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General Comments on the Nuclear Decay Data Requirements for JEFF-3 and the Evaluation Procedure.

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Abstract:

The purpose of this note is an attempt:

- to identify the main decay data needs for current fission reactors and emerging nuclear systems.
- to underline the difficulties to proceed to a rigorous and complete evaluation procedure, involving a sophisticated treatment of uncertainties propagation using variance-covariance (data often lacking in current libraries) and a qualification of the decay data against integral reactor parameters.
- to suggest a method to ensure the internal (and external) consistency of a Decay Data File in ENDF-6 format, and in particular to stress the data lacking in this format to proceed to these internal consistency tests.
- to review briefly some of the libraries to complement the JEF-2.2 file.

1°) Introduction:

We are not dealing here with the evaluation procedure of the data recorded in the General Purpose File of any ENDF library as specified in the ENDF-6 format (see ref. ENDF-102), mainly devoted to cross-section data (and some of the fission data), but with the decay data available (together with the fission yields) in the ENDF Special Purpose File.

However, in order to build the decay chains for the inventory calculation of both the stable and radioactive nuclides in fission reactors, it is necessary to know all the channels energetically possible for the production and disappearing modes by radioactive decay and/or nuclear reaction of all the nuclides available in the ENDF libraries. It is in particular necessary to supplement the ENDF (cross-section data) library (e.g. JEF or ENDF/B) with other external nuclear data files such as:

- the EAF file to take into account all the activation reactions actually known, e.g. as it is done in the DARWIN inventory code developed at CEA.
- the ENSDF and AME-95 data files for nuclear structure, decay, and mass data. Such basic nuclear data are for instance required for the prediction (using nuclear models as available in the GNASH code) of unmeasured cross-sections for high-spin nuclides involved in new nuclear systems to transmute long-lived radioactive wastes.

That is why a master file or a so-called Decay Chains File (DCF) would be very useful to test the complementarity and the consistency of these required nuclear data files, and to ensure the traceability of the progress made.

2°) Data Required to build the Decay Chains in Nuclear Energy Systems

The summary list of physical data required for this purpose, without consideration of available libraries or format, is the following:

a°) For any type of nuclide:

- spin, parity, level state (isomeric or ground state) with excitation energy if necessary.
- masse excess
- abundance (for stable nuclide) or half-life (for radioactive nuclide).
- production mode: fission, activation, decay, natural element.
- disappearing mode: decay (modes, intensities, decay energy Q_i), fission, or other nuclear reaction (MT index in ENDF format).
- binding and reaction energies: S_n , S_p , S_{2n} , S_{2p} , Q_α , $2\beta^-$, Q_{ep} , $Q(\beta^-, n)$, $Q4\beta^-$, $Q(d, \alpha)$, $Q(p, \alpha)$, $Q(n, \alpha)$...

NB: these data are important to state about the energetically open reactions/disintegrations to build the decay chain for the inventory calculation.

- evaluation source. NSR reference (Burrows-1990) and also CINDA, but NSR is more appropriate for Decay data and Fission yields

b°) For radioactive nuclides:

- list of emitted particles, associated spectra and decay energies
- deviation in the energy balance between: $Q_{\text{eff}} = \sum_i Q_i B_i$ and $Q_{\text{calc}} = \sum_i P_i E_i$

c°) For Fission Products:

- independant and cumulative fission yield (for spontaneous fission, or neutron induced fission on thermal, fast, or fusion spectrum).
- interpolation law versus energy of the incident neutron.

NB:

data may be required for gammas or charged particles induced fission (e.g. transmutation of wastes based on accelerator driven system), especially for high-spin nuclides (see intermediate Energy Data specialist's Meeting;)

d°) For Delayed Neutron precursor nuclides: P_n value and DN spectrum.

e°) For fissile nuclides, a consistent set of fission data including:

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- i) energy release due to fission E_f (with all prompt and delay components)
- ii) ν_p, ν_d, ν_t , and associated neutron spectra
- iii) $\{\lambda_i, \alpha_i\}, \beta_{eff}$,
- iiii) list of available Fission Yield Data sets in the ENDF library.

3°) Criteria to determine Priority Levels for Data Requirement.

Of course we have to distinguish different priority levels in the nuclear decay data requirements according to the following (quantitative/qualitative) features:

- the required number of nuclides to be evaluated:

The decay chain length may differ from one nuclear application to another one. It is sometimes feasible (resp. necessary) to simplify (resp. to extend) the decay chains describing only the main nuclides contributing to the capture effect, decay heat, radiotoxicity ...

