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**High Priority Nuclear Data Request List.
The Data for Long-lived Fission Products, Minor Actinides and the Thorium Cycle.**

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Abstract.

The history of the High Priority Request List is described first, followed by a brief discussion of the specification of accuracy requirements. The requirements for incineration of long-lived fission products and minor actinide isotopes, and for the thorium cycle, are discussed in general terms and the work needed to define requirements more precisely is indicated. Subgroups of the WPEC are reviewing aspects of the data and their recommendations will help in the definition of requirements.

1. Introduction.

The first international request list for nuclear data was produced about 40 years ago by the European-American Nuclear Data Committee (this committee later becoming the NEA Nuclear Data Committee, the NEANDC). The list developed to become WRENDA, the World Request List for Nuclear Data, and the Nuclear Data Section of the IAEA took responsibility for compiling it. Each country submitted its requirements, with national nuclear data committees going through the painful process of reviewing each item of their own list, every year or two, and deciding which had been met, which were no longer required and considering the justification for new requirements.

Some countries were more rigorous than others in considering the justification for requests. Because of concerns about the size and realism of the requests in WRENDA one country went so far as to propose that all requests should be justified by a cost-benefit analysis, a very difficult (impossible?) thing to do, and also proposed that requesters should be prepared to pay for measurements. When this suggestion was put to a meeting of the IAEA International Nuclear Data Committee it was firmly rejected because it was considered that the free exchange of measured nuclear data would be affected. We can note that although the results of differential nuclear data measurements are published openly, this isn't the case for integral measurements, although simple integral measurements (thermal spectrum averages and resonance integrals, for example) are usually published. However, even when the results of integral measurements are not openly published, conclusions relating to nuclear data, which have been drawn from the analyses of integral measurements, are usually published. There was a time when evaluated nuclear data libraries were restricted in their availability but at the present time they are distributed freely (in some cases following a period of testing).

2. The High Priority Request List.

Because of concerns about the number of requests in WRENDA, and the justification for some of them, about 20 years ago the NEA Committee on Reactor Physics (NEACRP), together with the NEANDC, proposed the production of a High Priority Nuclear Data

Request List. The aim was for the NEACRP to try to reach agreement on what are the most important requirements and for the NEANDC to closely monitor the progress made towards meeting them. It could also provide a framework for collaborative work between the nuclear data laboratories. Agreement was reached between French, German and UK participants on a set of high priority requirements and contributions were also received from Japan and the USA. More recently the coordination of the production of the list has become the responsibility of a subgroup of the NEA Working Party on International Evaluation Cooperation, the WPEC Subgroup C. Progress in meeting the requirements is reviewed at an annual joint meeting of the WPEC and the Working Party on International Nuclear Data Measurement Activities, the WPMA.

In the past 10 years there has been a reorientation of fission reactor programmes in most countries. Germany, the UK and the USA have stopped their fast reactor development programmes (although maintaining an interest in continuing work in other countries). Because of this, different countries now have different priorities for improved nuclear data relating to fission reactors (although there is agreement between countries on some requirements, for example, those relating to fuel storage, transport and reprocessing). No attempt has been made in recent years to produce an internationally agreed set of high priority requirements for fission reactor technology. However, several countries have cooperated to produce an agreed list for fusion reactor technology and for intermediate energy applications.

What sort of data requirements are to be included? Requirements can be for:

- differential measurements,
- integral measurements,
- nuclear theory calculations or
- analysis and evaluation of existing measured data.

The requester should indicate which of these he is requesting. However, it is sometimes difficult for the requester to know by which of these methods his requirements for data are to be met. One of the purposes of the joint meetings of the WPMA and the WPEC is to give advice on this – indeed to advise on whether it is feasible to meet the request at all. There are various aspects to be considered – whether the required measurement accuracy can be achieved using existing facilities, the availability of suitable samples, etc.

The current High Priority Request List can be viewed at the web site of the NEA. It is divided into sections:

- *Section 1.* Standards, and requirements for the interpretation of measurements.
- *Section 2.* Dosimetry.
- *Section 3.* Fusion reactor technology.
- *Section 4.* Fission reactor technology.
- *Section 5.* Requirements for medical and industrial applications.
- *Section 6.* Intermediate energy requirements.
- *Section 7.* Japanese requirements for waste transmutation

In the following sections requirements for fission reactor technology applied to the transmutation of radiotoxic waste are discussed. For the fission product isotopes and minor actinide isotopes there is an emphasis on validation of the existing evaluations using integral measurements. However, it is also desirable to update the evaluations to include recent measurements and to apply improved

theoretical models. For some items there are requirements for differential cross-section measurements.

