JAEA-Research
2016-019
DOI:10.11484/jaea-research-2016-019

Application of Probability Generating Function to the Essentials of Nondestructive Nuclear Materials Assay System using Neutron Correlation

Takashi HOSOMA

Technology Development Department
Tokai Reprocessing Technology Development Center
Nuclear Fuel Cycle Engineering Laboratories
Sector of Decommissioning and Radioactive Waste Management

January 2017
Japan Atomic Energy Agency
日本原子力研究開発機構

JAEA-Research

JAEA-Research 2016-019

Application of Probability Generating Function to the Essentials of Nondestructive Nuclear Materials Assay System using Neutron Correlation

Takashi HOSOMA

Technology Development Department
Tokai Reprocessing Technology Development Center
Nuclear Fuel Cycle Engineering Laboratories
Sector of Decommissioning and Radioactive Waste Management
Japan Atomic Energy Agency
Tokai-mura, Naka-gun, Ibaraki-ken

(Received November 7, 2016)

In the previous research (JAEA-Research 2015-009), essentials of neutron multiplicity counting mathematics were reconsidered where experiences obtained at the Plutonium Conversion Development Facility were taken into, and formulae of multiplicity distribution were algebraically derived up to septuplet using a probability generating function to make a strategic move in the future. Its principle was reported by K. Böhnel in 1985, but such a high-order expansion was the first case due to its increasing complexity.

In this research, characteristics of the high-order correlation were investigated. It was found that higher-order correlation increases rapidly in response to the increase of leakage multiplication, crosses and leaves lower-order correlations behind, when leakage multiplication is > 1.3 that depends on detector efficiency and counter setting. In addition, fission rates and doubles count rates by fast neutron and by thermal neutron in their coexisting system were algebraically derived using a probability generating function again. Its principle was reported by I. Pázsit and L. Pál in 2012, but such a physical interpretation, i.e. associating their stochastic variables with fission rate, doubles count rate and leakage multiplication, is the first case. From Rossi-alpha combined distribution and measured ratio of each area obtained by Differential Die-Away Self-Interrogation (DDSI) and conventional assay data, it is possible to estimate: the number of induced fissions per unit time by fast neutron and by thermal neutron; the number of induced fissions (< 1) by one source neutron; and individual doubles count rates. During the research, a hypothesis introduced in their report was proved to be true. Provisional calculations were done for UO_2 of 1~10 kgU containing ~ 0.009 wt% ^{244}Cm.

Keywords: Nondestructive Assay, Neutron, Correlation, Coincidence, Multiplicity, Differential Die-away Self-interrogation, DDSI, Probability Generating Function, Feynman-alpha, Rossi-alpha, *Y*-value, Safeguards, Nuclear Nonproliferation

中性子相関を利用する核物質非破壊測定システムの基礎への確率母関数の適用

日本原子力研究開発機構　バックエンド研究開発部門
核燃料サイクル工学研究所
再処理技術開発センター　技術部

細馬　隆

(2016年11月7日受理)

前回の報告（JAEA-Research 2015-009）では、中性子多重相関計数法の数理的基礎について、プルトニウム転換技術開発施設で得られたウラン・プルトニウム混合二酸化物の計量管理の経験を含めて、中性子多重相関計数法の数理的基礎について再考し、また将来への布石として七重相関までの多重相関分布式を、確率母関数を用いて代数的に導いた。導出の原理は K. Böhnel が 1985 年に報告しているが、複雑さのため七重相関までの拡張は初めてであった。

今回、高次の多重相関の基本的性質を調べた結果、高次相関は漏れ増倍率の増大に応じて急速に増大し、検出器の効率や設定によるが、漏れ増倍率が 1.3 を超えると、より低次の相関と交わり追い越してゆくことを見出した。続いて、高速中性子と熱中性子が共存する系のそれぞれの単位時間あたり核分裂数と二重相関計数率を、再度、確率母関数を用いて代数的に導いた。導出の原理は I. Pázsit と L. Pál が 2012 年に報告しているが、物理量との関連付け、則ち彼らの用いた確率変数を核分裂数や二重相関計数率及び漏れ増倍率と結びつけるのは初めてである。これにより Differential Die-Away Self-Interrogation (DDSI) 法により得られる Rossi-alpha 二重分布とそれらの面積比及び従来法の測定値から：高速中性子と熱中性子それぞれの単位時間あたりの誘導核分裂数；ソース中性子 1 個あたりのそれぞれの誘導核分裂数（< 1）; 及びそれぞれの二重相関計数率を求めることができる。また、彼らの報告で導入されていた仮説が正しいことを証明し、更に ^{244}Cm を 0.009 wt% 程度含む UO_2 1~10 kgU について、暫定的な計算を行った。

核燃料サイクル工学研究所：〒319-1194　茨城県那珂郡東海村村松 4-33

Contents

目次

Nomenclature

Neutrons arising from a fission does not include delayed neutrons from a fission product.

ν	The number of neutrons arising from a fission ($\overline{\nu}$ or $<\nu>$ is the expectation)
p	Probability of an event where a neutron produces an induced fission
l	Probability of an event where a neutron leaks from a sample
P_m	Probability mass distribution of multiple leakage neutrons composed of m neutrons
G	Probability generating function
p_ν	Probability distribution of the number of neutrons from a spontaneous fission
f_ν^1	Probability distribution of the number of neutrons from a fast induced fission
f_ν^2	Probability distribution of the number of neutrons from a thermal induced fission
ϵ	Efficiency of counter (counts per fission)
ϵ_n	Efficiency of counter (counts per neutron)
α	Rate of decay regarding prompt neutrons in a system or a sample ‘$e^{-\alpha t}$ will be the expected number of neutrons present at time t due to one primary neutron introduced into boiler’ by Feynman, Hoffmann and Serber (1956).
τ	Mean time between fissions
$1/(\alpha\tau)$	Total number of fissions by one source neutron
α_r	Ratio of neutrons arising from (α, n) reaction to those from spontaneous fission
λ	Probability of reactions (fission, detection, etc.) for a neutron for a unit time
Y	Measure of the excess in counting fluctuation expected in Poisson process
$\overline{F_1}$	Expected number of fast induced fission and spontaneous fission per unit time
$\overline{F_2}$	Expected number of thermal induced fission per unit time
$\overline{\nu_{s1}}$	First factorial moment (expected number of neutrons) of spontaneous fission
$\overline{\nu_{i1}^1}$	First factorial moment (expected number of neutrons) of fast induced fission
$\overline{\nu_{i1}^2}$	First factorial moment (expected number of neutrons) of thermal induced fission
$\overline{\nu_{s2}}$	Second factorial moment (twice of expected neutron pairs) of spontaneous fission
$\overline{\nu_{i2}^1}$	Second factorial moment (twice of expected neutron pairs) of fast induced fission
$\overline{\nu_{i2}^2}$	Second factorial moment (twice of expected neutron pairs) of thermal induced fission
α_1	Rate of decay for neutrons arise from fast induced fission and spontaneous fission
α_2	Rate of decay for neutrons arise from thermal induced fission
τ_1	Mean time between fast induced fissions or spontaneous fissions
τ_2	Mean time between thermal induced fissions
S	Count rate of neutrons = Count rate of singlets unrelated to correlation ≡ Singles
D	Count rate of neutron pairs = Count rate of doublets ≡ Doubles
T	Count rate of neutron pairs composed of three neutrons (triplets) ≡ Triples
D_1	Count rate of neutron pairs arise from fast induced fission and spontaneous fission
D_2	Count rate of neutron pairs arise from thermal induced fission
f_d	Fraction of gate effectiveness to count doublets (doubles gate fraction)
M_T	Total multiplication
M_L	Leakage multiplication

記号表

ここで「核分裂で生じる中性子」には核分裂片から生じる遅発中性子を含まない。

ν	1 回の核分裂で生じる中性子の数（$\overline{\nu}$ 又は $<\nu>$ はその期待値）
p	1 個の中性子が誘導核分裂反応を生じる事象の確率
l	1 個の中性子が試料から漏れる事象の確率
P_m	中性子 m 個 1 組から成る漏れ中性子の多重相関の組数の確率分布
G	確率母関数
p_ν	1 回の自発核分裂で生じる中性子の数の確率分布
f_ν^1	1 回の誘導核分裂（高速中性子起源）で生じる中性子の数の確率分布
f_ν^2	1 回の誘導核分裂（熱中性子起源）で生じる中性子の数の確率分布
ϵ	検出効率（counts per fission）
ϵ_n	検出効率（counts per neutron）
α	系又は試料中の即発中性子減衰定数
τ	核分裂の平均的な時間間隔
$1/(\alpha\tau)$	1 個の中性子から生じる核分裂の総数
α_r	（α, n）反応由来中性子数の自発核分裂由来中性子数に対する比
λ	単位時間あたり 1 個の中性子が反応（核分裂や検出等）を生じる確率
Y	計数率のゆらぎの Poisson 過程からの外れを示す指標
$\overline{F_1}$	単位時間あたり誘導核分裂（高速中性子起源）の数の期待値
$\overline{F_2}$	単位時間あたり誘導核分裂（熱中性子起源）の数の期待値
$\overline{\nu_{s1}}$	自発核分裂の一次階乗モーメントで、生じる中性子数の期待値
$\overline{\nu_{i1}^1}$	誘導核分裂（高速中性子起源）の一次階乗モーメントで、 生じる中性子数の期待値
$\overline{\nu_{i1}^2}$	誘導核分裂（熱中性子起源）の一次階乗モーメントで、 生じる中性子数の期待値
$\overline{\nu_{s2}}$	自発核分裂の二次階乗モーメントで、生じる中性子ペア数の期待値の二倍
$\overline{\nu_{i2}^1}$	誘導核分裂（高速中性子起源）の二次階乗モーメントで、 生じる中性子ペア数の期待値の二倍
$\overline{\nu_{i2}^2}$	誘導核分裂（熱中性子起源）の二次階乗モーメントで、 生じる中性子ペア数の期待値の二倍
α_1	自発及び誘導核分裂（高速中性子起源）の即発中性子減衰定数
α_2	誘導核分裂（熱中性子起源）の即発中性子減衰定数
τ_1	自発又は誘導核分裂（高速中性子起源）の平均的な時間間隔
τ_2	誘導核分裂（熱中性子起源）の平均的な時間間隔
S	中性子の計数率 = 相関の有無を考えない単味（singlets）計数率 ≡ Singles
D	中性子ペア（２個 1 組）の計数率 = 二重相関（doublets）計数率 ≡ Doubles
T	中性子ペア（３個 1 組）の計数率 = 三重相関（triplets）計数率 ≡ Triples
D_1	自発及び誘導核分裂（高速中性子による）による中性子ペアの計数率
D_2	誘導核分裂（熱中性子による）による中性子ペアの計数率
f_d	二重相関計数時のゲートの有効割合（doubles gate fraction）
M_T	全増倍率
M_L	漏れ増倍率

1. Preface

Neutron coincidence/multiplicity assay systems have been used for accounting nuclear materials and for safeguards to determine plutonium weight in a small and unshaped material (ex. in waste and in glovebox holdup). Several assay systems for such a material composed of U-Pu (U/Pu=1) mixed dioxide powder have been installed, jointly used with inspectorate and improved[1,2,3] over two decades at the Plutonium Conversion Development Facility (PCDF) adjacent to the Tokai Reprocessing Plant in the Japan Atomic Energy Agency (JAEA). It is known that Passive Nondestructive Assay Manual called PANDA[4], its 2007 Addendum[5] and Neutron Fluctuations[6] are comprehensive wisdom. However, we need more essentials to cover the experiences in the field (ex. accidental pairs) and to develop devices may be required in the future. So, essentials were re-examined, confirmed and expanded in the previous report[7]. This report is the addendum.

2. Multiplicity distribution up to septuplet

2.1 Introduction

Equations for neutron leakage multiplicity correlations up to triplets are frequently referred in papers for neutron coincidence/multiplicity assay system. There are two correlations, i.e. true correlation resulting from a fission cascade and accidental correlation resulting from different fission cascades. For a doublet, the former was called 'coupled pair' and the latter 'accidental pair' by Hoffmann[8] in 1949. These pairs and Rossi-alpha distribution are formulated in the Appendix a). The frequently referred equations are associated with true correlations only, and derivation was given in several ways, ex. by Hage and Cifarelli[9] in 1985. Another viewpoint associated with Y-value was given by Croft[10] in 2012. However, it is difficult to apply these methods to higher correlations greater than quadruplets. There is another way using probability generating function (PGF) discussed by Böhnel[11] in 1985, which might be applied to higher correlations to make a strategic move in the future. In this section, derivation of higher correlations by PGF is explained then equations up to septuplet are derived assisted in doing mathematics by a computer, then characteristics of higher correlations are investigated.

It is sure that counting technique for high-order correlations has not been established because of large uncertainty of counting statistics due to thermalization and small leakage multiplication of a sample. However, it could be possible if fast neutron detecting/counting technique could be established without moderator and a sample could have larger leakage multiplication within criticality control.

