Delayed Neutron Energy Spectrum from a Specific Precursor

(O. Schwerer et al., Memo CP-C/429, V. Semkova, et al., Memo CP-D/837)

Memo CP-C/429 (2014-05-23)

After discussions during and after the NRDC meeting, we propose some modifications and additions to the coding proposed in 4C-3/396 and CP-D/837, as follows:

The delayed neutron emission probability (=probability for emission of at least one beta-delayed neutron), P_n, is coded as `,PN` with units `NO-DIM`.
The probability of emission of N beta-delayed neutrons, P_{Nn}, is coded as `NUM,PN` with units `NO-DIM`. *(No change from 4C-3/396)*
The delayed-neutron emission multiplicity, <n>, is coded as `,MLT,DN` with units `PRT/DECAY` or `PC/DECAY`.
The energy spectrum of delayed neutrons emitted by a specific precursor is coded as `,PN/DE` with units of dimension `1/E`. *(Modified from CP-D/837)*

Appended below are the proposed revised Lexfor page D.6 and a summary of the necessary dictionary updates.

Revised LEXFOR page D.6 (last part of chapter „Delayed Fission Neutrons")

Delayed-Neutron Emission Probability (P_n value)

Definition: Probability for emission of at least one β-delayed neutron

REACTION Coding: `(Z-S-A(0,B-)[Z+1]-S'-A,,PN)`

Units: `NO-DIM`

For delayed neutron emission probabilities see for example, Amarel [4], Tomlinson [5], and Asghar [6].

Probability of emission of N β-delayed neutrons (P_{Nn})

Definition: Probability to emit N neutrons after β decay

REACTION Coding: `(Z-S-A(0,B-)[Z+1]-S'-A,NUM,PN)`

Units: `NO-DIM`

The number of emitted neutrons is given under the data heading `PART-OUT` with units `NO-DIM`.

Delayed-neutron emission multiplicity <n>

Definition: Multiplicity of delayed neutrons per decay

$$<n> = P_{1n} + 2*P_{2n} + 3*P_{3n} + \ldots.$$

REACTION Coding: `(Z-S-A(0,B-)[Z+1]-S'-A,MLT,DN)`

Units: `PRT/DECAY` or `PC/DECAY`

Energy spectrum of delayed neutrons emitted by a specific precursor

REACTION Coding: `(Z-S-A(0,B-)[Z+1]-S'-A,,PN/DE)`

Units: a code from Dictionary 25 with dimension `1/E` (*e.g.*, `1/KEV`)

1.

1. From the above definitions follows $P_n = P_{1n} + P_{2n} + P_{3n} + \ldots$

If only one neutron emission is energetically possible, $P_n = P_{1n} = <n>$, the coding `,PN` must be used.

1. Data not Presently Compiled in EXFOR

- The energy spectrum of all delayed neutrons together, which is time dependent, due to the contributions from the different half-life groups.
- The delayed-neutron equilibrium spectrum as found in a steady-state reactor.

References

[1.] S. Amiel, *IAEA Panel on Fission-product Nuclear Data*, Bologna, 1973, IAEA report IAEA-169, Vol. II (1973) p. 33

[2.] G.R. Keepin, Physics of Nuclear Kinetics (Addison-Wesley, 1965) Chapter 4

[3.] E.K. Hyde, The Nuclear Properties of Heavy Elements, Vol. III (Prentice Hall, 1964) p. 261 ff.

[4.] I. Amarel, *et al*., J. Inorg. Nuc. Chem., **31**, 577 (1969)

[5.] L. Tomlinson, *et al*., J. Inorg. Nuc. Chem., **33**, 3609 (1971)

[6.] M. Asghar, *et al*., Nucl. Phys. **A247**, 359 (1975)

[7.] I. Dillmann, *et al.,* (ed.), INDC(NDS)-0643, IAEA, Vienna (2014)

(End of Lexfor page)

Related dictionary updates

Dictionary 25 (Data Units)

Change:

PC/DECAY *change definition to:* Particles per 100 decays

Add:
PRT/DECAY Particles per decay

Dictionary 33 (Particles)
Change:
DN *Add flag 7*

Dictionary 213 (Reaction Type)
Add:
PNE NUD MFQ 4* 3 Delayed neutron spectrum from individual precursor

Dictionary 236 (Quantities)
Change:
,PN *Change unit dimension to* NO
Add:
,MLT,DN MLT PN Delayed neutron emission multiplicity
NUM,PN PN NO Probability of emission of N delayed neutrons
,PN/DE PNE 1/E Spectrum of delayed neutrons emitted by specific precursor

Memo CP-D/837 (2014-04-09)

Beta-delayed neutrons emitted from some of the fission products play an important role in the control and in the safety aspects of nuclear reactors. Large uncertainties of the delayed neutron data used in reactor calculations lead to costly conservatism in the design operation of reactor control system. Beta-delayed neutron emission probabilities have been experimentally studied and evaluated during the last decades, yet experimental information is only available for less than half of the identified n-emitters (precursors), and unfortunately many of these measurements give only upper limits or approximate values. In recent years novel detectors have been built to operate at major accelerator facilities to measure the delayed-neutron decay characteristics of individual precursors, in synergy with quantification of aggregate properties involved in the fissile materials. Current research interest in this direction is motivated by the need for better calculations of the decay heat in reactors, planning of future advance fuel technology, anti-neutrino spectra from reactors, r-process nucleosynthesis, and nuclear structure physics. Although half-lives and P_n data can be found scattered in several compiled and evaluated libraries such as ENSDF, NUBASE, NuDat, Wallet Cards, etc., complete documentation of measurements is often missing. Moreover, measured neutron spectra are not available in any database.
The dynamics of the nuclear transformations through decay at different stages after nuclear fission process is measured by mass yields and charge dispersions as well as energy distributions. Such data are compiled in EXFOR as primary fission-fragment yield, secondary fission-fragment yields, independent fission-product yields, cumulative fission-product yields etc. However, regarding the beta-delayed neutrons emitted from some of the fission products, mainly data for the six-group presentation are compiled. Availability of data for individual precursors will allow more precise evaluations of total number of delayed neutrons per fission emitted from different

fission systems since the fission yields of the individual precursors vary. Therefore we would like to propose extension of the compilation scope to the delayed neutron spectrum for a specific precursor.
Although as it was mentioned the beta-delayed quantities are properties of the fission product nucleus and most of the experimental methods define the delay neutrons per decay of the individual precursor information about the process involved in the production of neutron reach isotopes could be useful for users. The following codes for the keyword METHOD are proposed NIFIS (neutron induced fission), PIFIS (proton induced fission), HIIFR (heavy-ion induced fragmentation), LIISP (light-ion induced spallation LCP<=4). Different measurement techniques are employed to measure beta-delayed neutron emission probabilities and spectral distributions such as: beta and neutron spectrometry, double gamma spectrometry, gamma and neutron spectrometry, beta-neutron coincidence technique, beta-gamma coincidence technique, BRICT beta-recoil ions coincidence technique. Summary of the measurement techniques from the Report INDC(NDS)-0599 is included in APPENDIX I. The following new codes for the keyword METHOD are proposed NSPEC (neutron spectrometry) and RISPC (recoil ion spectrometry).

In some articles neutron spectrum is reported in arbitrary units as relative intensity and in some articles the spectrum is normalised to number of the emitted delay neutrons per decay. Examples for the compilations of the two types of data are given below.

Example

```
(---(0,B-)SF4,SF5,SF6,,NPD)
```
β-delayed neutron spectrum normalized to the probability distribution
```
(---(0,B-)---,, PN/DE,,REL)
```
β-delayed neutron spectrum in arbitrary unit
```
(---(0,B-)---,,PN/DE)
```
β-delayed neutron spectrum in neutrons/100 decays/MeV

Table I. Measurements of Delayed Neutron Spectra for EXFOR compilation.