- the associated types of nuclear reactions/disintegrations to be described and therefore evaluated.

- the reliability (or confidence) level required for these nuclear data with respect to the safety related parameters of the application (or nuclear system under study), i.e. the achieved data uncertainty through the evaluation process compared to the target uncertainty:

i) compilation and assessment of the status of the existing data

ii) internal consistency (e.g. level scheme, energy balance), external consistency (comparison of the same data evaluated in different libraries) and identification of data discrepancies

iii) evaluation/recommendation of data, including a validation of microscopic data against integral reactor parameters). In some cases, it is not clear whether the data are resulting from a rigorous objective and complete evaluation procedure with a statistical treatment of discrepant data (and a careful analysis of error correlations using full variance-covariance data) or only based upon subjective assessments/assumptions of published experimental and theoretical data. This topic, and different objective procedures to evaluate the decay data and their uncertainties, are discussed in detail by Nichols and al - 1994, and Mac Mahon et al. - 1994.

Unfortunately, the full evaluation process is not rigorously completed because of a lack of information (e.g. measured data not available, or experimental covariance input data not provided), lack of manpower in this field of expertise, or just because it is too much time consuming. That is why some of the data stored in the Evaluated Nuclear Data Files are resulting from an evaluation without a full internal consistency required to fulfill some of the consistency checks with recommended data for the main nuclides of interest in reactor applications.

4°) Main Features and Limitations of the ENDF and ENSDF Decay Data Libraries and Format.

4.1 Utilization of ENSDF

One would be attempted to distinguish short-term and long term decay data requirement with two basic types of nuclear data library:

- a decay data library containing recommended values for current nuclear systems, fission reactors and safety related parameters. This Applied Data File has to be frozen for a given period, with a policy of regular updates to improve the data or to include new ones. This is in principle the aim of the radioactive decay data recorded in any ENDF (Evaluated Nuclear Data File) Special Purpose File (MF =8, MT = 457), as it is specified in the ENDF-6 format (ref. ENDF-102), e.g. the ENDF/B-VI or JEF-2 decay data files.
- a more comprehensive library (or Chart of nuclides), not connected with the application purpose, including the recent progress in nuclear spectroscopy, with an regularly updated inventory of all nuclides already known as it is done in ENSDF (the Evaluated Nuclear Structure Data File),

One also has to mention that now the data are available:

- ENSDF On-Line Retrieval System (BNL-NNDC, NEA). Two options can be used
 - Telnet facility
 - WEB (www.nndc.bnl.gov)
- New codes running on PC with data on Cdrom.
 - PCNUDAT and Papyrus NSR) to visualize the ENSDF/NSR data (free)
 - TABLE OF ISOTOPES (LBL , John Wiley) , Commercial, Cd rom
 - NUCLEUS with the NUBASE library, Orsay , AMDC, WEB

It would be then reasonable to think that the basic mean to update and to complement ENDF with consistent decay data would be to derive from ENSDF a new ENDF release containing the main decay properties required for most standard calculations of reactor parameters using the different programs operating on ENSDF, e.g. among them, the most important ones

- Codes for the conversion and the Physics analysis of the data

All these codes are also available on Web or Telnet (free)

Almost all can run on PC. A short description of some of them is given below.

. *RADLIST*: to calculate the energies, intensities, and dose rates of the atomic and nuclear radiations associated with the decay data of radionuclides.

. *LOGFT*: to calculate the data involved in the evaluation of the β^- , β^+ /e.c. (electronic capture), and internal bremsstrahlung spectra, such as the log (f.t) values, end-point β energies E_i^0 , $\langle E_{\beta i} \rangle$ and intensities $I_{\beta i}$.

. *GTOL*: to check the balance in the input and output gamma feeding on each level given in the nuclear decay scheme.

. *HSICC*: to calculate internal conversion and internal pair formation coefficients.

. *FMTCHK*: to check the ENSDF format.

. One could note that most of the JEF-2.2 decay data (JEF-Report 13) were already derived from ENSDF .

However, some other specific features and limitations of ENSDF have to be pointed out and compared to ENDF ones :

- ENSDF consists of approximately 3000 "data sets" arranged by mass (264) and ordered by Z and after by category (Adopted , Decay, Reactions, etc.. It is therefore a "*vertical*" evaluation, whereas an atomic mass evaluation (e.g. Audi et al. - 1995) or NUBASE is "*horizontal*" (set of properties over a range of mass).