2.2 Probability generating function for a neutron leakage chain

Figure 1 shows a neutron leakage chain resulting from a fission cascade where a spontaneous fission or a (α,n) reaction is a starting event. In this figure, expected number of spontaneous fission neutrons $\overline{\nu_{s1}}$ and expected number of induced fission neutrons $\overline{\nu_{i1}}$ are used where subscript '1' indicates a singlet that means neutron is counted individually, whereas a doublet ($\overline{\nu_{s2}}$, $\overline{\nu_{i2}}$) means neutron pair come from a spontaneous or induced fission is counted. Such usages of symbols are normal in conventional neutron coincidence/multiplicity counting, together with a ratio of the number of neutrons yields from (α,n) reaction to the number from spontaneous fission α_r, leakage probability l and induced fission probability p:

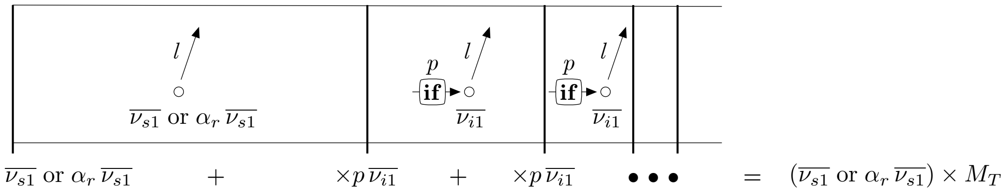


Figure 1 Neutron leakage chain resulting from a fission cascade

Neutron leakage multiplication M_L and total multiplication M_T are defined as[a]:

$$M_L = l\,M_T = l\,[1+p\,\overline{\nu_{i1}}+(p\,\overline{\nu_{i1}})^2\ldots] = \frac{l}{1-p\,\overline{\nu_{i1}}} = \frac{1-p-p_c}{1-p\,\overline{\nu_{i1}}} \approx \frac{1-p}{1-p\,\overline{\nu_{i1}}} \tag{1}$$

where p_c is neutron capture probability, ex. by (n,γ) reaction, however it is not so large (several percent of p) thus $l \approx 1-p$ is normally supposed. Now, let us suppose the PGF of leakage neutrons in Figure 1 when the chain starts from one neutron (not one fission/reaction). From the equations in the Appendix b), the PGF for a neutron is written as:

$$G_h\,(z) = (1-p)\sum_{n=1}^{1} z^n\,1 + \overbrace{p\sum_{n=0}^{\infty}[\,G_h\,(z)\,]^n\;P_{\nu_i}\,(\nu_i = n)}^{G_i\,(\,G_h\,(z)\,)} \tag{2}$$

where P_{ν_i} is probability of number of neutrons for induced fission and $\nu_i = \nu_{i1}$. The first term on the right side of the equation means direct leakage, thus $X = \{1\}$ and $P_X = 1$. The second term means leakages through induced fission as a composite of leakages in each ν_i, which results in the nest of the PGF for a neutron. The subscript 'h' comes from the one in Böhnel's equation (9) where the PGF is explained as 'the PGF for the number of neutrons of this first and all successive generations that leave a system'.

a. $\dfrac{M_L-1}{\overline{\nu_{i1}}-1} = \dfrac{p}{1-p\,\overline{\nu_{i1}}} = p\,M_T$, $\dfrac{M_L-1}{M_L\,\overline{\nu_{i1}}-1} = p$, $\dfrac{M_L\,\overline{\nu_{i1}}-1}{\overline{\nu_{i1}}-1} = M_T$ are useful transformations.

The equation (2) is simplified to:

$$G_h(z) = (1-p)\,z + p\,G_i(G_h(z)) \tag{3}$$

The k^{th} derivatives are solved algebraically assisted by a computer[b]:

$$
\begin{aligned}
G_h^{'} &= \frac{1-p}{1-p\,G_i^{'}} \\
G_h^{''} &= \frac{(1-p)^2\,p}{(1-p\,G_i^{'})^3}\,G_i^{''} \\
G_h^{(3)} &= \frac{(1-p)^3\,p}{(1-p\,G_i^{'})^4}\left[G_i^{(3)} + \frac{p}{1-p\,G_i^{'}}\,3\,(G_i^{''})^2\right] \\
G_h^{(4)} &= \frac{(1-p)^4\,p}{(1-p\,G_i^{'})^5}\left[G_i^{(4)} + \frac{p}{1-p\,G_i^{'}}\,10\,G_i^{(3)}\,G_i^{''} + \frac{p^2}{(1-p\,G_i^{'})^2}\,15\,(G_i^{''})^3\right] \\
G_h^{(5)} &= \frac{(1-p)^5\,p}{(1-p\,G_i^{'})^6}\left\{G_i^{(5)} + \frac{p}{1-p\,G_i^{'}}\left[15\,G_i^{(4)}G_i^{''} + 10\,(G_i^{(3)})^2\right]\right. \\
&\qquad \left. + \frac{p^2}{(1-p\,G_i^{'})^2}\,105\,G_i^{(3)}(G_i^{''})^2 + \frac{p^3}{(1-p\,G_i^{'})^3}\,105\,(G_i^{''})^4\right\} \\
G_h^{(6)} &= \frac{(1-p)^6\,p}{(1-p\,G_i^{'})^7}\left\{G_i^{(6)} + \frac{p}{1-p\,G_i^{'}}\left[21\,G_i^{(5)}G_i^{''} + 35\,G_i^{(4)}G_i^{(3)}\right]\right. \\
&\qquad + \frac{p^2}{(1-p\,G_i^{'})^2}\left[210\,G_i^{(4)}(G_i^{''})^2 + 280\,(G_i^{(3)})^2G_i^{''}\right] \\
&\qquad \left. + \frac{p^3}{(1-p\,G_i^{'})^3}\,1260\,G_i^{(3)}(G_i^{''})^3 + \frac{p^4}{(1-p\,G_i^{'})^4}\,945\,(G_i^{''})^5\right\} \\
G_h^{(7)} &= \frac{(1-p)^7\,p}{(1-p\,G_i^{'})^8}\left\{G_i^{(7)} + \frac{p}{1-p\,G_i^{'}}\left[28\,G_i^{(6)}G_i^{''} + 56\,G_i^{(5)}G_i^{(3)} + 35\,(G_i^{(4)})^2\right]\right. \\
&\qquad + \frac{p^2}{(1-p\,G_i^{'})^2}\left[378\,G_i^{(5)}(G_i^{''})^2 + 1260\,G_i^{(4)}G_i^{(3)}G_i^{''} + 280\,(G_i^{(3)})^3\right] \\
&\qquad + \frac{p^3}{(1-p\,G_i^{'})^3}\left[3150\,G_i^{(4)}(G_i^{''})^3 + 6300\,(G_i^{(3)})^2(G_i^{''})^2\right] \\
&\qquad \left. + \frac{p^4}{(1-p\,G_i^{'})^4}\,17325\,G_i^{(3)}(G_i^{''})^4 + \frac{p^5}{(1-p\,G_i^{'})^5}\,10395\,(G_i^{''})^6\right\}
\end{aligned} \tag{4}
$$

b. *Mathematica*® ver.5.0 by Wolfram Research.
In ref.11, Böhnel had shown up to 3rd derivatives in eq.(21), (22) and (23), where $h(u)$, $f(u)$, $H(u)$, $M_{(p)i} \to M_{(p)k}$ and $f_s(u)$ corresponds respectively to $G_h(z)$, $G_i(z)$, $G_H(z)$, $\overline{\nu_{ik}}$ and $G_s(z)$ are used.

The following relations are applied later to the above equations:

$$G_i^{(k)}|_{z=1} = \overline{\nu_i(\nu_i - 1)(\nu_i - 2)\cdots(\nu_i - k + 1)} = \overline{\nu_{ik}} \tag{5}$$

$$\frac{1-p}{1-p\,G_i^{'}} = \frac{1-p}{1-p\,\overline{\nu_{i1}}} = M_L \tag{6}$$

$$\frac{p}{1-p\,G_i^{'}} = \frac{p}{1-p\,\overline{\nu_{i1}}} = \frac{M_L - 1}{\overline{\nu_{i1}} - 1} \overset{\text{def}}{=} M_{L\nu} \tag{7}$$

2.3 Probability generating function considering the source of a neutron

The starting event of Figure 1 is a spontaneous fission or a (α,n) reaction. The PGF for a neutron is rewritten using $G_s\,(z)$ for spontaneous fission where P_{ν_s} is used instead of P_{ν_i} as:

$$G_H\,(z) = \frac{\alpha_r\,\overline{\nu_{s1}}}{1+\alpha_r\,\overline{\nu_{s1}}}\,G_h\,(z) + \frac{1}{1+\alpha_r\,\overline{\nu_{s1}}}\,G_s\,(G_h\,(z)) \tag{8}$$

where the subscript '*H*' comes from the one in Böhnel's equation (13). The coefficients of the first and the second terms on the right side of the equation, i.e. $\alpha_r\,\overline{\nu_{s1}}\,/(\,1+\alpha_r\,\overline{\nu_{s1}}\,)$ and $1/(\,1+\alpha_r\,\overline{\nu_{s1}}\,)$ means probabilities of a neutron from a (α,n) reaction or a neutron from a spontaneous fission, because (α,n) reaction occurs $\alpha_r\,\overline{\nu_{s1}}$ times more frequently than spontaneous fission. Same as the equation (5), the next transformation is applied later to the above equation:

$$G_s^{(k)}|_{z=1} = \overline{\nu_s(\nu_s - 1)(\nu_s - 2)\cdots(\nu_s - k + 1)} = \overline{\nu_{sk}} \tag{9}$$

The k^{th} derivatives are too long to be shown here, however it is possible to enclose the right side of each derivative by the term $(1-p)^k(1+\alpha_r\,\overline{\nu_{s1}})^{-1}(1-p\,\overline{\nu_{i1}})^{1-2k}$ after the equations (4), (5) and (9) are applied. Finally, the equations (6) and (7) are applied to eliminate $1-p$ and p, followed by replacing $(M_L-1)\,/(\overline{\nu_{i1}}-1)$ by $M_{L\nu}$ to express equations in short form.

To obtain probability mass distribution of multiple leakage neutrons (probability distribution of neutron pairs composed of m neutrons) derived from PGF for a spontaneous fission event, the following equation in the Appendix b) is applied:

$$P_m\,(m{=}k) = (1+\alpha_r\overline{\nu_{s1}})\;\frac{G_H^{(k)}|_{z=0}}{k!} \tag{10}$$

Example of the part of solving process (including 2.2), especially multiple derivation, by a computer are shown in the Appendix c).

2.4 Probability mass distribution of multiple leakage neutrons

Results of the equation (10) up to $k=3$ is enclosed by $M_L^m/m!$ same as the conventional equations. Terms in the enclosure are collected by $\overline{\nu_{s1}}$, $\overline{\nu_{s2}}$, $\overline{\nu_{s3}}$ $\cdots$ followed by collected by $M_{L\nu}$, $M_{L\nu}^2$, $M_{L\nu}^3$ $\cdots$. Obtained probability mass distribution of multiple leakage neutrons are shown below. There are another expressions in the Appendix d):

$$P_m\,(m{=}1) = M_L\,(1+\alpha_r)\,\overline{\nu_{s1}} \tag{11}$$

$$P_m\,(m{=}2) = \frac{M_L^2}{2}\,\left[\overline{\nu_{s2}} + \overline{\nu_{i2}}\,M_{L\nu}\,(1+\alpha_r)\,\overline{\nu_{s1}}\right] \tag{12}$$

$$P_m\,(m{=}3) = \frac{M_L^3}{6}\,\left[\overline{\nu_{s3}} + 3\,\overline{\nu_{i2}}\,M_{L\nu}\,\overline{\nu_{s2}} + \left(\overline{\nu_{i3}}\,M_{L\nu} + 3\,\overline{\nu_{i2}}^2\,M_{L\nu}^2\right)\,(1+\alpha_r)\,\overline{\nu_{s1}}\right] \tag{13}$$

The conventional equations frequently referred as singles count rate S, doubles count rate D and triples count rate T are:

$$S = F_p\,m_{\mathit{eff}}\,\epsilon_n\,(1+\alpha_r)\,\overline{\nu_{s1}}\,M_L \tag{14}$$

$$D = \frac{F_p\,m_{\mathit{eff}}\,\epsilon_n^2\,f_d}{2}\left[\overline{\nu_{s2}} + \overline{\nu_{i2}}\,(1+\alpha_r)\,\overline{\nu_{s1}}\,\frac{M_L-1}{\overline{\nu_{i1}}-1}\right]M_L^2 \tag{15}$$

$$T = \frac{F_p\,m_{\mathit{eff}}\,\epsilon_n^3\,f_t}{6}\left\{\overline{\nu_{s3}} + \frac{M_L-1}{\overline{\nu_{i1}}-1}\,\left[(1+\alpha_r)\,\overline{\nu_{s1}}\,\overline{\nu_{i3}} + 3\,\overline{\nu_{s2}}\,\overline{\nu_{i2}}\right] + 3\,(1+\alpha_r)\,\overline{\nu_{s1}}\left(\frac{M_L-1}{\overline{\nu_{i1}}-1}\right)^2\overline{\nu_{i2}}^2\right\}M_L^3 \tag{16}$$

$$\text{where}\quad \overline{\nu_{s/i\,m}} \overset{\text{def}}{=} m!\sum_{\nu_{s/i}=m}^{\max}\binom{\nu_{s/i}}{m}P_\nu = \overline{\nu_{s/i}\,(\nu_{s/i}-1)\,(\nu_{s/i}-2)\cdots(\nu_{s/i}-m+1)} \tag{17}$$