Author	Reference	Laboratory
Rudstam+	J,NSE,80,238,1982	2SWDSWR
Rudstam+	J,NSE,64,749,1977	2SWDSWR
Kratz+	J,NP,317,335,1979	2GERMNZ
Franz+	J,PRL,333,859,1974	2GERMNZ
Shalev+	J,NP/A,230,153,`974	2SWDSWR
Kratz+	INDC(NDS)-107/G,103,1979	2GERMNZ
Batchelor+	J,JNE,3,7,1956	2SWDSWR
Rudstam+	J,NIM,120,333,1974	2SWDSWR
Rudstam+	J,NP/A,235,397,1974	2SWDSWR
Shalev+	J,NP/A, 275,76,1977	2SWDSWR
Greenwood+	J,NSE,91,305,1985	1USABNL
Greenwood+	J,NSE,126,324,1997	1USABNL
Kratz+	J,ZP/A,312,33,1983	2GERMNZ
Reeder+	J,NSE,75,140,1980	1USABNW
Franz+	J,NIM,144,253,1977	2GERMNZ
Shalev+	J,PRL,28,697,1972	2SWDSWR
Ohm+	J,NP/A,274,45,1976	2GERMNZ

References:

L. Mathieu, O. Serot, T. Materna, A. Bail, U. Koster, H. Faust, O. Litaize, E. Dupont, C. Jouanne, A. Letourneaud and S. Panebiancod, “New neutron long-counter for delayed neutron investigations with the LOHENGRIN fission fragment separator” JINST 7 P08029 doi:10.1088/1748-0221/7/08/P08029


```
ENTRY          31739   20140317                                    31739  0    1
SUBENT        31739001   20140317                                  31739  1    1
BIB                11        31                                    31739  1    2
TITLE       Investigation of beta strength functions by neutron    31739  1    3
            and gamma-ray spectroscopy (I). The decay of 87Br,     31739  1    4
           137I, 85As and 135Sb                                    31739  1    5
AUTHOR     (K.-L.Kratz,W.Rudolph,H.Ohm,H.Franz,M.Zendel,           31739  1    6
            G.Herrmann,S.G.Prussin,F.M.Nuh,A.A.Shihab-Eldin,       31739  1    7
            D.R.Slaughter,W.Halverson,H.V.Klapdor)                 31739  1    8
INSTITUTE  (2GERMNZ,1USAUCX,2GERMPH)                               31739  1    9
REFERENCE  (J,NP/A,317,335,1979)                                   31739  1   10
           (J,PRL,28,687,1972) Pulse-height distribution of        31739  1   11
           delayed neutrons from 137Xe (precursor 137I) on graph.  31739  1   12
           (J,PRL,33,859,1974)Delayed-neutron spectra following    31739  1   13
           decay from 85As and 135Sb on graphs.                    31739  1   14
FACILITY   (REAC,2GERMNZ) Mainz TRIGA reactor operated in a        31739  1   15
         pulse mode FWHM 30 msec integrated fluxes 10**14 n/cm2    31739  1   16
METHOD     (CHSEP,JET)                                             31739  1   17
           (NIFIS) Fission products were produced by neutron-ind.  31739  1   18
            235U fission. Samples 0.1-1.0 mg 235U dissolved in HCL 31739  1   19
            or HNO3.                                               31739  1   20
DETECTOR   (HE3SP) Two identical 3He ionization chambers.          31739  1   21
            Resolution of about 12 keV for thermal neutrons and    31739  1   22
            20 keV for 1 MeV neutrons.                             31739  1   23
REL-REF    (I,,H.Franz+,J,NIM,144,253,1977) Detectors and data     31739  1   24
           analysis.                                               31739  1   25
CORRECTION Neutrons from bulk fission products less that 0.4% of   31739  1   26
            total count rate and less that 0.02% had energy        31739  1   27
            greater than 40 keV.                                   31739  1   28
            Scattered neutrons from the sample less that 7%.       31739  1   29
ERR-ANALYS No information on sources of uncertainties.             31739  1   30
           (E-ERR-DIG) Error in dig. the secondary energy values.  31739  1   31
           (ERR-DIG) Error in digitizing data values.              31739  1   32
HISTORY    (20140317C)                                             31739  1   33
ENDBIB             31         0                                    31739  1   34
COMMON              2         3                                    31739  1   35
E-ERR-DIG  ERR-DIG                                                 31739  1   36
KEV        ARB-UNITS                                               31739  1   37
2.         2.                                                      31739  1   38
ENDCOMMON           3         0                                    31739  1   39
ENDSUBENT          38         0                                    31739  199999
SUBENT        31739002   20140317                                  31739  2    1
BIB                 3         4                                    31739  2    2
```

```
REACTION   (33-AS-85(0,B-)34-SE-85,,PN/DE,,REL)                31739  2    3