3. Definition of Accuracy Requirements.

How are the accuracy requirements to be chosen and what do they mean? In the case of cross-sections it is the error which is systematic over a range of energies which is important. This is because in reactor calculations one is mainly concerned with reactor spectrum averaged values. Sensitivity calculations can be used to guide the choice of accuracy requirement. First one must specify the accuracies required in the reactor properties to be calculated using the nuclear data. Then one must decide how the total uncertainty is to be partitioned between the contributing items of nuclear data. In order to decide on this partitioning some judgement must be made about the relative costs (or relative difficulties) of obtaining different types of nuclear data, for different materials, to different accuracies. Usually a simple assumption is made and the accuracy requirement is taken to be inversely proportional to the sensitivity. It might be better to aim for a higher accuracy than this for the less sensitive items, or materials, however, so as to eliminate them as sources of uncertainty. If a measurement is required it might be better to measure it to the achievable accuracy for that type of data, rather than to a lower accuracy and then, when the measurement has been made, find a higher accuracy is now needed.

In most cases the accuracy requested has been based on a broad judgement rather than extensive sensitivity calculations. Indeed, for many proposed new developments the designs are not precisely defined and so the sensitivities cannot be accurately calculated. Also, the role of integral measurements in meeting the requirements isn't well defined in advance.

In some cases measurements are required to resolve discrepancies between different measurements or with the conclusions drawn from the analysis of integral measurements. Measurements can be required to fill in gaps in the data when interpolation is found to be unreliable. Measurements can also provide parameters for the theoretical models used to calculate cross-sections, or can be made to validate theoretical methods (for example, the methods used to calculate inelastic scattering by fission product nuclei).

4. Priorities.

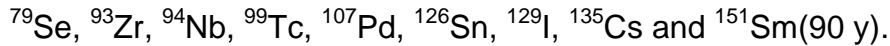
The latest edition of WRENDA (93/94) contains 71 pages of requests, 720 requests in all. They are divided into three categories, or priorities. The High Priority Request List has grown to about 40 pages of requests and as a consequence it has been considered necessary, in some sections, to assign priorities. This is now the case for the fission reactor technology requests.

5. Nuclear Data Requirements for Studies on the Transmutation of Fission Product Isotopes.

Fission product nuclear data are being reviewed by subgroups of WPEC. Subgroup 10 (coordinator, Kawai) is reviewing inelastic scattering data and plans to report shortly. Subgroup 17 (coordinator, Gruppelaar) has carried out intercomparisons of cross-sections averaged in a fast reactor spectrum (ECN-R-98-014). A new subgroup (coordinator, Gulliford) will review fission product cross-sections for thermal reactors. However, the

emphasis in these subgroups has been on data for conventional reactors, rather than for transmutation of long-lived fission products.

The long-lived isotopes considered as potentially suitable for transmutation are:



with primary consideration being given to ^{93}Zr , ^{99}Tc , ^{107}Pd , ^{129}I and ^{135}Cs . Of greatest interest are ^{99}Tc and ^{129}I .

Consideration has also been given to ^{90}Sr (29 y) and ^{137}Cs (30 y). For both of these isotopes there are significant differences between the capture data in JEF-2.2, ENDF/B-VI and JENDL-3.2. There have been recent measurements, by Harada et al (1994), of the thermal capture cross-sections and resonance integrals and the JENDL-3.2 values are consistent with these.

The JEF-2.2 evaluations for ^{99}Tc and ^{129}I are revisions, made by van der Kamp and Gruppelaar (1992), to the earlier evaluations. JENDL-3.2 also includes revisions to the resonance region data for ^{99}Tc .

In addition to data for these isotopes the cross-sections of other fission product isotopes of the element, and their reaction products, are required. However, consideration is being given first to the data for the above 9 isotopes.