Real count rates are equal to the product of probability mass distribution of multiple leakage neutrons and $F_p\,m_{\mathit{eff}}\,\epsilon_n^m\,(1/f_d/f_t\cdots/f_7)$ where F_p is the number of spontaneous fissions per unit mass and time, m_{eff} is effective mass of spontaneous fission nuclei (^{238}Pu, ^{240}Pu and ^{242}Pu) , ϵ_n is counting efficiency (counts per neutron), f_d and f_t are characteristic values depend on counter setting called gate fraction. Our results up to $k=3$ is absolutely consistent with conventional equations. Results up to $k=7$ are given as:

$$\begin{aligned}P_m\,(m{=}4) = \frac{M_L^4}{24}\,\Big[&\overline{\nu_{s4}} + 6\,\overline{\nu_{i2}}\,M_{L\nu}\,\overline{\nu_{s3}} \\ &+ \left(4\,\overline{\nu_{i3}}\,M_{L\nu} + 15\,\overline{\nu_{i2}}^2\,M_{L\nu}^2\right)\,\overline{\nu_{s2}} \\ &+ \left(\overline{\nu_{i4}}\,M_{L\nu} + 10\,\overline{\nu_{i3}}\,\overline{\nu_{i2}}\,M_{L\nu}^2 + 15\,\overline{\nu_{i2}}^3\,M_{L\nu}^3\right)\,(1+\alpha_r)\,\overline{\nu_{s1}}\Big]\end{aligned} \tag{18}$$

$$
\begin{aligned}
P_m\,(m=5) = \frac{M_L^5}{120}\ \{ & \overline{\nu_{s5}} + 10\,\overline{\nu_{i2}}\,M_{L\nu}\,\overline{\nu_{s4}} \\
& + \left(10\,\overline{\nu_{i3}}\,M_{L\nu}\ + 45\,\overline{\nu_{i2}}^2\,M_{L\nu}^2 \right)\ \overline{\nu_{s3}} \\
& + \left(5\,\overline{\nu_{i4}}\,M_{L\nu}\ + 60\,\overline{\nu_{i3}}\,\overline{\nu_{i2}}\,M_{L\nu}^2\ + 105\,\overline{\nu_{i2}}^3\,M_{L\nu}^3 \right)\ \overline{\nu_{s2}} \\
& + \Big[\overline{\nu_{i5}}\,M_{L\nu} + \left(15\,\overline{\nu_{i4}}\,\overline{\nu_{i2}} + 10\,\overline{\nu_{i3}}^2 \right)\ M_{L\nu}^2 \\
& \qquad + 105\ \overline{\nu_{i3}}\,\overline{\nu_{i2}}^2\,M_{L\nu}^3 + 105\,\overline{\nu_{i2}}^4\,M_{L\nu}^4 \Big]\ (1+\alpha_r)\,\overline{\nu_{s1}}\ \}
\end{aligned}
\tag{19}
$$

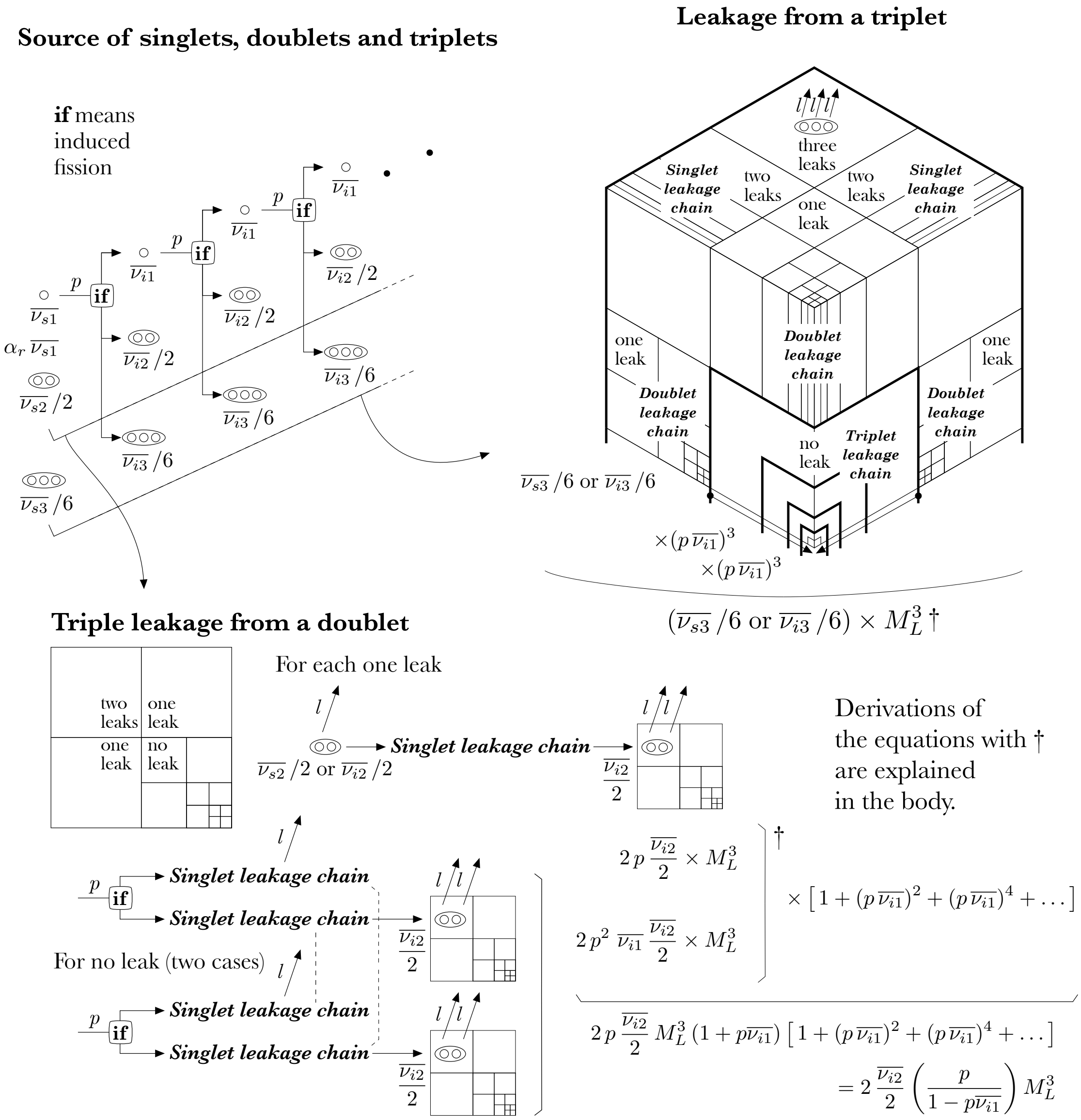
$$
\begin{aligned}
P_m\,(m=6) = \frac{M_L^6}{720}\ \{ & \overline{\nu_{s6}} + 15\,\overline{\nu_{i2}}\,M_{L\nu}\,\overline{\nu_{s5}} \\
& + \left(20\,\overline{\nu_{i3}}\,M_{L\nu}\ + 105\,\overline{\nu_{i2}}^2\,M_{L\nu}^2 \right)\ \overline{\nu_{s4}} \\
& + \left(15\,\overline{\nu_{i4}}\,M_{L\nu}\ + 210\,\overline{\nu_{i3}}\,\overline{\nu_{i2}}\,M_{L\nu}^2\ + 420\,\overline{\nu_{i2}}^3\,M_{L\nu}^3 \right)\ \overline{\nu_{s3}} \\
& + \Big[6\,\overline{\nu_{i5}}\,M_{L\nu} + \left(105\,\overline{\nu_{i4}}\,\overline{\nu_{i2}} + 70\,\overline{\nu_{i3}}^2 \right)\ M_{L\nu}^2 \\
& \qquad + 840\ \overline{\nu_{i3}}\,\overline{\nu_{i2}}^2\,M_{L\nu}^3 + 945\,\overline{\nu_{i2}}^4\,M_{L\nu}^4 \Big]\ \overline{\nu_{s2}} \\
& + \Big[\overline{\nu_{i6}}\,M_{L\nu} + (\,21\,\overline{\nu_{i5}}\,\overline{\nu_{i2}} + 35\,\overline{\nu_{i4}}\,\overline{\nu_{i3}}\,)\ M_{L\nu}^2 \\
& \qquad + \left(210\ \overline{\nu_{i4}}\,\overline{\nu_{i2}}^2 + 280\ \overline{\nu_{i3}}^2\,\overline{\nu_{i2}} \right)\ M_{L\nu}^3 \\
& \qquad\quad + 1260\ \overline{\nu_{i3}}\,\overline{\nu_{i2}}^3\,M_{L\nu}^4 + 945\,\overline{\nu_{i2}}^5\,M_{L\nu}^5 \Big]\ (1+\alpha_r)\,\overline{\nu_{s1}}\ \}
\end{aligned}
\tag{20}
$$

$$
\begin{aligned}
P_m\,(m=7) = \frac{M_L^7}{5040}\ \{ & \overline{\nu_{s7}} + 21\,\overline{\nu_{i2}}\,M_{L\nu}\,\overline{\nu_{s6}} \\
& + \left(35\,\overline{\nu_{i3}}\,M_{L\nu}\ + 210\,\overline{\nu_{i2}}^2\,M_{L\nu}^2 \right)\ \overline{\nu_{s5}} \\
& + \left(35\,\overline{\nu_{i4}}\,M_{L\nu}\ + 560\,\overline{\nu_{i3}}\,\overline{\nu_{i2}}\,M_{L\nu}^2\ + 1260\,\overline{\nu_{i2}}^3\,M_{L\nu}^3 \right)\ \overline{\nu_{s4}} \\
& + \Big[21\,\overline{\nu_{i5}}\,M_{L\nu} + \left(420\,\overline{\nu_{i4}}\,\overline{\nu_{i2}} + 280\,\overline{\nu_{i3}}^2 \right)\ M_{L\nu}^2 \\
& \qquad + 3780\ \overline{\nu_{i3}}\,\overline{\nu_{i2}}^2\,M_{L\nu}^3 + 4725\,\overline{\nu_{i2}}^4\,M_{L\nu}^4 \Big]\ \overline{\nu_{s3}} \\
& + \Big[7\,\overline{\nu_{i6}}\,M_{L\nu} + (\,168\,\overline{\nu_{i5}}\,\overline{\nu_{i2}} + 280\,\overline{\nu_{i4}}\,\overline{\nu_{i3}}\,)\ M_{L\nu}^2 \\
& \qquad + \left(1890\ \overline{\nu_{i4}}\,\overline{\nu_{i2}}^2 + 2520\ \overline{\nu_{i3}}^2\,\overline{\nu_{i2}} \right)\ M_{L\nu}^3 \\
& \qquad\quad + 12600\ \overline{\nu_{i3}}\,\overline{\nu_{i2}}^3\,M_{L\nu}^4 + 10395\,\overline{\nu_{i2}}^5\,M_{L\nu}^5 \Big]\ \overline{\nu_{s2}} \\
& + \Big[\overline{\nu_{i7}}\,M_{L\nu} + \left(28\,\overline{\nu_{i6}}\,\overline{\nu_{i2}} + 56\,\overline{\nu_{i5}}\,\overline{\nu_{i3}} + 35\,\overline{\nu_{i4}}^2 \right)\ M_{L\nu}^2 \\
& \qquad + \left(378\ \overline{\nu_{i5}}\,\overline{\nu_{i2}}^2 + 1260\ \overline{\nu_{i4}}\,\overline{\nu_{i3}}\,\overline{\nu_{i2}} + 280\ \overline{\nu_{i3}}^3 \right)\ M_{L\nu}^3 \\
& \qquad + \left(3150\ \overline{\nu_{i4}}\,\overline{\nu_{i2}}^3 + 6300\ \overline{\nu_{i3}}^2\,\overline{\nu_{i2}}^2 \right)\ M_{L\nu}^4 \\
& \qquad\quad + 17325\ \overline{\nu_{i3}}\,\overline{\nu_{i2}}^4\,M_{L\nu}^5 + 10395\,\overline{\nu_{i2}}^6\,M_{L\nu}^6 \Big]\ (1+\alpha_r)\,\overline{\nu_{s1}}\ \}
\end{aligned}
\tag{21}
$$

Singles, Doubles, Triples, Quadruples, Quintuples, Sextuples, Septuples are given as:

$$
\underbrace{S/D/T/Q_r/Q_t/S_x/S_p}_{m=1,2,3,4,5,6,7} = F_p\,m_{eff}\,\epsilon_n^m\,f_m \times P_m\,(m = 1/2/3/4/5/6/7)
\tag{22}
$$

For reference, probability branching scheme of triplets is shown in Figure 2. It is complicated and difficult to extend the scheme to higher-order correlations.



For triplet :

$$\overbrace{\left(\begin{array}{l}\overline{\nu_{s3}}/6\\ +\,(\overline{\nu_{i3}}/6)\;p\,\overline{\nu_{s1}}\,\left[1+p\overline{\nu_{i1}}+(p\,\overline{\nu_{i1}})^2\ldots\right]\\ +\,(\overline{\nu_{i3}}/6)\;p\,\alpha_r\,\overline{\nu_{s1}}\,\left[1+p\overline{\nu_{i1}}+(p\,\overline{\nu_{i1}})^2\ldots\right]\end{array}\right)}^{\text{Evaluation of the chain of sources}}M_L^3$$

$$+\;\overbrace{\left(\begin{array}{l}(\overline{\nu_{s2}}/2)\\ +\,(\overline{\nu_{i2}}/2)\;p\,\overline{\nu_{s1}}\,\left[1+p\overline{\nu_{i1}}+(p\,\overline{\nu_{i1}})^2\ldots\right]\\ +\,(\overline{\nu_{i2}}/2)\;p\,\alpha_r\,\overline{\nu_{s1}}\,\left[1+p\overline{\nu_{i1}}+(p\,\overline{\nu_{i1}})^2\ldots\right]\end{array}\right)}^{\text{Evaluation of the chain of sources}}\;2\,\frac{\overline{\nu_{i2}}}{2}\,\frac{p}{1-p\overline{\nu_{i1}}}M_L^3$$

$$=\;\frac{1}{6}\left\{\overline{\nu_{s3}}+\frac{p}{1-p\overline{\nu_{i1}}}\left[(1+\alpha_r)\,\overline{\nu_{s1}}\,\overline{\nu_{i3}}+3\,\overline{\nu_{s2}}\,\overline{\nu_{i2}}\right]+\,3\,(1+\alpha_r)\,\overline{\nu_{s1}}\,\left(\frac{p}{1-p\overline{\nu_{i1}}}\right)^2\overline{\nu_{i2}}^{\,2}\right\}M_L^3$$

Figure 2 Probability branching scheme of triplets (reprinted from JAEA-Research 2015-009)

2.5 Dependence of high-order correlations on leakage multiplication

The parameters F_p, ϵ_n, f_d, $f_t \cdots f_7$ and α_r were supposed to be 474 $g^{-1}s^{-1}$, 0.4, 0.67, $(0.67)^2$ … $(0.67)^6$ and 0.7, respectively, with reference to the measurement conditions to count S, D of U-Pu mixed dioxide powder at PCDF. For $\overline{\nu_{s/i\,m}}$, ^{240}Pu and ^{239}Pu (2 MeV neutron) were selected because of their large contribution on neutron yield. The $\overline{\nu_{s/i\,m}}$ were calculated using P_ν consolidated/proposed by Zucker and Holden[12,13,14] in 1984-1986. They are shown in Table 1 and 2:

Table 1 P_ν of ^{240}Pu and ^{239}Pu (2 MeV neutron)

	ν=0	1	2	3	4	5	6	7
^{240}Pu	0.0632	0.2320	0.3333	0.2528	0.0986	0.0180	0.0020	0.00006[†]
^{239}Pu (2 MeV)	0.0063	0.0612	0.2266	0.3261	0.2588	0.0956	0.0225	0.0026

†. Estimated by Gaussian fit of the distribution[c] based on Terrell[15] in 1957.

