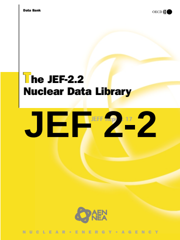

JEF/DOC-727

Remarks on Measurements

on long-lived radioactive Fission Products.

H. Weigmann

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Remarks on measurements on long-lived radioactive fission products.

The present note is a response to a demand expressed during the JEF meeting in Paris in June 1997. It gives a brief overview of problems encountered in neutron data measurements on long-lived radioactive fission products. The note is organised in two chapters:

1. General considerations.
2. A brief review of the status for a few prominent cases.

Chapter 1 : General Considerations.

Long-lived radioactive fission products present a major concern for geological waste depositories. Therefore their incineration in waste transmutation facilities is considered. In addition, some of these isotopes have large capture cross sections and therefore contribute significantly to neutron absorption in reactors at high burn-up. To determine their effect on reactor performance and to judge the feasibility of their incineration, the neutron cross sections of these isotopes are needed; in particular, the capture cross sections must be known with an accuracy of typically 5 - 10 %.

The special difficulties met with measurements on radioactive fission products are connected with availability of the sample material and with its radioactivity.

The non-availability of suitable sample material is the most severe problem met in any attempt to perform measurements of fission product cross sections. In principle, plenty of fission products are of course present in the waste stream of fuel reprocessing plants. However, separation of individual fission product elements from the bulk waste stream would mean a modification of the routine operational procedure of the plant, and this is generally not accepted. In the early years of nuclear energy when the fuel cycle was not industrialised to the degree it is today, the availability of samples has apparently been less of a problem. Therefore, a number of measurements on fission product nuclei have been performed years ago which today would meet severe difficulties from the simple problem of availability of the base material.

Moreover, even if a given element has been separated from the fission product waste, it will come in the isotopic mixture as it is being produced in fission. Measurement of reactions on the isotope of interest will suffer from interference due to reactions on the other isotopes present.

The difficulties connected with the radioactivity of the sample are very dependent on the isotope in question and on the cross section to be measured, but they are often less severe than what might at first glance be expected. The long half-life of the isotopes in question is connected with a low beta decay Q-value, and this in turn has the consequence that often no or only very low energy gamma-rays are emitted (see table 1; data taken from R.B. Firestone et al., Table of Isotopes [1]). Thus e.g. a sample of 10 g ^{99}Tc, although it has an activity of about $6.4*10^9$ Bq, provides only moderate difficulties even for direct capture measurements.

Table 1: Decay Properties of Fission Product Nuclei.

Isotope	half-life [y]	beta decay Q-value [keV]	Gamma-rays energy [keV]	Gamma-rays branching ratio [%]
^{79}Se	$< 6.5 * 10^4$	150.7	-	-
^{93}Zr	$1.5 * 10^6$	91.1	30.8	> 95.
^{99}Tc	$2.1 * 10^5$	293.5	89.7	0.0016
^{126}Sn	$1. * 10^5$	380.	various, highest energy 87.6	
^{129}I	$1.6 * 10^7$	194.	39.6	100.
^{135}Cs	$2.3 * 10^6$	268.6	-	-
^{147}Sm	$1.1 * 10^{11}$	(2310.)**	-	-
^{151}Sm	90.0	76.8	21.5	0.91

** ^{147}Sm decays by alpha- rather than beta-decay.

Of course, the experimental difficulties also depend on the cross section type to be measured. Direct measurement of the capture cross section may meet difficulties with the radioactivity of the sample, but in a transmission measurement where the detector is far away from the sample, there is no technical difficulty, the only possible problem being the handling of the sample. In this connection it is to be mentioned that if a transmission measurement has sufficient resolution to allow shape analysis of a reasonable number of resonances, a reliable value for the average radiative width may be extracted; this value can then be very useful at higher energies to deduce the capture cross section from the measured total cross section when a direct measurement of the capture cross section is not available.