The fission yield and radioactive decay data are comparatively well known. The primary requirement for the studies on the reductions in radiotoxicity of waste by means of transmutation is for validated capture cross-section data. In the resonance region total cross-section data are also required so that resonance shielding effects can be calculated. The scattering data are less important because the fission product isotope being transmuted is usually relatively dilute in the incineration assembly, and the scattering cross-sections of the matrix materials have the main effect in determining the spectrum.

The primary requirement for the isotopes of interest is for validation of the existing evaluations using the results of thermal and fast reactor integral measurements. Evaluations are available in JEF-2.2 and ENDF/B-VI for all the isotopes excepting ^{79}Se , whereas JENDL-3.2 includes evaluations for all the isotopes. There are fast spectrum integral measurements for many of the isotopes of interest. For these the requirement is for validation of the evaluations using the available measurements. In some cases there are inconsistencies between integral measurements, and further measurements would then be valuable.

For thermal reactor applications, measurements of the 2200m/s and resonance integrals are required. A review of the measurements prior to about 1980 is given by Mughabghab (1981 and 1984) and in many cases these data have been taken into account in the evaluations. More recent measurements are available for several isotopes. In some cases there are inconsistencies between the integral values and the differential measurements. Reactivity worth measurements in thermal reactor spectra have been made in MINERVE at Cadarache, and in DIMPLE at Winfrith, in the CERES programme of measurements, for the fission product isotopes of importance in fuel transport and reprocessing. These included measurements for ^{99}Tc .

Comparing the thermal values and resonance integrals in JEF-2.2 and JENDL-3, from JEF Report 14, gives some indication of the status of the thermal region data:

Isotope	JEF-2.2		JENDL - 3.2	Measur e		Res.	
	2.2km/s	R.I.	2.2km/s	R.I.	2.2km/s	R.I.	Params. to
⁷⁹ Se	Absent		50.0	60.6			None
⁹³ Zr	1.78	33.0	2.24	18.1			1.7 keV
⁹⁴ Nb	13.6	117	15.8	125			3 res.
⁹⁹ Tc	19.1	304	19.7	311	22.9±1.3	398±38	4.2 keV
¹⁰⁷ Pd	1.81	105	2.01	111			3.5 keV
¹²⁶ Sn	0.300	0.160	0.090	0.130			None
¹²⁹ I	33.9	30.3	27.0	29.0	30.0±1.4	33.2±1.5	3.4 keV
¹³⁵ Cs	9.02	61.0	8.70	62.3			2 res.
¹⁵¹ Sm	15200	3465	15100	3407			250 eV

Table 1. Comparison of thermal capture cross-section values and resonance integrals (JEF Report 14). The recent measured values are those of Harada et al (1984).

We see that there are significant differences for ⁹³Zr and ¹²⁶Sn.

In addition to the requirement for validation using integral measurements, for ⁹⁹Tc and ¹²⁹I there is a requirement for more accurate differential data in the resonance region and in the keV region up to about 100 keV, for fast and thermal reactor transmutation studies.

Fast spectrum integral measurements have been made in a series of 5 cores built in the STEK facility at Petten (Veenema and Janssen, 1976). These were reactivity worth measurements, but the effect is dominated by the capture for most isotopes. This is the most comprehensive series of integral measurements for fission product isotopes, with measurements being made in 5 different spectra for several sizes of sample. Reactivity worth measurements for fission product isotopes have also been made in the fast-thermal coupled system RRR/SEG at Rossendorf (Dietze et al, 1994). Other fast spectrum measurements include the activation measurements made in the CFRMF facility, in the ERMINE facility (the ZONA-1 and RONA-3 programmes) and irradiations in PHENIX and in EBR-2.

In Table 2 the fast spectrum averaged capture cross-sections in JEF-2.2 and JENDL-3.2 are compared, and the availability of integral and differential measurements is noted.

There appears to be an absence of measured differential data for several isotopes, both in the resonance region and at keV energies. However, in some of these cases there are integral measurements which give confidence in the data files derived theoretically.