Table 2 $\overline{\nu_{sm}}$ of ^{240}Pu and $\overline{\nu_{im}}$ of ^{239}Pu (2 MeV neutron)

	m=1	2	3	4	5	6	7
$\overline{\nu_{sm}}$	2.1537	3.7889	5.2155	5.2965	3.7510	1.7423	0.3024
$\overline{\nu_{im}}$	3.1591	8.2116	17.1498	27.9672	34.2240	29.3040	13.1040

Higher-order factorial moments become larger than $\overline{\eta_{im}} = \overline{\nu_{im}}/(m\,!)$ which is the number of multiple neutron pairs for a fission shown in Table 3. See Appendix d) for detail.

Table 3 $\overline{\eta_{sm}}$ of ^{240}Pu and $\overline{\eta_{im}}$ of ^{239}Pu (2 MeV neutron)

	m=1	2	3	4	5	6	7
$\overline{\eta_{sm}}$	2.1537	1.8944	0.8692	0.2207	0.0313	0.0024	0.0001
$\overline{\eta_{im}}$	3.1591	4.1058	2.8583	1.1653	0.2852	0.0407	0.0026

Calculated $S/D/T/Q_r/Q_t/S_x/S_p$ for one gram spontaneous fission source are shown in Table 4 and Figure 3. Underline indicates the minimum of each M_L. It is interesting that the minimum moves to lower-order when M_L > 1.3 (ϵ_n=0.4, f_d=0.67).

---

c. $\mathrm{erf}\,x = \dfrac{2}{\sqrt{\pi}}\displaystyle\int_0^x \exp\left(-z^2\right) dz = \sqrt{\dfrac{2}{\pi}}\int_0^{\sqrt{2}x} \exp\left(-\dfrac{t^2}{2}\right) dt$ is applied to

$P_\nu = \displaystyle\sum_{m=0}^{\nu} P_m - \sum_{m=0}^{\nu-1} P_m = \dfrac{1}{\sqrt{2\pi}}\int_{(\nu-\overline{\nu}-0.5)/\sigma}^{(\nu-\overline{\nu}+0.5)/\sigma} \exp\left(-\dfrac{t^2}{2}\right) dt$ then we get

$P_\nu = \dfrac{1}{2}\left[\mathrm{erf}\left(\dfrac{\nu-\overline{\nu}+0.5}{\sigma\sqrt{2}}\right) - \mathrm{erf}\left(\dfrac{\nu-\overline{\nu}-0.5}{\sigma\sqrt{2}}\right)\right]$ where $\overline{\nu}$ and σ are mean and width of Gaussian distribution fit.

The width is given as 1.08 ± 0.01 by Terrell (ref.15) for all nuclei except for ^{252}Cf, however it is better to use the mean and the width in ref.7 pp.153-154 to fit precisely. The mean and the width of ^{240}Pu are 2.092 and 1.150, respectively. This mean is not equal to $\overline{\nu_{s1}}$ (2.1537 for ^{240}Pu).

Table 4 $S/D/T/Q_r/Q_t/S_x/S_p$ for one gram spontaneous fission source
($\epsilon_n = 0.4,\ \alpha_r = 0.7,\ f_d = 0.67$)

	S (m=1)	D (m=2)	T (m=3)	Q_r (m=4)	Q_t (m=5)	S_x (m=6)	S_p (m=7)
M_L=1.0	694	96.3	11.8	0.806	0.0306	0.00064	<u>0.000004</u>
M_L=1.1	764	159	42.4	12.5	4.23	1.54	<u>0.603</u>
M_L=1.2	833	241	102	51.7	29.5	18.1	<u>11.7</u>
M_L=1.3	902	342	205	150	124	109	<u>101</u>
M_L=1.4	972	466	371	<u>363</u>	399	470	579
M_L=1.5	1,041	<u>615</u>	621	776	1,087	1,632	2,563
M_L=1.6	1,111	<u>790</u>	984	1,519	2,628	4,871	9,450

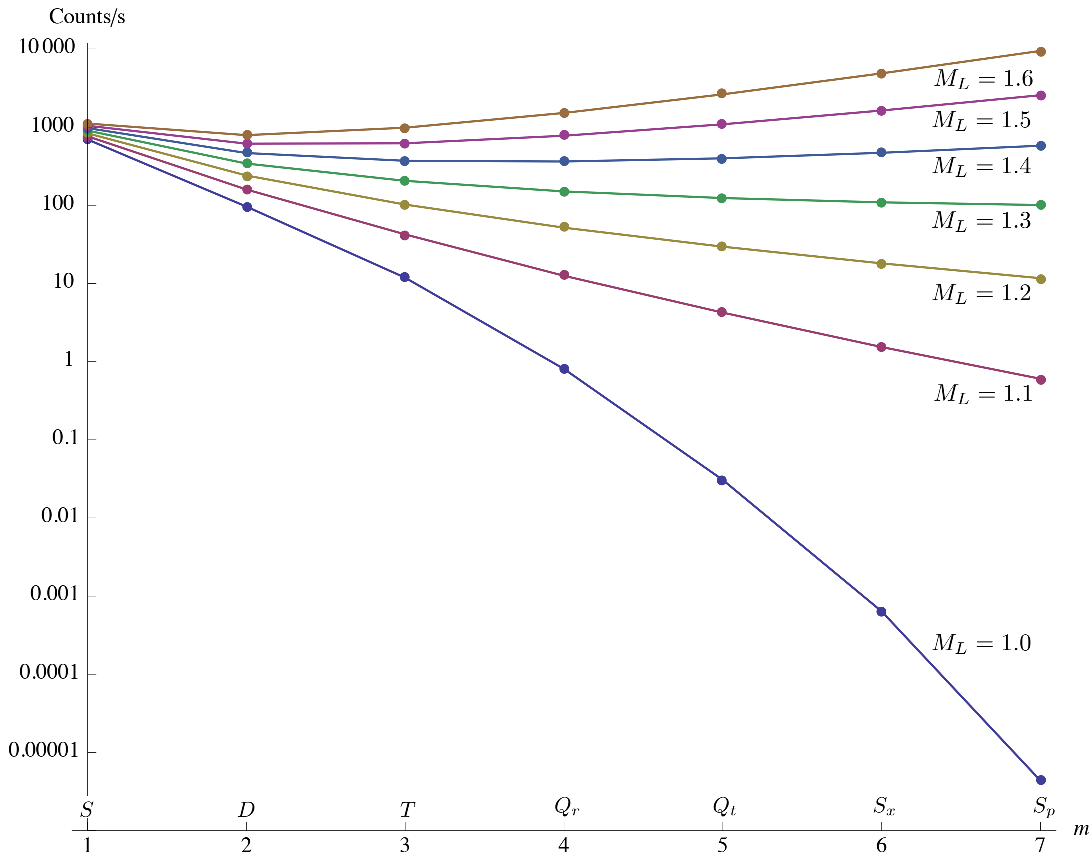


Figure 3 $S/D/T/Q_r/Q_t/S_x/S_p$ for one gram spontaneous fission source
($\epsilon_n = 0.4,\ \alpha_r = 0.7,\ f_d = 0.67$)

Count rate increases at an accelerated pace with increase of M_L, which becomes more remarkable when m is larger. Ratio of the maximum to the minimum in the same M_L reduces to < 10 when M_L > 1.3 ($\epsilon_n = 0.4,\ f_d = 0.67$). In addition, chemical composition, isotopic composition, ratio of the number of induced fission nuclei to the number of spontaneous nuclei, energy of a neutron, mean free path, density, shape and size of a sample and so on does not appear in the calculation of $S/D/T/Q_r/Q_t/S_x/S_p$, because they affect variable p in the equation (6) then affect M_L.

2.6 Crossing of high-order correlations at high-leakage multiplication

Figure 4a is another view of Figure 3. Figure 4b, 4c and 4d are examples of parameter study. It is clear that high-order correlations increase rapidly and cross and leave lower-order correlations behind. This tendency is enforced by ϵ_n and f_d ($f_t \cdots f_7$ are supposed to be $f_d^{\,m}$), which results in moving the cross-point to lower M_L. Instead of log scale, linear scale was adopted to show the cross-points clearly.

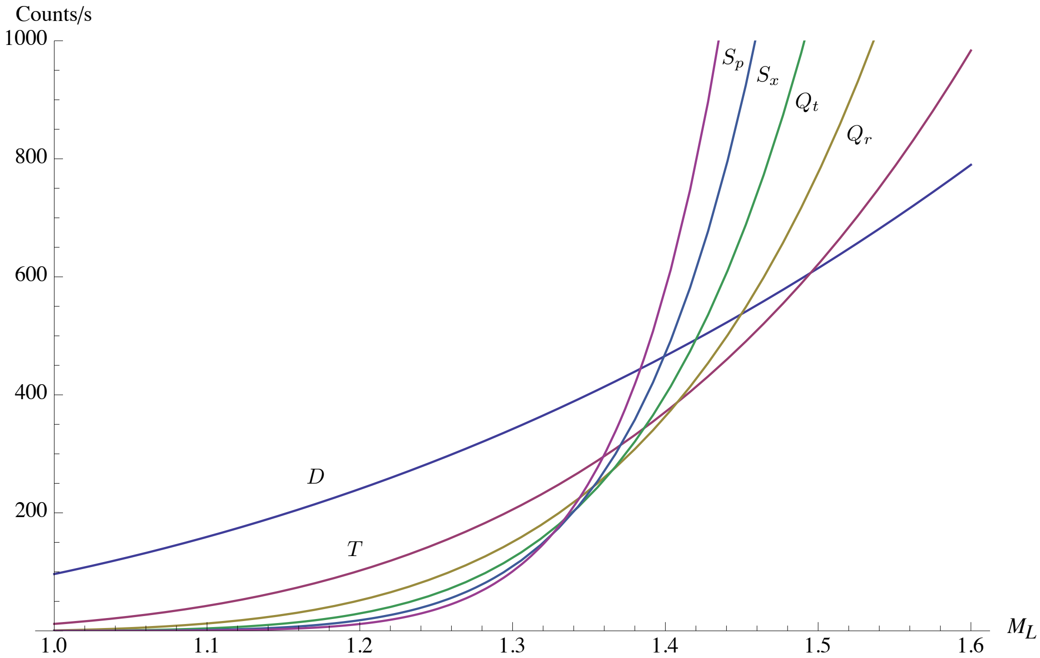


Figure 4a Crossing of $D/T/Q_r/Q_t/S_x/S_p$ (ϵ_n=0.4, α_r=0.7, f_d=0.67)

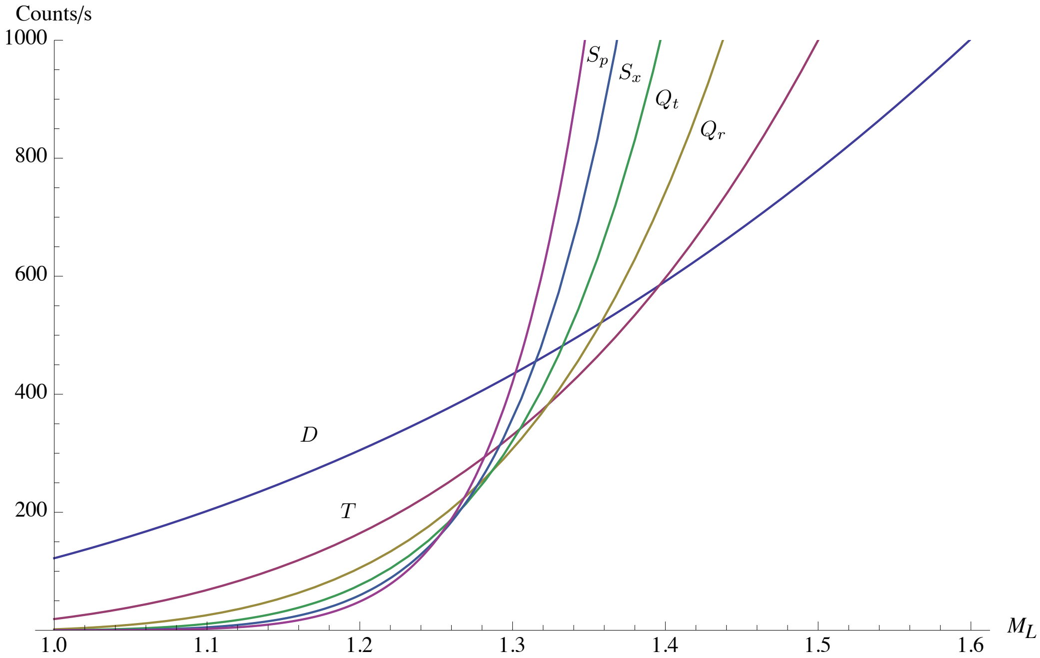


Figure 4b Crossing of $D/T/Q_r/Q_t/S_x/S_p$ (ϵ_n=0.4, α_r=0.7, f_d=0.85)