DECAY-DATA (33-AS-85,2.05SEC)                                  31739  2    4
STATUS     (CURVE) Scanned from Fig. 1 (upper part) of Nucl.   31739  2    5
            Phys. A317 (1979) 335.                             31739  2    6
ENDBIB            4          0                                 31739  2    7
NOCOMMON          0          0                                 31739  2    8
DATA              2        595                                 31739  2    9
E          DATA                                                31739  2   10
KEV        ARB-UNITS                                           31739  2   11
     15.91      3.00                                           31739  2   12
     20.87     15.64                                           31739  2   13

ENTRY            31743   20140403                              31743  0    1
SUBENT        31743001   20140403                              31743  1    1
BIB               9         21                                 31743  1    2
TITLE          Energy spectra of delayed neutrons from the     31743  1    3
               precursors79(Zn, Ga),80Ga, 81Ga,94Rb, 95Rb,129In, and 31743  1    4
               130In                                           31743  1    5
AUTHOR     (G.Rudstam,E.Lund)                                  31743  1    6
INSTITUTE  (2SWDSWR)                                           31743  1    7
REFERENCE  (J,NSE,64,749,1977)                                 31743  1    8
FACILITY   (REAC,2SWDSWR)                                      31743  1    9
METHOD     (NIFIS) Precursors were produced by neutron-induced 31743  1   10
            235U fission.                                      31743  1   11
           (OLMS) The separation of delayed neutron spectra from 31743  1   12
           isobaric precursors requires a proper timing of the 31743  1   13
           experiment and that an unfolding procedure using    31743  1   14
           half-lives of the preqursors.                       31743  1   15
           (NSPEC)                                             31743  1   16
REL-REF    (I,,G.Rudstam,J,NIM,120,333,1974) Details on the    31743  1   17
           techniques used in the measurements                 31743  1   18
DETECTOR   (HE3SP) Energy resolution (FWHM) ~20 keV for thermal 31743  1   19
            neutrons and ~35 keV for 1 MeV neutrons. The response 31743  1   20
            function determined by monoenergetic neutrons from 31743  1   21
            7Li(p,n)7Be and T(p,n)3He reactions.               31743  1   22
HISTORY    (20140403C) VS                                      31743  1   23
ENDBIB           21          0                                 31743  1   24
NOCOMMON          0          0                                 31743  1   25
ENDSUBENT        24          0                                 31743  199999
SUBENT        31743002   20140403                              31743  2    1
BIB               4          6                                 31743  2    2
REACTION   (31-GA-80(0,B-)32-GE-80,,PN/DE)                     31743  2    3
ASSUMED    (ASSUM,31-GA-80(0,B-),,PN)                          31743  2    4
ERR-ANALYS (E-ERR-DIG) Digitizing error of neutron energy      31743  2    5
           (ERR-DIG) Error in digitized values.                31743  2    6
STATUS     (CURVE) Data scanned from Fig.2b in Nucl.Sci. Eng. 64, 31743  2    7
            749(1977).                                         31743  2    8
ENDBIB            6          0                                 31743  2    9
COMMON            1          3                                 31743  2   10
ASSUM      E-ERR-DIG ERR-DIG                                   31743  2   11
```

```
PER-CENT KEV     PER-CENT                             31743  2   12
10.         2.         2.                             31743  2   13
ENDCOMMON          3         0                        31743  2   14
DATA             3         6                          31743  2   15
E-MIN     E-MAX     DATA                              31743  2   16
KEV       KEV       PC/DEC/KEV                        31743  2   17
   70.        80.     7.95E-05                        31743  2   18
  120.       130.     5.74E-05                        31743  2   23
ENDDATA            8         0                        31743  2   24
ENDSUBENT         23         0                        31743  299999
ENDENTRY           2         0                        3174399999999
```