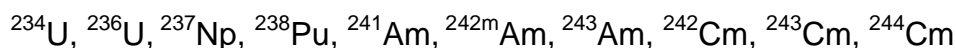
Isotope	JEF-2.2	JENDL-3.2	STEK/JEF	Other Int.	differential
⁷⁹ Se	Absent				
⁹³ Zr	0.135	0.106	Discrepant		ORNL
⁹⁴ Nb					None
⁹⁹ Tc	0.630	0.592	Good	CFRMF	Chou; Macklin
¹⁰⁷ Pd	1.057	1.052	Good		Macklin
¹²⁶ Sn					None
¹²⁹ I	0.364	0.384	Good (?)	CFRMF	ORNL
¹³⁵ Cs	0.238	0.228	Good		None
¹⁵¹ Sm	3.36	2.11	C/E ~ 1.2	PHENIX	None

Table 2. Comparison of fast spectrum averaged capture cross-sections. Cross-section data from the report of WPEC Subgroup 17, ECN-R-98-014.

We see that there are some isotopes for which no integral measurements have been made and it would be helpful if measurements could be made for these. However, the number of facilities available for making integral measurements is now much fewer than it was 5 or 10 years ago.

6. Requirements for the Minor Actinide Isotopes.

Reactor designs for plutonium incineration require more accurate data for the higher plutonium isotopes, as well as for americium and curium isotopes. For uranium and plutonium fuelled reactors recycling plutonium and the isotopes of Np, Am and Cm there are data requirements for:



as well as for more accurate data for the higher plutonium isotopes: ²⁴⁰Pu, ²⁴¹Pu and ²⁴²Pu.

Subgroup 8 of the WPEC (coordinators, Nakagawa and Takano) is reviewing the minor actinide data and plans to report at the end of this year.

For the minor actinide isotopes it is the fission and capture cross-sections which are of main interest, rather than the fission neutron yields, fission neutron spectra and scattering cross-sections.

We look first at the consistency of the values in the files, using data in JEF Report 14. For the isotopes which are fissile only at high energies the fission spectrum averaged values (given in JEF Report 14) of the fission cross-sections give a good indication of the relative values. Apart from ^{242m}Am and ²⁴³Cm, the fission cross-sections are comparatively small in the thermal and resonance ranges. The fission spectrum averages are compared in Table 3:

Isotope	JEF-2.2	ENDF/B-VI	JENDL-3.2
²³⁴ U	1.215	1.215	1.211
²³⁶ U	0.582	0.574	0.586
²³⁷ Np	1.290	1.305	1.321
²³⁸ Pu	1.968	1.965	1.984
²⁴¹ Am	1.323	1.387	1.359
^{242m} Am	1.842	2.222	1.844
²⁴³ Am	1.100	1.112	1.066
²⁴² Cm (163 d)	1.647	0.992	1.830
²⁴³ Cm (29.1 y)	2.174		1.946
²⁴⁴ Cm (18.1 y)	1.608	1.549	1.564

Table 3. Comparison of fission spectrum averaged fission cross-sections.

Agreement between the files doesn't mean that the data are accurate, but more probably that the same measured data have been the basis of the cross-sections adopted in the different files. The agreement is good, apart from ^{242m}Am (which is fissile at thermal energies) and ²⁴²Cm.

Comparing next the thermal values and resonance integrals:

Isotope	JEF-2.2		ENDF/B	-VI	JENDL-	3.2
	thermal	R.I.	thermal	R.I.	thermal	R.I.
²³⁴ U	0.465	0.618	0.465	0.618	0.0062	0.809
²³⁶ U	0.0469	4.38	0.0472	4.46	0.0613	4.36
²³⁷ Np	0.0180	0.207	0.0180	0.214	0.0225	0.886
²³⁸ Pu	17.3	22.7	17.1	21.0	17.9	22.9
²⁴¹ Am	3.18	9.77	3.15	8.26	3.02	7.37
^{242m} Am	6886	1630	6700	1874	6420	1549
²⁴³ Am	0.0496	1.19	0.0741	2.13	0.116	2.246
²⁴² Cm	5.00	4.48	3.02	0.492	5.07	11.2
²⁴³ Cm	432	1771			618	1553
²⁴⁴ Cm	1.04	11.9	1.04	5.96	1.03	5.72

Table 4. Comparison of thermal fission cross-sections and resonance integrals.

One notes that there are some very large differences in between thermal values and resonance integrals when these are small, but for ^{242m}Am the differences between the thermal values are small and between the resonance integrals they are about 20%. For ²⁴³Cm the difference between the two thermal fission values is about 30%.