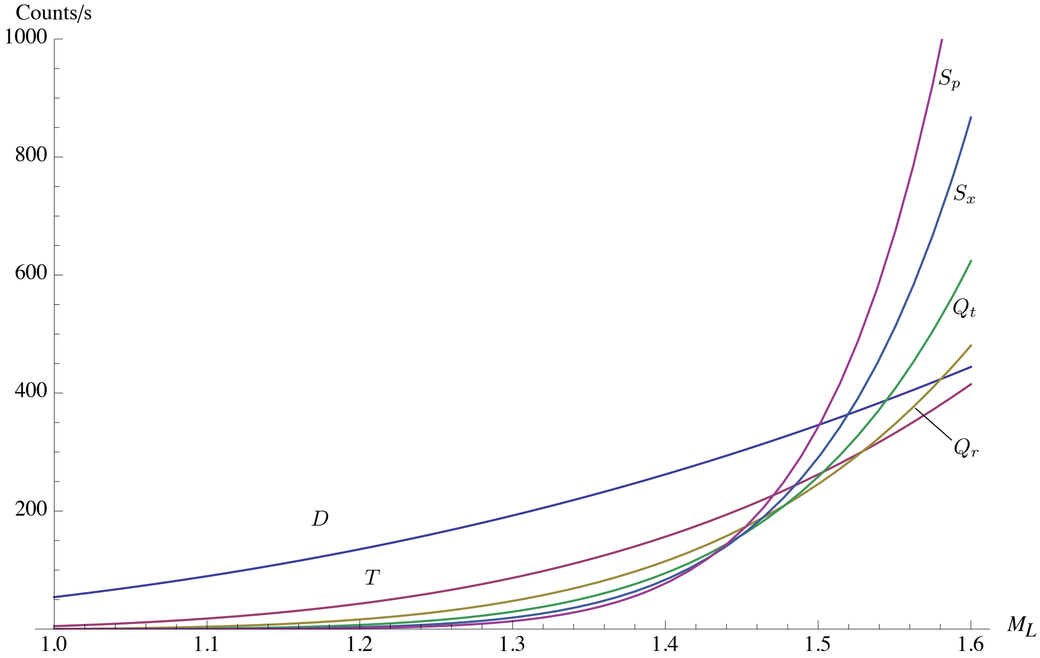


Figure 4c Crossing of $D/T/Q_r/Q_t/S_x/S_p$ ($\epsilon_n = 0.3$, $\alpha_r = 0.7$, $f_d = 0.67$)

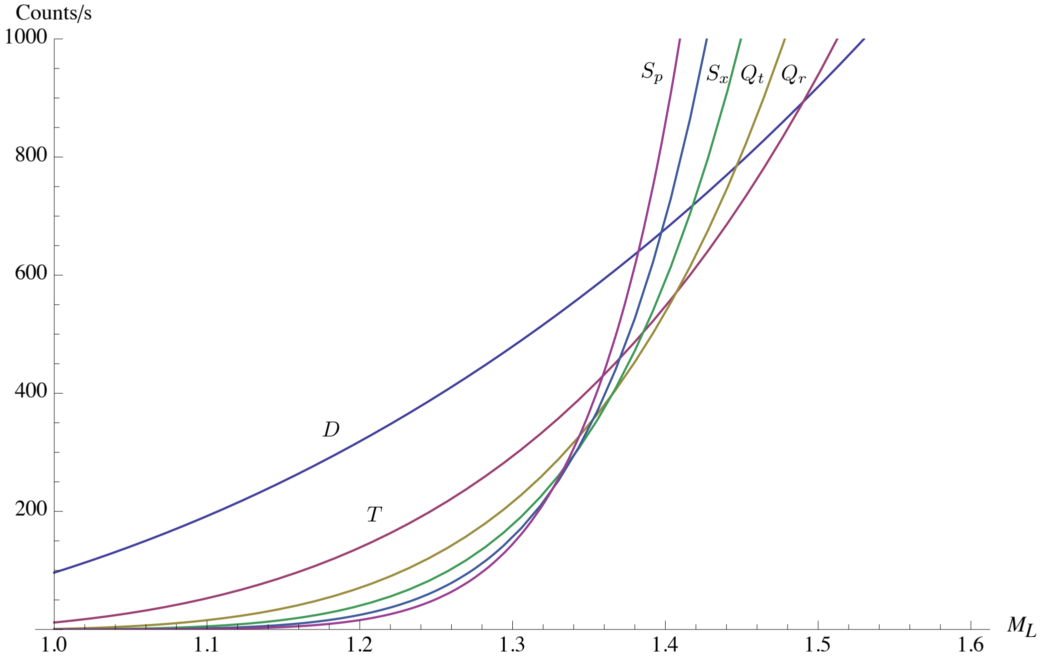


Figure 4d Crossing of $D/T/Q_r/Q_t/S_x/S_p$ ($\epsilon_n = 0.4$, $\alpha_r = 2.0$, $f_d = 0.67$)

In contrast, α_r increases count rate but moves cross-points very little. It should be noted that ratio of the maximum to the minimum in the same M_L is over 1000 when M_L < 1.1 ($\epsilon_n = 0.4$, $f_d = 0.67$) in conventional assay condition, but it reduces rapidly to < 10 when M_L > 1.3, which means observation of high-order correlation is possible if uncertainty is sufficiently small, for example, by fast neutron detection and counting without moderation.

Similar tendency had been reported[16] in 1997 by Ensslin for Pu metal (probably sphere) shown in Figure 5a. Our calculation at the same condition ($\epsilon_n=0.5$, $\alpha_r=0$, $f_d=0.67$) is shown in Figure 5b. This tendency depends constitutively on M_L, and ^{240}Pu mass has positive correlation with M_L. It is recognized that m_{eff} = 40~50 g is equivalent to M_L = 1.4 at the crossing point. Quantitative analysis is possible[d] but it is not so familiar.

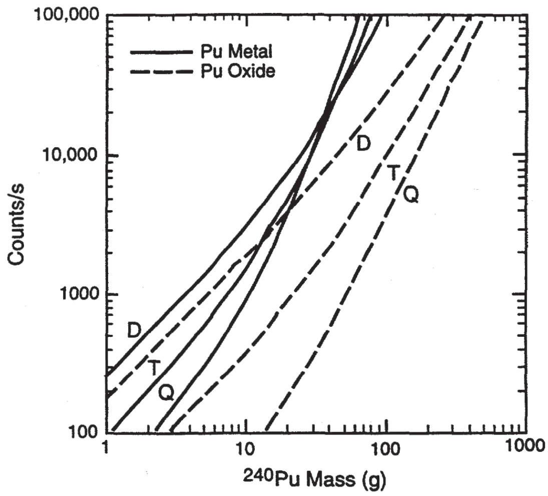


Figure 5a Crossing of $D/T/Q_r$ (reprinted from Fig. 1 of LA-UR-97-2716)[16].

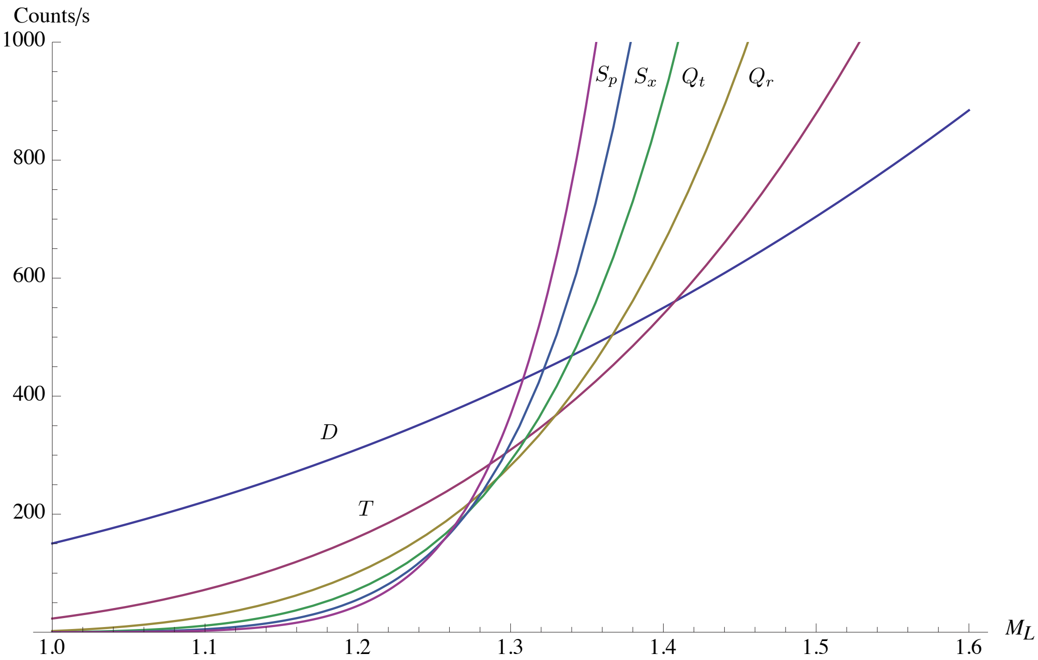


Figure 5b Crossing of $D/T/Q_r/Q_t/S_x/S_p$ ($\epsilon_n=0.5$, $\alpha_r=0$, $f_d=0.67$)

---

d. The ratio of fast fission cross section to total cross section is estimated to be 0.2~0.25, if isotopic composition of ^{239}Pu/^{240}Pu/^{241}Pu is 0.9/0.095/0.005. Probability of induced fission p is calculated inversely from M_L=1.4 to be 0.117. Then P_E, probability of leak without collision (pp.106-127 of ref.7), is estimated to be 0.557~0.602. By the way, mean free path of 2 MeV neutron is ~ 2.77 cm. From P_E and the path, radius of Pu metal sphere is estimated to be 1.56~1.80 cm. Estimated weight is 315~483 g and estimated m_{eff} is 30~46 g. Therefore, Figure 5b and 5a are highly consistent.

3. Fission rates and doubles count rates in fast neutron and thermal neutron coexisting system

3.1 Introduction

Differential die-away self-interrogation (DDSI) is an interesting technique for next generation technology for safeguards in Japan, where a sample of U-Pu-Cm mixed oxide in water will have to be measured and verified in the near future. One-point theory for fast neutron applied to a sample in the air and conventional neutron coincidence counting is difficult to be applied because fast neutron and thermal neutron coexists. There is another problem that neutrons from ^{244}Cm spontaneous fission is massive, so neutrons from U and Pu fast fission are dimmed. DDSI can distinguish thermal fission neutrons from fast/spontaneous fission neutrons by observing Rossi-alpha combined distribution, which is ranked as the second choice to measure and verify a spent fuel[17] in water. The device is similar to the conventional one, which is important for use in the near future.

In this section, Kolmogorov forward approach for the variance to mean (called Feynman-alpha) presented in ESARDA 2011[18] and published[19,20] in 2012 by Anderson, Pál and Pázsit was modified and solved again as two-point theory for DDSI. The process to solve Kolmogorov forward equation using PGF was explained step-by-step, and new formulae expressed by their stochastic variables were obtained, followed by a hypothetical stochastic variable (equal to the result of subtracting 1 from 'variance to mean' in their papers) corresponds to Y-value (introduced in ref.8) was derived. Then, some combinations of stochastic variables were interpreted based on the hypothesis to equations expressed by conventional variables i.e. singles count rate, doubles count rate, efficiency (counts per neutron) and leakage multiplication, etc. From these equations and observed Rossi-alpha combined distribution, it is possible to estimate: the number of induced fissions by fast and by thermal individually; the number of induced fission (< 1) by one neutron individually; and individual doubles count rate (depends on detector and counter setting), and so on. At the end of research, the hypothesis was proved to be true. Provisional calculations were done for UO_2 of 1~10 kgU containing ~ 0.009 wt% ^{244}Cm.

3.2 Kolmogorov forward differential equation for neutron fluctuation

Kolmogorov forward differential equation is very useful to express Markov chain with countable-state spaces[e] where interarrival or waiting times of transition of an event distributes exponentially[21]. Poisson process matches the requirement, so it is possible to express neutron fluctuation by Kolmogorov forward differential equation.

e. So-called 'continuous-time Markov chain' against 'finite-state Markov chain' or 'discrete-time Markov chain'.

Figure 6 shows flow and balance of fast and thermal neutrons in steady state though fluctuations, where $P(N_1,N_2,Z_1,t)$ is the probability having N_1,N_2,Z_1 neutrons (fast, thermal and detected, respectively) at time t, each λs is probabilities of various reactions in the flow for a neutron for a unit time, ν is the number of neutrons from a fission, p_ν, f_ν^1, f_ν^2 are probability distributions (spontaneous, induced by fast and by thermal, respectively) and S_1 is the number of spontaneous fissions for a unit time.