LEXFOR update

Delayed Fission Neutrons

…

Decay Properties of Fission Product Nucleus *(new entry)*

There are delayed-neutron quantities that are not properties of the fissioning nucleus but of the fission-product nucleus that is the "precursor" of the delayed neutron, e.g., **delayed-neutron emission probability**, **delayed-neutron energy spectrum** for a specific precursor. They may be also compiled in EXFOR for users although they are not reaction data. Delayed neutron quantities for a specific precursor can be studied not only by production of the precursor by fission but can be also by other method (*e.g.*, light-induced spallation, heavy-ion induced fragmentation) [7].

Delayed-Neutron Emission Probability (Pn value)

….

Delayed-Neutron Energy Spectrum for a Specific Precursor (*New entry*)

REACTION Coding: `(Z-S-A(0,B-)Z′-S′-A′,,PN/DE)`

where: *`Z-S-A`* is the precursor nucleus before β decay);

Units: a code from Dictionary 25 with dimension `PNDE` (*e.g.*, `PC/DEC/MEV`)

Examples:

`(Z-S-A(0,B-)Z′-S′-A,,PN/DE)`	β-delayed neutron spectrum in neutrons/100 decays/MeV or neutrons/decay/MeV
`(Z-S-A(0,B-)Z′-S′-A′,,PN/DE,,NPD)`	β-delayed neutron spectrum normalized to the probability distribution
`(Z-S-A(0,B-)Z′-S′-A′,,PN/DE,,REL)`	β-delayed neutron spectrum in arbitrary unit

Data not Presently Compiled in EXFOR

- The energy spectrum of all delayed neutrons together, which is time dependent, due to the contributions from the different half-life groups.

- The delayed-neutron equilibrium spectrum as found in a steady-state reactor.
- ~~Delayed-neutron energy spectrum from individual precursor~~

References

…

[7] L. Mathieu et al., J. Instrum., **7**, P08029 (2012)

APPENDIX I

1.1.1 Methods for β-delayed neutron measurements and new data

1.1.1.1 Methods for measurements

Notation

Precursor (^{A}Z): **M**, mother

β-decay daughter ($^{A}Z+1$): **I**, intermediate

β-delayed neutron-daughter ($^{A-1}Z+1$): **F**, final nucleus (**F1** for β1n, **F2** for β2n)

$$P_n = N_{n\text{-}decays} / N_{decays}$$

1. "β/n coincidence method" (replaces "n/β" term to account for proper sequence of detection): Beta efficiency not required. Neutron efficiency is determined in absolute terms:

$$P_n = \frac{1}{\varepsilon_n} \frac{N_{\beta n}}{N_\beta}$$

   The main assumption in this method is that the number of counted β's is free of contaminations, i.e. the background is subtracted and the random noise is corrected.
   This method depends on the β-efficiency ε_β, e.g. Si detectors have a threshold of about 150 keV, so the ε_β-curve increases to typically about 25% detection efficiency at around 2 MeV.
   As for the neutron efficiency, the curve is assumed to be constant unless an energy-dependent efficiency curve is given. If the neutron energy distribution is very different from the calibrant isotope, this might induce systematic effects which cannot be corrected.
   Evaluators were advised to seek information on the β/n coincidence window. The correlation time should be long enough to capture also the high-energy neutrons which need more time to be moderated down to thermal energies.
   Evaluators were also advised that if neutron energy spectra are recommended from such measurements, they should be in linear scale, corrected for the efficiency, and normalized to 1.

   Conclusion: *"The n/β method is good if the efficiency of the neutron detector has been measured using a source with similar energy dependence as the precursor of interest."*

2. "n-β": Neutrons and β counted separately (no coincidences) but simultaneously (in the same experiment).