We look next at the thermal capture data in the files:

Isotope	JEF-2.2		ENDF/B	-VI	JENDL-	3.2
	thermal	R.I.	thermal	R.I.	thermal	R.I.
²³⁴ U	103	659	103	659	100	630
²³⁶ U	5.16	346	5.14	341	5.30	345
²³⁷ Np	181	658	181	659	165	664
²³⁸ Pu	547	143	564	152	540	153
²⁴¹ Am	616	1444	622	1385	601	1303
^{242m} Am	1809	262	1359	287	1256	245
²⁴³ Am	76.0	1811	75.4	1818	78.6	1820
²⁴² Cm	16.5	116	16.9	111	15.9	108
²⁴³ Cm	113	284			130	198
²⁴⁴ Cm	14.4	633	15.1	659	15.3	634

Table 5. Comparison of thermal capture cross-sections and resonance integrals.

The differences between the thermal capture cross-sections and resonance integrals are small, apart from ^{242m}Am and ²⁴³Cm.

As was the case for the fission product isotopes, the first requirement is for validation of the data using thermal and fast reactor integral measurements. Irradiation experiments provide data on the capture cross-sections and (n,2n) reactions. Fission cross-sections can be validated using relative fission rate measurements made in zero power facilities, using fission chambers and foils.

There have been several programmes of measurements in France, such as the PROFIL fast reactor spectrum irradiations, which give information on the capture cross-sections of the primary actinide isotopes, as well as ²³⁷Np, ²³⁸Pu, ²⁴¹Am and ²⁴³Am. There are also other fast spectrum irradiation measurements in the literature. There have also been irradiations of isotopic samples in thermal and epithermal spectra in France (the SHERWOOD and ICARE experiments). A number of minor actinide isotopes were irradiated, including ²³⁴U, ²³⁷Np, ²³⁸Pu, ²⁴¹Am, ²⁴³Am and ²⁴⁴Cm. The SHERWOOD measurements were used to define a benchmark, including the results for ²⁴¹Am, ²⁴³Am and ²⁴⁴Cm. However, the interpretation of the measurements is complicated and a reanalysis of these experiments would be valuable.

The compositions of irradiated fuel samples are sensitive to both the capture and fission cross-sections. The analysis of the composition as a function of burn-up can provide a valuable data check. Such analyses have been made, for example, by Takano et al (1990) and Mounier (1998). Mounier has also derived the adjustments to be applied to key cross-sections, including the capture cross-sections of ²³⁶U, ²³⁷Np, ²⁴¹Am, ^{242m}Am, ²⁴³Am, ²⁴²Cm, ²⁴³Cm, ²⁴⁴Cm and ²⁴⁵Cm, and the fission cross-sections of ^{242m}Am, ²⁴³Cm and ²⁴⁵Cm. The adjustments proposed to the thermal capture cross-sections of ^{242m}Am and ²⁴²Cm are substantial (-60% and +17%).

The results of the integral measurements are not readily accessible. It would be very helpful if these could be compiled in a form which would allow them to be analysed, although it is recognised that, in the case of reactor irradiations the complexity of the reactor composition could make the specification and the analysis complicated.

In addition to the requirements for integral data validation requests have been made for differential measurements, as follows:

Fission cross-sections. One can note the recent measurements by Fursov (Trieste Conf. and Gatlinburg Conf.). Measurements have been requested for ^{237}Np and ^{243}Am , although it is considered that these requirements have now been met. A review of the measurements is required.

Capture cross-sections. Probably the most important requirements are for the capture cross-sections, and the resonance structure. However, existing measured data might be adequate for some isotopes. There are requests for ^{236}U , ^{237}Np , ^{238}Pu , ^{241}Am , ^{243}Am .

Fission neutron spectra. The systematics has been reviewed by Ohsawa and Shibata (Gatlinburg Conf.). Measurements are requested by Japan. There is also a request for the delayed neutron spectrum of ^{244}Cm .

Fission neutron yield, η . Recent measurements have been made, for example, by Kokhlov (Gatlinburg Conf.). Measurements are requested for ^{237}Np , ^{241}Am and ^{243}Am , although these requirements might be met by the measurements of Kokhlov (^{237}Np and ^{243}Am).

Inelastic scattering, $(n,2n)$. Measurements are requested for ^{237}Np , ^{238}Pu , ^{241}Am , and ^{243}Am .