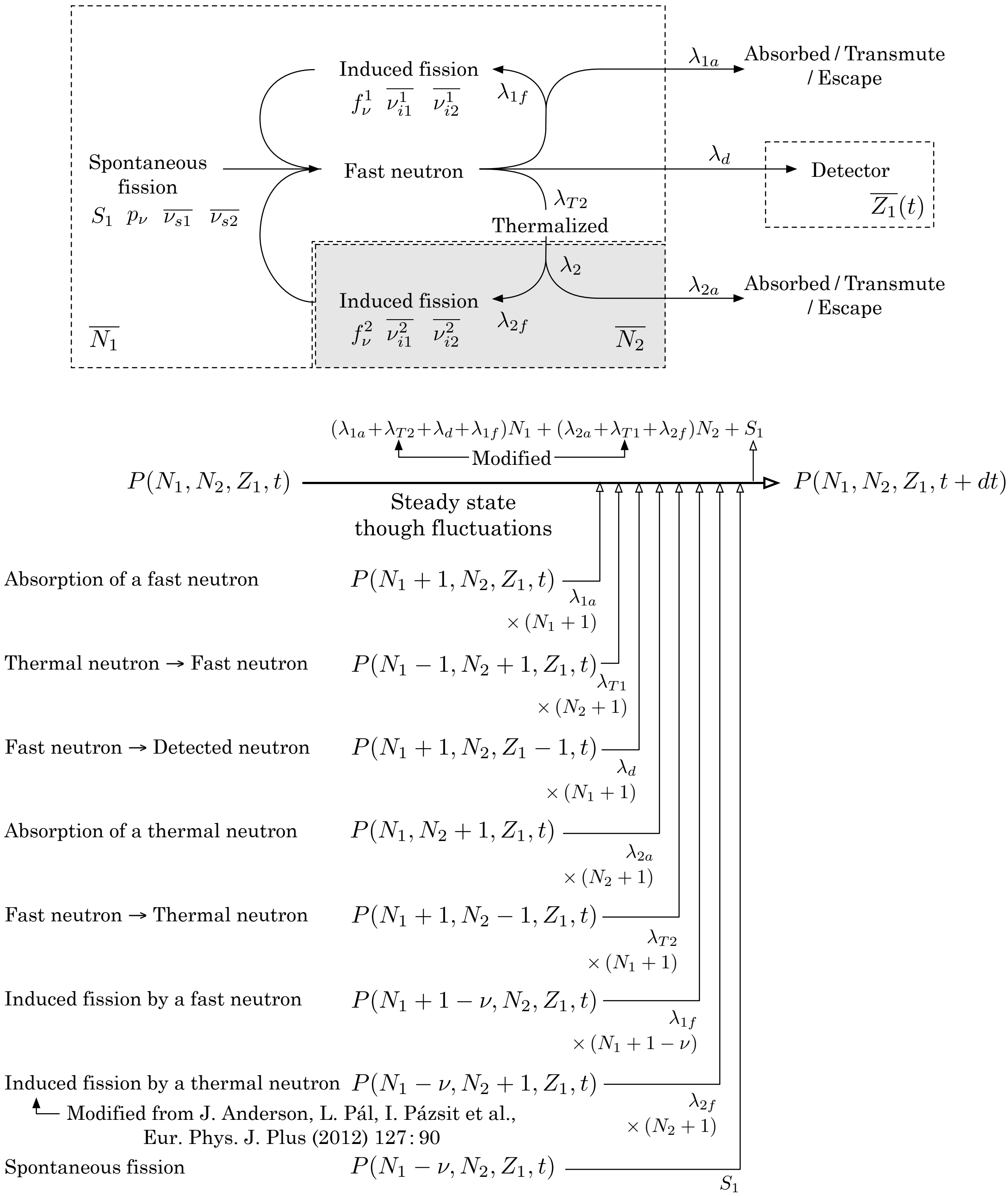


Figure 6 Flow and balance of fast and thermal neutrons

Kolmogorov forward differential equation in steady state though fluctuations is shown below (ref.20 is more recent than ref.19, but thermal fission term in ref.19 is correct):

$$\begin{aligned}
\frac{\partial P(N_1,N_2,Z_1,t)}{\partial t} = &+\lambda_{1a}(N_1+1)P(N_1+1,N_2,Z_1,t)\\
&+\lambda_{T1}(N_2+1)P(N_1-1,N_2+1,Z_1,t)\\
&+\lambda_{d}(N_1+1)P(N_1+1,N_2,Z_1-1,t)\\
&+\lambda_{2a}(N_2+1)P(N_1,N_2+1,Z_1,t)\\
&+\lambda_{T2}(N_1+1)P(N_1+1,N_2-1,Z_1,t)\\
&+\lambda_{1f}\sum_{\nu}(N_1+1-\nu)f^1_\nu P(N_1+1-\nu,N_2,Z_1,t)\\
&+\lambda_{2f}\sum_{\nu}(N_2+1)f^2_\nu P(N_1-\nu,N_2+1,Z_1,t)\\
&+S_1\sum_{\nu}p_\nu P(N_1-\nu,N_2,Z_1,t)\\
&-[(\lambda_{1a}+\lambda_{T2}+\lambda_d+\lambda_{1f})N_1\\
&\qquad+(\lambda_{2a}+\lambda_{T1}+\lambda_{2f})N_2+S_1]\,P(N_1,N_2,Z_1,t)
\end{aligned} \tag{23}$$

PGF for the probability $P(N_1,N_2,Z_1,t)$ is given as:

$$G(X,Y,Z,t)=\sum_{N_1}\sum_{N_2}\sum_{Z_1}X^{N_1}Y^{N_2}Z^{Z_1}P(N_1,N_2,Z_1,t) \tag{24}$$

PGF for other probabilities $(N_1+1)P(N_1+1,N_2,Z_1,t)$, … are given as[f]:

$$\frac{\partial G}{\partial X}=\sum_{N_1}\sum_{N_2}\sum_{Z_1}X^{N_1}Y^{N_2}Z^{Z_1}(N_1+1)P(N_1+1,N_2,Z_1,t) \tag{25}$$

$$\frac{\partial G}{\partial Y}=\sum_{N_1}\sum_{N_2}\sum_{Z_1}X^{N_1}Y^{N_2}Z^{Z_1}(N_2+1)P(N_1,N_2+1,Z_1,t) \tag{26}$$

$$X\frac{\partial G}{\partial Y}=\sum_{N_1}\sum_{N_2}\sum_{Z_1}X^{N_1}Y^{N_2}Z^{Z_1}(N_2+1)P(N_1-1,N_2+1,Z_1,t) \tag{27}$$

$$Y\frac{\partial G}{\partial X}=\sum_{N_1}\sum_{N_2}\sum_{Z_1}X^{N_1}Y^{N_2}Z^{Z_1}(N_1+1)P(N_1+1,N_2-1,Z_1,t) \tag{28}$$

$$Z\frac{\partial G}{\partial X}=\sum_{N_1}\sum_{N_2}\sum_{Z_1}X^{N_1}Y^{N_2}Z^{Z_1}(N_1+1)P(N_1+1,N_2,Z_1-1,t) \tag{29}$$

f. $\frac{\partial G}{\partial X}=\sum_{N_1=2}\sum_{N_2}\sum_{Z_1}X^{N_1-1}Y^{N_2}Z^{Z_1}N_1P(N_1,N_2,Z_1,t)=\sum_{N_1}\sum_{N_2}\sum_{Z_1}X^{N_1}Y^{N_2}Z^{Z_1}(N_1+1)P(N_1+1,N_2,Z_1,t)$

$Y\frac{\partial G}{\partial X}=\sum_{N_1}\sum_{N_2}\sum_{Z_1}X^{N_1}Y^{N_2+1}Z^{Z_1}(N_1+1)P(N_1+1,N_2,Z_1,t)=\sum_{N_1}\sum_{N_2=2}\sum_{Z_1}X^{N_1}Y^{N_2}Z^{Z_1}(N_1+1)P(N_1+1,N_2-1,Z_1,t)$

$$X^{\nu}\frac{\partial G}{\partial Y}=\sum_{N_1}\sum_{N_2}\sum_{Z_1}X^{N_1}Y^{N_2}Z^{Z_1}(N_2+1)P(N_1-\nu,N_2+1,Z_1,t) \tag{30}$$

$$X^{\nu}\frac{\partial G}{\partial X}=\sum_{N_1}\sum_{N_2}\sum_{Z_1}X^{N_1}Y^{N_2}Z^{Z_1}(N_1+1-\nu)P(N_1+1-\nu,N_2,Z_1,t) \tag{31}$$

$$\sum_{\nu}f_{\nu}^{2}X^{\nu}\frac{\partial G}{\partial Y}=\sum_{N_1}\sum_{N_2}\sum_{Z_1}X^{N_1}Y^{N_2}Z^{Z_1}\sum_{\nu}(N_2+1)f_{\nu}^{2}P(N_1-\nu,N_2+1,Z_1,t) \tag{32}$$

$$\sum_{\nu}f_{\nu}^{1}X^{\nu}\frac{\partial G}{\partial X}=\sum_{N_1}\sum_{N_2}\sum_{Z_1}X^{N_1}Y^{N_2}Z^{Z_1}\sum_{\nu}(N_1+1-\nu)f_{\nu}^{1}P(N_1+1-\nu,N_2,Z_1,t) \tag{33}$$

$$\sum_{\nu}p_{\nu}X^{\nu}G=\sum_{N_1}\sum_{N_2}\sum_{Z_1}X^{N_1}Y^{N_2}Z^{Z_1}\sum_{\nu}p_{\nu}P(N_1-\nu,N_2,Z_1,t) \tag{34}$$

3.3 Solving the equation using probability generating function

Master equation that is transformed from Kolmogorov forward differential equation using PGF is expressed as:

$$\begin{aligned}\frac{\partial G}{\partial t}&=(\lambda_{1a}+\lambda_{T2}Y+\lambda_{d}Z+\lambda_{1f}\sum_{\nu}f_{\nu}^{1}X^{\nu})\frac{\partial G}{\partial X}\\&\quad+(\lambda_{2a}+\lambda_{T1}X+\lambda_{2f}\sum_{\nu}f_{\nu}^{2}X^{\nu})\frac{\partial G}{\partial Y}+S_1\sum_{\nu}p_{\nu}X^{\nu}G\\&\quad-(\lambda_{1a}+\lambda_{T2}+\lambda_{d}+\lambda_{1f})X\frac{\partial G}{\partial X}-(\lambda_{2a}+\lambda_{T1}+\lambda_{2f})Y\frac{\partial G}{\partial Y}-S_1G\end{aligned} \tag{35}$$

where some PGF derivatives are associated with real variables or constants:

$$G|_{X=Y=Z=1}=\sum_{N_1}\sum_{N_2}\sum_{Z_1}P(N_1,N_2,Z_1,t)=1 \tag{36}$$

$$\left.\frac{\partial G}{\partial X}\right|_{X=1}=\overline{N_1} \qquad \text{Expected number of fast neutron} \tag{37}$$

$$\left.\frac{\partial G}{\partial Y}\right|_{Y=1}=\overline{N_2} \qquad \text{Expected number of thermal neutron} \tag{38}$$

$$\left.\frac{d}{dX}\sum_{\nu}f_{\nu}^{1}X^{\nu}\right|_{X=1}=\overline{\nu_{i1}^{1}} \qquad \text{1st factorial moment of induced fission by fast neutron} \tag{39}$$

$$\left.\frac{d}{dX}\sum_{\nu}f_{\nu}^{2}X^{\nu}\right|_{X=1}=\overline{\nu_{i1}^{2}} \qquad \text{1st factorial moment of induced fission by thermal neutron} \tag{40}$$

$$\left.\frac{d}{dX}\sum_{\nu}p_{\nu}X^{\nu}\right|_{X=1}=\overline{\nu_{s1}} \qquad \text{1st factorial moment of spontaneous fission} \tag{41}$$

First, apply equation (36) to the master equation and differentiate it with respect to X and Y, then $\partial G/\partial t$ and secondary-derivatives ($\partial^2 G/\partial X^2$, $\partial^2 G/\partial Y^2$, $\partial^2 G/(\partial X \partial Y)$) are replaced by zero. Then apply equations (37)~(41) and solve the simultaneous equation. As a result, expected number of fast neutrons and the number of thermal neutrons are given as:

$$\overline{N_1} = \frac{\lambda_2 S_1 \overline{\nu_{s1}}}{\lambda_1 \lambda_2 - \lambda_{1f} \lambda_2 \overline{\nu_{i1}^1} - \lambda_{2f} \lambda_{T2} \overline{\nu_{i1}^2} - \lambda_{T1} \lambda_{T2}} \tag{42}$$

$$\overline{N_2} = \frac{\lambda_{T2} S_1 \overline{\nu_{s1}}}{\lambda_1 \lambda_2 - \lambda_{1f} \lambda_2 \overline{\nu_{i1}^1} - \lambda_{2f} \lambda_{T2} \overline{\nu_{i1}^2} - \lambda_{T1} \lambda_{T2}} \tag{43}$$

$$\overline{Z_1} = \lambda_d \overline{N_1} t \tag{44}$$

where $\lambda_1 = \lambda_{1a} + \lambda_{T2} + \lambda_d + \lambda_{1f}$ and $\lambda_2 = \lambda_{2a} + \lambda_{T1} + \lambda_{2f}$

Next, apply equation (36) to the master equation and differentiate it to the second order with respect to X, Y and Z, then $\partial G/\partial t$ and thirdly-derivatives are replaced by zero, followed by applying equations (37)~(41) and (45)~(47):

$$\frac{d^2}{dX^2} \sum_\nu f_\nu^1 X^\nu \bigg|_{X=1} = \overline{\nu_i^1(\nu_i^1 - 1)} = \overline{\nu_{i2}^1} \quad \text{2nd factorial moment of induced fission by fast neutron} \tag{45}$$

$$\frac{d^2}{dX^2} \sum_\nu f_\nu^2 X^\nu \bigg|_{X=1} = \overline{\nu_i^2(\nu_i^2 - 1)} = \overline{\nu_{i2}^2} \quad \text{2nd factorial moment of induced fission by thermal neutron} \tag{46}$$

$$\frac{d^2}{dX^2} \sum_\nu p_\nu X^\nu \bigg|_{X=1} = \overline{\nu_s(\nu_s - 1)} = \overline{\nu_{s2}} \quad \text{2nd factorial moment of spontaneous fission} \tag{47}$$