   The remarks on the neutron efficiency mentioned in the previous method apply also here. In addition, the dependence of ε_β on the β-endpoint energy is not trivial and can cause complications in the determination of the β-efficiency.
   It was acknowledged that in the past there was no possibility of correcting for increasing noise in the data analysis afterwards. Nowadays modern digital data acquisition systems have time-stamps which allow these corrections to be made after the measurement.

3. "γ $^{A}Z+n$": Abundance of precursor determined via γ-counting of any β-decay daughter. It was suggested that the label of this method be changed to: "n-γ", for n and γ counting.

Several drawbacks of this method were mentioned. First of all, absolute γ intensities have to be known. When fragmentation reactions are used for the production of the precursor nucleus, the βn-daughter might also be produced. The γ counting would then have to be corrected to account for this. Contamination of the sample from the activity of the daughter nucleus would have to be known. Additional problems could arise if isomers are present.
In an upper limit for the half-life of the β-decay daughter nucleus of $t_{1/2}(^{A}Z{+}n) \sim 10^{*}\, t_{1/2}(M)$ was given to avoid too many decay corrections. After review of this limit, it was decided that this is not necessary if the half-life is well-known because it could then be corrected accordingly.

4. "$P_n\,^{A}Z$": Normalization with respect to a known P_n value from precursor ^{A}Z.
   In this method, care should be taken that the chosen standard has a neutron energy spectrum similar to the investigated isotope.
   Only neutron counting is used. The P_n value is deduced by comparison of the investigated neutron rate to the rate of a neutron emitter with known P_n value:

$$P_n = \frac{N_n(isotope)}{N_n(standard)}$$

   If the reference isotope is not measured simultaneously then the normalization requires the use of production yields. In the latter case, the results are affected by the uncertainties in e.g. the fission yields.
   This method is not recommended for measurements with a cocktail beam in which several unknown neutron-emitters with very similar half-lives can be present.

5. "ion": Counting of number of precursors N_{ion}. The amount of βn daughters $N_{\beta n\text{-}daughter}$ is determined by any suitable method, and the P_n value deduced from this:

$$P_n = \frac{N_{\beta n-daughter}}{N_{ion}}$$

   The efficiency of the "ion counting device" should be carefully determined and known for both species, mother and daughter. This method also needs corrections for βn-daughters already present in the beam cocktail.
   There can be further subdivisions depending on the identification method: Fragmentation ranging-out, ΔE-TOF, ΔE-E, ion-βγ, and the detection methods: traps and storage rings.

6. "fiss": Determination of the number of precursors by fission yields

   As this method is strongly affected by the uncertainties in the fission yields, evaluators were advised not to use it unless it was the only available measurement. In such a case, they should bear in mind that the results would have to be adapted to the most recent evaluations of fission yields.

7. "γ-γ": pure γ-counting technique to determine both the number of mothers and βn-nuclei (granddaughters) produced.

   Absolute γ-intensities are required, that means a complete knowledge of the decay scheme including γ's going to the ground state or eventually competing γ-decays from levels above the neutron separation energy.

$$P_n = \frac{\varepsilon_{\gamma,d} \cdot I_{\gamma,abs,d} \cdot N_{\gamma,gd}}{\varepsilon_{\gamma,gd} \cdot I_{\gamma,abs,gd} \cdot N_{\gamma,d}}$$

   where d= "daughter" nucleus; gd= "granddaughter" nucleus.

   Another issue with this method is the direct β-feeding of the ground-state. The previous comment about counting time vs. half-life holds as in method 1 and 2: if half-lives are well-known, the granddaughter activity can be decay-corrected.

8. Ion-recoil method: This method includes the trap measurements It uses the recoil ions and time-of-flight measurement to deduce the neutron spectrum and can be complemented with γ-detectors. However, it is only feasible for P_{1n} measurements.

The aforementioned coincidence methods can be complemented with triple coincidences, e.g. βnγ or βγγ.