The data are given by final level of the daughter nucleus. Therefore, the data sets of different decay modes have to be merged in input of the RADLIST computer program to process the decay properties and emitted radiations of a parent nucleus .

In ENDF format, the data are gathered for all decay modes of each radionuclide ().

Some of the uncertainties on half-lives are given asymmetrically and have to be symmetrized (A procedure has been developed in NUBASE for this purpose)

The propagation of uncertainties in an ENSDF evaluation is difficult due to a large amount of data, the correlations between quantities, and a lack of information provided by the authors (the propagation of large uncertainties based on a first-order Taylor expansion may not be valid). The variance-covariance matrices are not included in ENSDF. It is however possible to have available covariance data in ENDF decay data sub-section only when a continuous spectra is given.

On the other hand, although ENSDF may be considered as the main basic source of nuclear structure and decay data, some nuclides are not yet experimentally known by classical spectroscopic investigations and therefore not available in ENSDF. These data may however be required for a correct estimation of safety related reactor parameters (e.g. the decay heat, or the reactor kinetics controlled by the delayed neutron emission), and have to be evaluated in ENDF by other means (e.g. continuous spectra are measured in units of probability/eV or some data may completely be calculated by theoretical models). The lack for instance of decay data for short-lived fission products is leading to the underestimation of decay heat prediction by a summation method (Studsvik-87). Another case concerns the decay data that are already available in ENSDF but with an inconsistent level scheme because some nuclear levels may be missing by classical spectroscopy, leading to "Pandemonium" effect on the derived average decay energies, e.g. problem of balance between $\langle E_{\beta} \rangle$ and $\langle E_{\gamma} \rangle$ for short-lived and neutron riched fission products far from the β line of stability (Reich-85). New experiments have already improved the quality of data (see Rb 93 Contribution to this meeting)

That is why the evaluation procedure is much more complex to fulfil the nuclear data requirements, and one has to stress the features and limitations (or weaknesses) of both ENDF and ENSDF data libraries. We have therefore to distinguish the following cases (as described in next chapters):

- Data already available in ENDF but are still not satisfactory (even for current reactor applications, fuel cycle systems, and safety related parameters)
 - evaluation process still ongoing to improve any nuclear data (iterative)

- Data required in ENDF for current reactor applications but not available in ENSDF. (e.g. decay data for short-lived fission products).
- Data required for checking the internal ENDF consistency, but not available in ENDF format (auxiliary data could be taken from ENSDF or from an other source).
- Data neither evaluated (or not yet included in the format) in recent ENDF data libraries (JEF-2 or ENDF/B-VI) nor in ENSDF, but required for the study of emerging fuel cycle systems (the number of evaluated nuclides or/and physical parameters are to be enlarged).

4.2°) **Data already available in ENDF but still not satisfactory**

(even for current reactor applications, fuel cycle systems, and safety related parameters)

One has to underline that some of the decay data are already available in the applied ENDF data libraries but may not be satisfactory/reliable because of :

- a large uncertainty on the data compared to the target data uncertainty (some of the data are measured with a low accuracy or are sometimes not yet experimentally available and derived from theoretical prediction).
- the existence of an obvious discrepancy, e.g. wrong normalisation in a level scheme as detected by an internal consistency check, or as born out by the users' feedback (in the calculation of an integral parameter and a sensivity study of this parameter to the suspicious datum).
- some data are available but discrepant and a best estimate has to be provided with the associated final uncertainty taking into account systematic/statistical errors, and therefore correlations among uncertainties. Two basic tools are available, the so-called "generalized error propagation law" and "the least squares procedure" to solve an overdetermined system of equations to get the best estimator of the observed data (Winkler-92). Different procedures have been developped to weight the discrepant decay data (as summarised by Nichols-94). This is in practice done (Winkler-92) for isolated data with unconstrained (weighted) averaging, e.g. for the half-life (see MacMahon et al. -1994 for the half-life data of ^{137}Cs , ^{90}Sr , ^{154}Eu , and ^{252}Cf) or some simple decay schemes (when a single-path approach can be followed in the evaluation, and assuming the input data as uncorrelated). It is much more difficult to treat the interrelated data with constrained fits. This is especially true for the evaluation of the energies of the emitted radiations (level-scheme correlations) and their intensities (balance conditions). That is why processing the full covariance information is still not the common procedure for both the input and output parameters in the decay data evaluation. One should also notice that in some evaluations, it is not clear wether the data are resulting from a rigorous objective and complete evaluation procedure with a statistical treatment of discrepant data or based on subjective assessments/assumptions of published experimental and theoretical data (Nichols-94). One has anyway to be very careful with an automatic processing of a nuclear level scheme and associated emitted radiations.