Then the secondary-derivatives ($\frac{\partial^2 G}{\partial X^2}, \frac{\partial^2 G}{\partial Y^2}, \frac{\partial^2 G}{\partial Z^2}, \frac{\partial^2 G}{\partial X \partial Y}, \frac{\partial^2 G}{\partial X \partial Z}, \frac{\partial^2 G}{\partial Y \partial Z}$) are replaced by the symbols μ_{XX}, μ_{YY}, μ_{ZZ}, μ_{XY}, μ_{XZ}, μ_{YZ}, respectively. These symbols are neither mean nor variance nor self-covariance/covariance, only simple replacement proved in the Appendix e). As a result, a set of simultaneous equations are obtained:

$$\frac{\partial}{\partial t} \mu_{XX} = 2\left(-\lambda_1 + \lambda_{1f} \overline{\nu_{i1}^1}\right) \mu_{XX} + 2\left(\lambda_{T1} + \lambda_{2f} \overline{\nu_{i1}^2}\right) \mu_{XY} + \lambda_{1f} \overline{\nu_{i2}^1}\, \overline{N_1} + \lambda_{2f} \overline{\nu_{i2}^2}\, \overline{N_2} + S_1 \overline{\nu_{s2}} \tag{48}$$

$$\frac{\partial}{\partial t} \mu_{YY} = -2\, \lambda_2\, \mu_{YY} + 2\, \lambda_{T2}\, \mu_{XY} \tag{49}$$

$$\frac{\partial}{\partial t} \mu_{ZZ} = 2\, \lambda_d\, \mu_{XZ} \tag{50}$$

$$\frac{\partial}{\partial t}\,\mu_{XY} = (-\lambda_1 - \lambda_2 + \lambda_{1f}\overline{\nu_{i1}^1})\,\mu_{XY} + \lambda_{T2}\,\mu_{XX} + (\lambda_{T1} + \lambda_{2f}\overline{\nu_{i1}^2})\,\mu_{YY} \tag{51}$$

$$\frac{\partial}{\partial t}\,\mu_{XZ} = (-\lambda_1 + \lambda_{1f}\overline{\nu_{i1}^1})\,\mu_{XZ} + \lambda_d\,\mu_{XX} + (\lambda_{T1} + \lambda_{2f}\overline{\nu_{i1}^2})\,\mu_{YZ} \tag{52}$$

$$\frac{\partial}{\partial t}\,\mu_{YZ} = -\lambda_2\,\mu_{YZ} + \lambda_d\,\mu_{XY} + \lambda_{T2}\,\mu_{XZ} \tag{53}$$

where $\frac{\partial}{\partial t}\,\mu_{XX} = \frac{\partial}{\partial t}\,\mu_{YY} = \frac{\partial}{\partial t}\,\mu_{XY} = \frac{\partial}{\partial t}\,\mu_{XZ} = \frac{\partial}{\partial t}\,\mu_{YZ} = 0$ and $\frac{\partial}{\partial t}\,\mu_{ZZ} \neq 0$, therefore:

$$\mu_{XX} = \frac{(\omega_1\,\omega_2 + \lambda_2^2)(\lambda_{1f}\overline{\nu_{i2}^1}\,\overline{N_1} + \lambda_{2f}\overline{\nu_{i2}^2}\,\overline{N_2} + S_1\overline{\nu_{s2}})}{2(\lambda_1 + \lambda_2 - \lambda_{1f}\overline{\nu_{i1}^1}\,)\,\omega_1\,\omega_2} \tag{54}$$

$$\mu_{YY} = \frac{\lambda_{T2}^2}{\omega_1\,\omega_2 + \lambda_2^2}\,\mu_{XX} \tag{55}$$

$$\mu_{XY} = \frac{\lambda_2\,\lambda_{T2}}{\omega_1\,\omega_2 + \lambda_2^2}\,\mu_{XX} \tag{56}$$

$$\omega_1\,\omega_2 = \lambda_1\lambda_2 - \lambda_{1f}\lambda_2\overline{\nu_{i1}^1} - \lambda_{2f}\lambda_{T2}\overline{\nu_{i1}^2} - \lambda_{T1}\lambda_{T2} \tag{57}$$

where equation (57) is the denominator of $\overline{N_1}$ and $\overline{N_2}$. These results are equal to the ones in ref.19, when $\lambda_{1f} \to 0$ because induced fission by fast neutron is not considered in ref.19, $\lambda_{T1} \to 0$ because probability of thermal to fast is little (only by delayed fission) and $\lambda_{T2} \to \lambda_R$ because λ_R is used instead of λ_{T2}. The results below are obtained:

$$\begin{aligned}\mu_{XZ} &= \frac{\lambda_d\,(\lambda_{T1} + \lambda_{2f}\overline{\nu_{i1}^2})}{\omega_1\,\omega_2}\,\mu_{XY} + \frac{\lambda_d\,\lambda_2}{\omega_1\,\omega_2}\,\mu_{XX} \\ &= \frac{\lambda_d\,\lambda_2^2}{2\,\omega_1^2\,\omega_2^2}\,(\lambda_{1f}\overline{\nu_{i2}^1}\,\overline{N_1} + \lambda_{2f}\overline{\nu_{i2}^2}\,\overline{N_2} + S_1\overline{\nu_{s2}})\end{aligned} \tag{58}$$

$$\mu_{ZZ} = \frac{\lambda_d^2\,\lambda_2^2}{\omega_1^2\,\omega_2^2}\,(\lambda_{1f}\overline{\nu_{i2}^1}\,\overline{N_1} + \lambda_{2f}\overline{\nu_{i2}^2}\,\overline{N_2} + S_1\overline{\nu_{s2}})\;t \tag{59}$$

Here, a measure is introduced to eliminate t:

$$\frac{\mu_{ZZ}}{\overline{Z_1}} = \frac{\lambda_d\,\lambda_2^2}{\omega_1^2\,\omega_2^2}\,(\lambda_{1f}\overline{\nu_{i2}^1} + \lambda_{2f}\overline{\nu_{i2}^2}\,\frac{\overline{N_2}}{\overline{N_1}} + \overline{\nu_{s2}}\,\frac{S_1}{\overline{N_1}}) \tag{60}$$

The measure is modified using $\epsilon = \lambda_d/\lambda_{1f}$ (counts per fast fission) as:

$$\frac{\mu_{ZZ}}{\overline{Z_1}} = \epsilon\left(\frac{\lambda_2\,\lambda_{1f}}{\omega_1\,\omega_2}\right)^2\left(\overline{\nu_{i2}^1} + \frac{\lambda_{2f}\,\overline{N_2}}{\lambda_{1f}\,\overline{N_1}}\,\overline{\nu_{i2}^2} + \frac{S_1}{\lambda_{1f}\,\overline{N_1}}\,\overline{\nu_{s2}}\right) \tag{61}$$

It should be noted that the measure is proportional to the linear coupling of $\overline{\nu_{i2}^1}$, $\overline{\nu_{i2}^2}$ and $\overline{\nu_{s2}}$, i.e. the 2nd factorial moments of fast, thermal and spontaneous fission.

3.4 A hypothetical stochastic variable corresponding to Y-value

Y-value in excess of random Poisson fluctuations by Hoffmann[8] in 1949 is defined as:

$$\frac{\overline{c^2} - (\overline{c})^2}{\overline{c}} \stackrel{\text{def}}{=} 1 + Y \tag{62}$$

$$Y = \frac{\epsilon\,(\overline{\nu^2} - \overline{\nu})}{(\alpha\tau)^2} = \frac{\epsilon\,\overline{\nu(\nu-1)}}{(\alpha\tau)^2} = \epsilon\left(\frac{1}{\alpha\tau}\right)^2 \overline{\nu_2} \tag{63}$$

where c is count rate, τ is mean time between fissions, ϵ is efficiency (counts/fission), $\overline{\nu_2}$ is 2nd factorial moment of induced fission and α is rate of decay regarding prompt neutrons. $e^{-\alpha t}$ will be the expected number of neutrons present at time t due to one primary neutron introduced into boiler by Feynman, Hoffmann and Serber[22] in 1956. Equation (62) means variance to mean of count rate c, and Y-value is zero for Poisson processes where variance to mean is one. Y-value is different from variable Y of PGF.

The measure in the previous subsection and the Y-value looks like physically equivalent though the measure is extended (three types of fission included) and unusual because Z is PGF variable and Z_1 is real variable. Anyway, a hypothetical stochastic variable shown below is introduced:

$$\frac{\mu_{ZZ}}{\overline{Z_1}} = Y \tag{64}$$

By the way, there is a simple relationship between Y-value, singles count rate S and doubles count rate D[g], where f_d is same as the one in equation (15):

$$D = \frac{1}{2}\, f_d\, S\, Y \tag{65}$$

$$\frac{D}{f_d} = \frac{F\,\epsilon^2\,(\overline{\nu^2} - \overline{\nu})}{2\,\alpha^2\,\tau^2}\left(1 - \frac{1 - e^{-\alpha t}}{\alpha t}\right) \tag{66}$$

$$S = F\epsilon = \frac{\overline{Z_1}}{t} = \lambda_d\,\overline{N_1} \tag{67}$$

$$Y = \frac{\epsilon\,(\overline{\nu^2} - \overline{\nu})}{(\alpha\tau)^2}\left(1 - \frac{1 - e^{-\alpha t}}{\alpha t}\right) \tag{68}$$

For reference, 'variance to mean' in ref.19 and ref.20 is equal to variance of Z (not μ_{ZZ} but σ_{ZZ}^2) to mean of Z_1, thus subtracting 1 from 'variance to mean' means Y-value.

g. Refer to the Reference 10 and Appendix a).

3.5 Relations of stochastic variables with conventional singles, doubles, etc.

The term $\left(1-\dfrac{1-e^{-\alpha t}}{\alpha t}\right)$ in equation (66) and (68) becomes to one if $t \to \infty$, therefore Y-value in equation (63) and (64) can only be applied to a stable state. In such a case, the term $1/(\alpha\tau)$ means the number of induced fissions yields from one neutron (mentioned in ref.8), and $\overline{\nu}/(\alpha\tau)$ means the number of induced neutrons yields from one neutron. In a stable state, an important relation below is derived where the hypothetical stochastic variable serves as a medium:

$$\frac{D}{f_d} = \frac{\mu_{ZZ}}{2\,t} = \frac{1}{2}\,\frac{\lambda_d^2\,\lambda_2^2}{\omega_1^2\,\omega_2^2}\,(\lambda_{1f}\overline{\nu_{i2}^1}\,\overline{N_1}+\lambda_{2f}\overline{\nu_{i2}^2}\,\overline{N_2}+S_1\overline{\nu_{s2}}) \tag{69}$$

where the physical meaning of λ_2 is given by Figure 6 as a probability of input thermal neutron based on $\overline{N_2}$:

$$\lambda_2\,\overline{N_2} = \lambda_{T2}\,\overline{N_1} \;\to\; \lambda_2 = \frac{\overline{N_1}}{\overline{N_2}}\,\lambda_{T2} \tag{70}$$

Here, expected number of induced fissions by fast neutron per unit time $\overline{F_1} = \lambda_{1f}\,\overline{N_1}$ and expected number of induced fissions by thermal neutron per unit time $\overline{F_2} = \lambda_{2f}\,\overline{N_2}$ are introduced:

$$\frac{D}{f_d} = \frac{1}{2}\underbrace{\lambda_d^2\,\frac{\lambda_{T2}^2}{\omega_1^2\,\omega_2^2}\left(\frac{\overline{N_1}}{\overline{N_2}}\right)^2}_{\substack{\text{Square of [Efficiency (counts/neuteon)}\\ \times\text{ leakage mulplication]}}}\underbrace{\left(\overline{F_1}\,\overline{\nu_{i2}^1}+\overline{F_2}\,\overline{\nu_{i2}^2}+S_1\,\overline{\nu_{s2}}\right)}_{\substack{\text{Sum of fission rate} \times \text{2nd factorial moment}\\ \text{that means twice of the number of neutron pairs}}} \tag{71}$$

also:

$$S = \lambda_d\,\frac{\lambda_{T2}}{\omega_1\,\omega_2}\,\frac{\overline{N_1}}{\overline{N_2}}\,S_1\,\overline{\nu_{s1}} \qquad \left[\; S \neq \lambda_d\,\frac{\lambda_{T2}}{\omega_1\,\omega_2}\,\frac{\overline{N_1}}{\overline{N_2}}\left(\overline{F_1}\,\overline{\nu_{i1}^1}+\overline{F_2}\,\overline{\nu_{i1}^2}+S_1\,\overline{\nu_{s1}}\right)\;\right] \tag{72}$$

and:

$$\frac{\lambda_d\,\lambda_2}{\omega_1\,\omega_2} = \lambda_d\,\frac{\lambda_{T2}}{\omega_1\,\omega_2}\,\frac{\overline{N_1}}{\overline{N_2}} = \epsilon_n\,M_L \tag{73}$$

where $\epsilon_n\,(\neq\epsilon)$ is efficiency (counts/neutron) normally used in coincidence counting for safeguards and M_L is leakage multiplication. S is not expressed by linear coupling of $\overline{\nu_{i1}^1}$, $\overline{\nu_{i1}^2}$ and $\overline{\nu_{s1}}$, because M_L is composed of the linear coupling, i.e. the 1st factorial moments of fast, thermal and spontaneous fission, proved later.