Delayed neutron yields. There are requests for ^{237}Np , ^{238}Pu and ^{241}Am .

7. Requirements for the Thorium Cycle.

The isotopes of particular interest are ^{232}Th , ^{231}Pa , ^{233}Pa , ^{233}U and ^{234}U . The evaluations for ^{232}Th and ^{233}U in the JEF-2.2 library are quite old, dating from ~1967. The ENDF/B-VI evaluations date from 1978, with the addition of Olsen's resonance region analysis (1982) for ^{232}Th . The JENDL-3.2 evaluations were partly updated in 1993/94; the resonance region data were revised to include the measurements of Kobayashi (1987) for ^{232}Th and the evaluation of Derrien (1994) for ^{233}U . There are more recent measurements, such as the Geel total cross-section measurements, 1.5 to 18 MeV (Gatlinburg, 1994) still to be taken into account. The first requirement is for a comprehensive review of the status of the measured data and the second is for integral data analyses.

Subgroup 14 of the WPEC (coordinator, Ignatyuk) is reviewing data for the thorium cycle and has prepared 4 thorium benchmark specifications. Probably the most important requirement at the present time is a compilation of integral data suitable for validating the data for the thorium cycle.

Comparing the high energy fission cross-sections in the files:

Isotope	JEF-2.2	ENDF/B-VI	JENDL-3.2
²³² Th	0.0707	0.0743	0.0777
²³¹ Pa	0.981	0.981	0.826
²³³ Pa	0.460		0.323
²³³ U	1.841	1.911	1.949
²³⁴ U	1.215	1.215	1.211

Table 6. Comparison of fission spectrum averaged fission cross-sections.

For the thermal energies we have:

Isotope	JEF-2.2		ENDF/B	-VI	JENDL-	3.2
	thermal	R.I.	thermal	R.I.	thermal	R.I.
²³³ U	526	752	529	746	531	763

Table 7. Comparison of thermal fission cross-sections and resonance integrals for ²³³U.

We see that the thermal fission values for ²³³U don't differ markedly between the files.

For thermal capture the values are as follows:

Isotope	JEF-2.2		ENDF/B	-VI	JENDL-	3.2
	thermal	R.I.	thermal	R.I.	thermal	R.I.
²³² Th	7.41	85.0	7.42	85.5	7.40	83.9
²³¹ Pa	227	594	228	595	201	593
²³³ Pa	41.5	854			40.0	861
²³³ U	46.0	134	46.0	136	45.3	138
²³⁴ U	103	659	103	659	100	630

Table 8. Comparison of thermal capture cross-sections and resonance integrals.

There are no marked differences between the files so far as these parameters are concerned.

There are requests in the High Priority List for measurements of the capture cross-sections of ²³²Th, ²³¹Pa, ²³³Pa and ²³³U, and for inelastic scattering in ²³²Th. There is also a request for measurements of the delayed neutron yields in ²³²Th and ²³³U.

The data for ²³²Th and ²³³U have not been the subject of the same in depth analyses as have the cross-sections for ²³⁵U, ²³⁸U and ²³⁹Pu. Data of comparable accuracy are required for ²³²Th and ²³³U but it is unclear, in the absence of such reviews, that further capture and fission cross-section measurements are required. We note the recent capture measurements for ²³²Th by Kobayashi (1987) and the resonance region analysis for ²³³U by Derrien.

8. Concluding Remarks.

A document is needed giving the background to each request in the High Priority Request List. In many cases it is simply a statement that this item of data is important and the requester isn't aware of the status of the data.

It is important that estimates of uncertainties in the data be included in the files. The evaluator is in the best position to assess the uncertainties and it shouldn't be left to the user to make his own judgement. Without information on the present uncertainties deciding on the need for measurements is made more difficult.

Integral measurements have an important role in assessing the accuracy of the fission product and minor actinide data in the files. Compilations of integral measurements suitable for making these assessments are needed.

Differential measurement requirements should be limited to a few important isotopes, such as cross-sections of ^{99}Tc , ^{129}I , ^{237}Np , ^{241}Am , ^{232}Th and ^{233}U , until more comprehensive assessments of the requirements and the status have been carried out. However, there are requirements for integral measurements in different spectra for all of the isotopes of interest. In many cases the integral measurements have already been made, although they aren't always readily available.

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