One has first to mention the IAEA international nuclear data programme to provide standard reference nuclear data files:

- the Nuclear Data Standards for Nuclear Measurements, NEANDC/INDC Nuclear Standards File, (OECD/NEA report, NEANDC-311 "U", INDC (SEC)-101, OECD/NEA 1992).
- the International Reactor Dosimetry File, IRDF (IRDF-90, IAEA-NDS-141).

Concerning the decay data, the international efforts were undertaken in the scope of two different working groups:

- the international network of Nuclear Structure and Decay Data NSDD (IAEA-INDC(NDS)-307).
- the International Committee for Radionuclide Metrology (Winkler-1994, ICRM-95)

In particular, two Coordinated Research Programs (CRPs) have been undertaken to produce a reference data file for:

- Transactinide nuclear data (IAEA-N° 261, 1986).
- X-ray and Gamma-ray Standards for Detector Calibration (IAEA-TECDOC-619, IAEA, Vienna, September, 1991).

We also have to mention the work of an IAEA specialists' group for the Development of an International Nuclear Decay Data and Cross-section Database (IAEA-INDC(NDS)-329).

However, among the important data to be further improved for practical applications and safety related problems, here are some typical examples:

- the nuclear data of short-lived products, e.g. fission products yields and decay data involved in decay heat prediction or delayed neutron data for the calculation of reactor dynamics (Michaudon-92, Dickens-91, Rowlands-92&96, Das-94).
- the radiation and decay characteristics of very long-lived activation products to be well-known in particular for fission reactor decommissioning (Kocherov-93, Chechev-91), e.g. large discrepancies for the half-life of ^{118m}Ag (three different values are available: 127 ± 7 y, 418 ± 15 y and 310 ± 132 y) and ^{59}Ni (five different values in between $7,58 \cdot 10^4$ - $7,58 \cdot 10^5$ y) as pointed out by Manokhin (Manokhin-94 in INDC(NDS)-329).
- the decay data of important long-lived fission products (see MacMahon et al. -1994 for the half-life data of ^{137}Cs , ^{90}Sr).
- the decay data of transactinides nuclides (IAEA-TECDOC-336-1984, IAEA-TRS-261-1986), and

That is why a general High Priority Request List (HPRL) for JEFF-3 has been established (Rowlands-96), and a special one has also been produced (JEF/DOC- 603) for Decay and Fission Yield Data (Special Purpose File in ENDF-6 format) based on national request lists from UK (Nichols-96, JEF/DOC-601&602) and France (Blachot et al., JEF-DOC/507).

It is also important to mention the list of 214 isotopes requiring reference data as selected by the USNDN sub-task force on nuclear decay data (IAEA-INDC(NDS)-307,) but only **one nuclide was evaluated in 1996**

4.3°) Data required in ENDF for current reactor applications but not available in ENS DF.

(e.g. decay data for short-lived fission products).

Some decay data are not experimentally available and than have to be derived partially or totally from theoretical models or empirical equations (e.g. missing levels in a decay scheme, or mean decay energies and Pn values not measured ...). One has to mention in particular new means to estimate the β strength function (Klapdor-83&91, Yamada-88, Kratz-83).

Some other data are resulting from continuous β and γ spectroscopic measurement (Rudstam-92) leading directly to the mean β and γ values on all the nuclear levels of the daughter nuclide decay scheme. In both cases, the data are external to ENSDF.

A typical example is the evaluation of the β^- and (β^- , n) decay data of short-lived fission products (Dickens-91, Das-94), e.g. in the JEF-2.2 (Blachot et al -92) and ENDF/B-VI (England et al -92a) data files. The supplemented new data have an obvious impact on integral safety related parameters such as the decay heat and delayed neutron emission (Michaudon-92, England-92b, Storrer-94, Blachot et al. -97).

4.4°) Data required for checking the internal ENDF consistency, but not available in ENDF format (auxiliary data could be taken from ENSDF).

The purpose of this chapter is to underline the lack of data in the ENDF-6 format to check the internal consistency of an ENDF decay data library, or the lack of information in the ENDF-6 procedure to use correctly the data.

