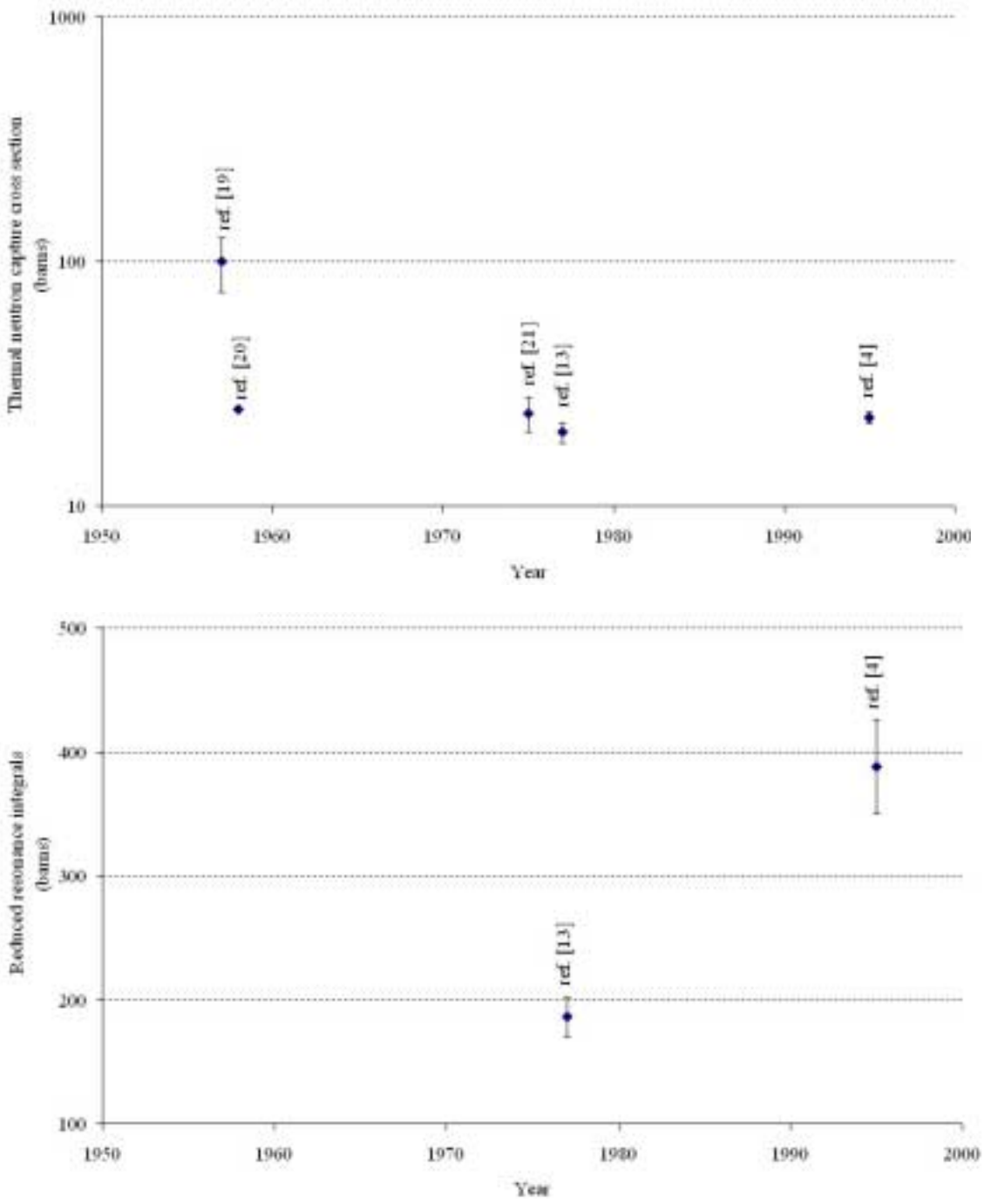
3.3 Development of a new spectroscopic method for neutron cross-sections in a wide energy region

In order to efficiently determine neutron cross-sections of LLFP and minor actinides over a broad energy range from thermal to MeV region, some new experiments will be required. The present authors are planning to start the international collaborations from 2001 Japanese fiscal year.

Table 1. **Neutron capture cross-sections at 2 200 m/s neutron energy and resonance integrals for some important fission product nuclides, obtained by the present authors as well as other researchers**

Nuclide	Half-life (year)	Previous data (barns) (Authors and published year)	Data obtained by JNC (barns)
^{137}Cs	30	$\sigma_{eff} = 0.11 \pm 0.03$ (Stupegia 1960 [11])	$\sigma_0 = 0.25 \pm 0.02$ $I_0 = 0.36 \pm 0.07$ (1990 [1], 1993 [2])
^{90}Sr	29	$\sigma_{eff} = 0.8 \pm 0.5$ (Zeisel 1966 [12])	$\sigma = (15.3 + 1.3 - 4.2) \times 10^{-3}$ $I_0 \leq 0.16$ (1994 [3])
^{99}Tc	2.1×10^5	$\sigma_0 = 20 \pm 2$ $I'_0 = 186 \pm 16$ (Lucas 1977 [13])	$\sigma_0 = 22.9 \pm 1.3$ $I = 398 \pm 38$ ($I' = 388 \pm 38$) (1995 [4])
^{129}I	1.6×10^7	$\sigma_0 = 27 \pm 2$ $I_0 = 36 \pm 4$ (Eastwood 1958 [14])	$\sigma_0 = 30.3 \pm 1.2$ $I_0 = 33.8 \pm 1.4$ (1996 [5])
^{127}I	(stable)	$\sigma_0 = 4.7 \pm 0.2$ $I_0 = 109 \pm 5$ (Friedmann 1983 [15])	$\sigma_0 = 6.40 \pm 0.29$ $I_0 = 162 \pm 8$ (1997 [6])
^{135}Cs	2.3×10^6	$\sigma_0 = 8.7 \pm 0.5$ $I_0 = 61.7 \pm 2.3$ (Baerg 1958 [16])	$\sigma_0 = 8.3 \pm 0.3$ $I_0 = 38.1 \pm 2.6$ (1997 [7])
^{134}Cs	2	$\sigma_{eff} = 134 \pm 12$ (Bayly 1958 [17])	$\sigma_{eff} = 141 \pm 9$ (1999 [8])
^{133}Cs	(stable)	$\sigma_0 = 30.4 \pm 0.8$ $I_0 = 461 \pm 25$ (Baerg 1960 [18])	$\sigma_0 = 29.0 \pm 1.0$ $I_0 = 298 \pm 16$ (1999 [9])

Figure 1. **Thermal neutron capture cross-sections (upper) and resonance integrals (lower) of ^{99}Tc nuclide**



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