The difficulty with the measurement of the capture cross section of a radioactive sample is not only the detector background itself; in order to reduce this background it may be necessary to shield the low energy background gamma radiation from the sample by introducing a lead shield in between sample and detector. This however will modify the response function of the detector, especially in the case of the type of detector most commonly used today, i.e. the "total energy detector" employing a weighting function for the efficiency determination.

Generally, very little experimental information is available on cross sections other than total or capture (one exception is ^{147}Sm which has a large (n,alpha) cross section at thermal and in the resonance region and has extensively been studied at Dubna [2,3]). The main reason probably is that such data are rarely requested. For the same reason, also in this note these cross sections are mentioned only in general terms:

- Measurement of (n,n') and (n,2n) cross sections by neutron detection will meet the usual problem of the availability of a suitable sample. Also the radioactivity of the sample may pose a difficulty similar to the one in capture measurements.
- Measurement of the (n,n') cross section by detection of gamma-rays after inelastic scattering will generally be more difficult than a capture measurement because of the low energy of the gamma-rays to be detected which has as a consequence that the detector cannot be effectively shielded against the radioactivity of the sample.
- Difficulties with the measurement of (n,2n), (n,p) or (n,alpha) cross sections by activation will again depend on the individual case. One condition for the feasibility is that the half-life of the reaction product must be sufficiently short: For example, if the half-life is longer than a few hours, even with a sizeable cross section and an intense neutron source will the induced activity be less than about 10^{-4} of the activity of the sample material and may thus be difficult to measure. But again, this will depend on the decay properties (gamma energies and branching ratios) of both, the reaction product and the sample material. In the case of (n,p) and (n,alpha) cross sections when chemical separation of the reaction products can be applied (and does not pose too big problems with the highly active sample material), these background problems may be largely overcome.
- If (n,p) and (n,alpha) reactions cannot be measured by activation, direct detection of the emitted proton or alpha-particles e.g. in an ionisation chamber, would be a feasible alternative. As only mg amounts of sample material would be needed, difficulties with its activity should not be too large.

Chapter 2: Brief Review of Selected Isotopes.

In this chapter a rough overview of the status of experimental data for a few long-lived radioactive fission products is given. This is by no means meant to be complete: In particular, no literature prior to 1965 and no integral experiments are considered.

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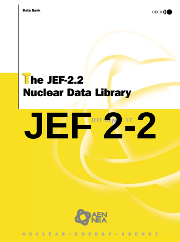

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Again the status of data as of 1992 is presented in the paper of Van der Kamp and Gruppelaar [6]: For the thermal capture cross section and the capture resonance integral they adopt experimental values of Friedmann and Aumann [11]. In the resonance region, apart from few older measurements over limited energy ranges, an extensive measurement by Macklin [12] exists. It utilised a sample of (including impurities) about 19 g of PbI_2 which contained about 3.8 g of ^{129}I. The measurement covered the energy range from 11 eV to 500 keV; a resonance analysis yielded capture areas for resonances up to 3.4 keV. With these data and the evaluation of Van der Kamp and Gruppelaar, the status of data for ^{129}I looks rather satisfactory; however, for both the thermal data and the resonance and keV regions, it rests on only one measurement each.

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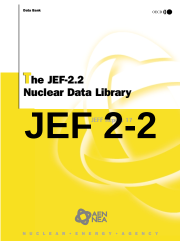

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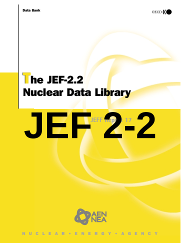

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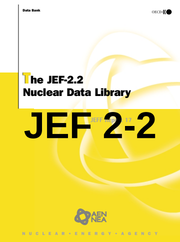

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In this chapter a rough overview of the status of experimental data for a few long-lived radioactive fission products is given. This is by no means meant to be complete: In particular, no literature prior to 1965 and no integral experiments are considered.

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