As already mentioned, one of the basic procedure to check the internal consistency of a proposed decay scheme is to verify the following equation:

$$Q_{\text{eff}} = \sum_i Q_i B_i \quad \cong \quad Q_{\text{calc}} = \sum_i P_i E_i$$

The separated energy components not available in the ENDF-6 format to proceed to this simple consistency check are the energy of the:

- neutrinos (not released but not negligible in the energy balance).
- internal bremsstrahlung.
- recoil nucleus (generally quite small)

Moreover, it would be necessary to have available the log ft values to check the β and neutrinos energy balance.

It is also sometimes confusing to have both discrete and continuous spectra given without a recommended one. In such a case, the average decay energy derived from the discrete spectrum is not consistent with the datum given in ER_{AV} . Is it necessary to renormalize the discrete spectrum to the actual ER_{AV} given, unless we may have discrepant results between:

- the β (and/or γ) decay heat calculation by a summation methods (using the pseudo α , β , and γ average decay energies).

and

- the β (and/or γ) spectrum of aggregated FPs (using the ER_i for the discrete energy levels).

One should also note that the spectrum is in some cases not available whereas Q_i and ER_i are not equal to zero.

Some other internal inconsistencies were detected in JEF2.2 decay data applying some simple tests as described in the contribution of B. Nimal (Consistency tests of JEF2-2)

4.5°) Data neither in ENDF nor in ENSDF but required for the study of emerging fuel cycle systems:

The study of new strategies for the back end of the fuel cycle and emerging technology to incinerate the radioactive wastes (including calculations of activation and gas production in nuclear facility materials, transmutation of actinides and fission products in reactors or accelerator driven systems) expands the type and quantity of nuclear data required for both stable and radioactive nuclides because of the wider range of energy investigated (up to ~ 100 MeV for neutrons and ~ 1 GeV for protons in the entrance channel), leading to new nuclear reactions involved (e.g. spallation) and high-spin nuclides produced (some of them populated in isomeric states). Therefore nuclear data are needed for about 2500~3000 nuclides and the associated decay chains.

Because most of the exotic nuclear reactions involved in these new nuclear systems are not experimentally available, comprehensive nuclear model are utilized to perform cross-section calculations.

For that purpose, basic data are required as input parameters, e.g. atomic mass, ground-state spin and parity data, nuclear structure data, (and optical model transmission coefficients, see GNASH input data, P.G. Young-1992).

5°) Available Data Libraries to supplement JEF-2 decay Data Library and Proposal for a JEFF-3 Evaluation Strategy

A possible policy to update JEF2-2 decay data and future JEF3 would be (as a first step):

- to update and complement JEF2.2 with a critical compilation of the ENSDF, NUBASE, UKHEDD-2, UKPADD-
- to include the new measured data for short-lived FPs.
- to supplement the JEF2.2 cross-section data file with the EAF activation file to build the more comprehensive decay/reaction chains.
- to process a kind of Master File or "Decay Chains File" associated with a rigorous book-keeping for the date and source of evaluated data, and a statement about the reliability.

It is very important to provide reactor physicists with a kind of Master File or Decay Chains File for the study of new nuclear technology to transmute the long-lived radioactive wastes.

Conclusion

The aim of this note is to look for a common strategy for the evaluation of the JEFF-3 Decay Data File and to stress the questionmarks about the means to achieve such a task (manpower, evaluation procedure, input data source, checking codes, priority-levels).

We have attempted to review the main features and limitations of two important types of decay data libraries, i.e. the Special Purpose File of any ENDF library (JEF-2.2 or ENDF/B-VI), and the ENSDF nuclear structure data file.

ENSDF associated with the RADLST program could be one of the important mean to check the internal consistency of the decay scheme of some radionuclides suspected in JEF-2.2.

The external consistency check would be to compare the JEF evaluation with the data evaluated in other libraries such as UKHEDD-2 (for heavy elements), UKPADD-3 (for activation products), and NUBASE.

It is however necessary to ensure a constant effort for the evaluation of :

- the data already available in JEF-2.2 but requiring an improvement, especially for short-lived and very long-lived radionuclides. The problem in particular for short-lived fission products is the lack of information in ENSDF about their level scheme as derived from classical spectroscopic measurement. Theoretical evaluations or data from continuous measurement of beta decay properties are therefore to be searched.
- the mass and basic nuclear data for nuclides and activation products involved in the study of new nuclear systems for the wastes transmutation. There is an obvious need to extend the decay chains for both stable and radioactive nuclides produced by the nuclear reactions induced by neutrons and protons at high energy. That is why it would be very helpful to provide the reactor physicists with a kind of Master File or a comprehensive "Decay Chains File" in order to make an inventory of the known nuclides, nuclear reaction and disintegration processes energetically opened for their production and disappearance.

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