As a result, equation (71) and (72) are expressed using conventional variables:

$$\frac{D}{f_d} = \frac{1}{2}\,\epsilon_n^2\,M_L^2\,\left(S_1\overline{\nu_{s2}} + \overline{F_1}\,\overline{\nu_{i2}^1} + \overline{F_2}\,\overline{\nu_{i2}^2}\right) \tag{74}$$

$$S = \epsilon_n\,M_L\,S_1\,\overline{\nu_{s1}} \tag{75}$$

To separate ϵ_n and M_L, physical meaning of ϵ_n is considered, and ϵ_n is given as:

$$\epsilon_n = \frac{\lambda_d}{\lambda_{1f}\left(\overline{\nu_{i1}^1} + \frac{\overline{F_2}}{\overline{F_1}}\,\overline{\nu_{i1}^2} + \frac{S_1}{\overline{F_1}}\,\overline{\nu_{s1}}\right)} = \frac{\lambda_d\,\lambda_2}{\lambda_1\,\lambda_2 - \lambda_{T1}\,\lambda_{T2}} \tag{76}$$

If λ_{T1} is supposed to be zero due to its small probability of delayed neutron from fission product, the following equations are derived:

$$\epsilon_n = \frac{\lambda_d}{\lambda_1} \tag{77}$$

$$M_L = \frac{\lambda_1\,\lambda_2}{\omega_1\,\omega_2} = 1 + \frac{\lambda_2\,\lambda_{1f}\,\overline{\nu_{i1}^1} + \lambda_{T2}\,\lambda_{2f}\,\overline{\nu_{i1}^2}}{\omega_1\,\omega_2} \quad \geq 1 \tag{78}$$

The important equation below regarding M_L is obtained:

$$M_L = \frac{1}{S_1\,\overline{\nu_{s1}}}\,\left(S_1\,\overline{\nu_{s1}} + \overline{F_1}\,\overline{\nu_{i1}^1} + \overline{F_2}\,\overline{\nu_{i1}^2}\right) \tag{79}$$

$$\because\ \omega_1\,\omega_2 = \frac{\lambda_{T2}\,S_1\,\overline{\nu_{s1}}}{\overline{N_2}} = \frac{\lambda_2\,S_1\,\overline{\nu_{s1}}}{\overline{N_1}} \tag{80}$$

For equation (80), $\omega_1\,\omega_2$ means physically a rate of neutron supply from spontaneous fission (source of all neutrons) to keep the number of thermal neutrons be constant, which results in keeping the number of fast neutrons be constant. So, $\omega_1\,\omega_2$ is closely associated with the rate of decay α in *Y*-value. From the similarity between equation (61) and (63), equations below are derived for induced fission by fast neutron:

$$\frac{1}{\alpha\tau} = \frac{\lambda_2\,\lambda_{1f}}{\omega_1\,\omega_2} \tag{81}$$

$$S = \epsilon\,\frac{1}{\alpha\,\tau}\,S_1\,\overline{\nu_{s1}} \ \rightarrow\ \frac{1}{\alpha\,\tau} = \frac{\epsilon_n}{\epsilon}\,M_L \tag{82}$$

From equation (82), it becomes clear that equation (81) corresponds to the 'net' number of fast induced fissions by one source neutron, because S is count rate of fast neutron involving thermal fission neutrons and M_L includes both fast and thermal.

From equation (79), it is clear that M_L is composed of linear coupling of $\overline{\nu_{i1}^1}$, $\overline{\nu_{i1}^2}$ and $\overline{\nu_{s1}}$. It should be noted that equation (79) looks like different from the conventional definition of M_L (equation (1) and Figure 7), though equation (79) is intuitively and truly correct as 'net' multiplication. $\overline{N_1}$ and $\overline{N_2}$ are at steady state as a result of leakage, so $\overline{F_1}$ and $\overline{F_2}$ are also the result of leakage, that is the meaning of 'net' multiplication. Obtained M_L and conventional M_L is same although viewpoint is different. See Appendix f) for detail.

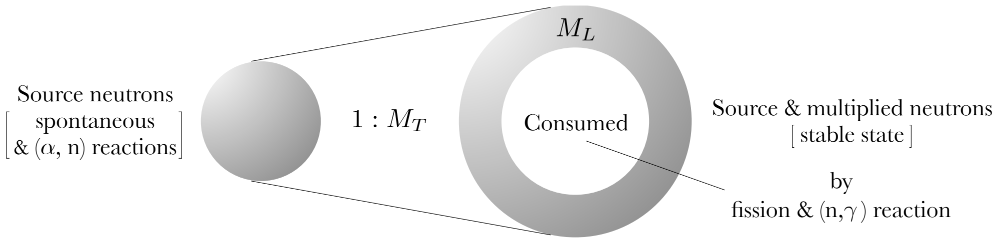


Figure 7 Conventional definition of M_L

3.6 Separating contribution of thermal neutron

It is possible to separate the contribution of fast neutron on Y-value and the one of thermal neutron on Y-value. First, efficiencies (counts/fission) based on $\overline{N_1}$ and on $\overline{N_2}$ are defined:

$$\epsilon_1 = \epsilon = \frac{\lambda_d}{\lambda_{1f}} = \frac{S}{\overline{F_1}} \qquad \text{for } \overline{N_1} \tag{83}$$

$$\epsilon_2 = \frac{\lambda_d}{\lambda_{2f}} \frac{\overline{N_1}}{\overline{N_2}} = \frac{S}{\overline{F_2}} \qquad \text{for } \overline{N_2} \tag{84}$$

$$\frac{\mu_{ZZ}}{\overline{Z_1}} = \underbrace{\epsilon_1 \left(\frac{\lambda_2\,\lambda_{1f}}{\omega_1\,\omega_2} \right)^2 \left(\overline{\nu_{i2}^1} + \frac{S_1}{\overline{F_1}} \overline{\nu_{s2}} \right)}_{Y_1 \text{ based on } \overline{N_1}} + \underbrace{\epsilon_2 \left(\frac{\lambda_2\,\lambda_{2f}}{\omega_1\,\omega_2} \frac{\overline{N_2}}{\overline{N_1}} \right)^2 \overline{\nu_{i2}^2}}_{Y_2 \text{ based on } \overline{N_2}} \tag{85}$$

From the view point of similarity between equation (63) and (85), the following equation is derived:

$$\alpha_1\,\tau_1 = \frac{\omega_1\,\omega_2}{\lambda_{1f}\,\lambda_2} \qquad \text{for } Y_1 \tag{86}$$

$$\alpha_2\,\tau_2 = \frac{\omega_1\,\omega_2}{\lambda_2\,\lambda_{2f}} \frac{\overline{N_1}}{\overline{N_2}} = \frac{\omega_1\,\omega_2}{\lambda_{2f}\,\lambda_{T2}} \qquad \text{for } Y_2 \tag{87}$$

where $1/(\alpha_1\,\tau_1)$ is the 'net' number of fast induced fissions by one source neutron, whereas $1/(\alpha_2\,\tau_2)$ is the 'net' number of thermal induced fissions by one source neutron.

Next, apply equation (57) to equation (86) and (87) to eliminate $\omega_1 \omega_2$. The following equations are obtained assuming $\lambda_{T1} \to 0$:

$$\alpha_1 \tau_1 = \frac{1}{\overline{F_1}} \left[\frac{\lambda_1}{\lambda_{1f}} \overline{F_1} - \overline{F_1}\, \overline{\nu_{i1}^1} - \overline{F_2}\, \overline{\nu_{i1}^2} \right] \tag{88}$$

$$\alpha_2 \tau_2 = \frac{1}{\overline{F_2}} \left[\frac{\lambda_1}{\lambda_{1f}} \overline{F_1} - \overline{F_1}\, \overline{\nu_{i1}^1} - \overline{F_2}\, \overline{\nu_{i1}^2} \right] \tag{89}$$

An important set of equations is obtained using a parameter r:

$$\overline{F_1} = \frac{r}{\alpha_1 \tau_1} \tag{90}$$

$$\overline{F_2} = \frac{r}{\alpha_2 \tau_2} \tag{91}$$

$$M_L - 1 = \frac{1}{\alpha_1 \tau_1} \overline{\nu_{i1}^1} + \frac{1}{\alpha_2 \tau_2} \overline{\nu_{i1}^2} = \frac{1}{r} \left(\overline{F_1}\, \overline{\nu_{i1}^1} + \overline{F_2}\, \overline{\nu_{i1}^2} \right) \tag{92}$$

$$\begin{aligned} r &= \frac{\lambda_1}{\lambda_{1f}} \overline{F_1} - \overline{F_1}\, \overline{\nu_{i1}^1} - \overline{F_2}\, \overline{\nu_{i1}^2} = \lambda_1 \overline{N_1} - \underbrace{\left(\overline{F_1}\, \overline{\nu_{i1}^1} + \overline{F_2}\, \overline{\nu_{i1}^2} \right)}_{\text{multiplication}} \\ &= S_1 \overline{\nu_{s1}} \end{aligned} \tag{93}$$

$$r\, M_L = \underbrace{r}_{\text{source}} + \underbrace{r\,(M_L - 1)}_{\text{multiplication}} = \lambda_1 \overline{N_1} \tag{94}$$

From equation (86), (87), (90) and (91), it becomes clear that one source neutron mentioned in equation (81), (86) and (87) is a neutron from spontaneous fission, and r means the total number of source neutrons per unit time. It also becomes clear that $\overline{F_1}$ is 'net' number of fast induced fissions in the system affected by thermal induced fission and leakage shown in Figure 6. $\overline{F_1}$ is different from the one simply estimated from fissile density, thickness, cross-section and neutron flux in a large sample without moderation. It is also possible to express Y_1 and Y_2 using r:

$$Y_1 = \frac{1}{r} \epsilon_n M_L \left(\overline{F_1}\, \overline{\nu_{i2}^1} + S_1 \overline{\nu_{s2}} \right) = \frac{1}{r} \epsilon_1 \frac{1}{\alpha_1 \tau_1} \left(\overline{F_1}\, \overline{\nu_{i2}^1} + S_1 \overline{\nu_{s2}} \right) \tag{95}$$

$$Y_2 = \frac{1}{r} \epsilon_n M_L \overline{F_2}\, \overline{\nu_{i2}^2} = \frac{1}{r} \epsilon_2 \frac{1}{\alpha_2 \tau_2} \overline{F_2}\, \overline{\nu_{i2}^2} \tag{96}$$

Equation (95) and (96) satisfies the relation $\dfrac{\mu_{ZZ}}{\overline{Z_1}} = Y = Y_1 + Y_2$. These equations are useful to obtain ϵ_1 and ϵ_2 independently after determining Y_1 and Y_2 independently in the next subsection. Y is given by equation (65).

3.7 Evaluating induced fission rates by fast neutron and by thermal neutron

First, equation (69) is divided to separate the contribution of fast neutron on D_1/f_d and the one of thermal neutron on D_2/f_d. It is important that the ratio of D_1 to D_2 is independent from the stochastic parameters $\lambda_{(\mathrm{s})}$. To apply the same f_d both to D_1 and to D_2, it is necessary to adjust parameters of counting unit, if not, the following equation have to be modified a little. In any case, the ratio is independent from the stochastic parameters $\lambda_{(\mathrm{s})}$.

$$\frac{D_1}{f_d} = \frac{1}{2}\,\frac{\lambda_d^2\,\lambda_2^2}{\omega_1^2\,\omega_2^2}\left(\overline{F_1}\,\overline{\nu_{i2}^1} + S_1\overline{\nu_{s2}}\right) \tag{97}$$

$$\frac{D_2}{f_d} = \frac{1}{2}\,\frac{\lambda_d^2\,\lambda_2^2}{\omega_1^2\,\omega_2^2}\,\overline{F_2}\,\overline{\nu_{i2}^2} \tag{98}$$

$$\frac{D_1}{D_2} = \frac{\overline{F_1}\,\overline{\nu_{i2}^1}}{\overline{F_2}\,\overline{\nu_{i2}^2}} + \frac{S_1\,\overline{\nu_{s2}}}{\overline{F_2}\,\overline{\nu_{i2}^2}} \tag{99}$$

From equation (92):

$$\overline{F_2} = \frac{1}{\overline{\nu_{i1}^2}}\left[r\,(M_L - 1) - \overline{F_1}\,\overline{\nu_{i1}^1}\right] \tag{100}$$

Therefore, the number of induced fissions $\overline{F_1}$ and $\overline{F_2}$ are obtained using the ratio of D_1 to D_2 (D_1/D_2), M_L, S_1, 1st and 2nd factorial moments. This equation is useful because determination of (D_1/D_2) is easier than determination of D_1 and D_2 independently:

$$\overline{F_1} = S_1\,\frac{(D_1/D_2)\,\overline{\nu_{i2}^2}\,\overline{\nu_{s1}}\,(M_L - 1) - \overline{\nu_{i1}^2}\,\overline{\nu_{s2}}}{(D_1/D_2)\,\overline{\nu_{i1}^1}\,\overline{\nu_{i2}^2} + \overline{\nu_{i1}^2}\,\overline{\nu_{i2}^1}} \tag{101}$$

$$\overline{F_2} = S_1\,\frac{\overline{\nu_{i2}^1}\,\overline{\nu_{s1}}\,(M_L - 1) + \overline{\nu_{i1}^1}\,\overline{\nu_{s2}}}{(D_1/D_2)\,\overline{\nu_{i1}^1}\,\overline{\nu_{i2}^2} + \overline{\nu_{i1}^2}\,\overline{\nu_{i2}^1}} \tag{102}$$

Simultaneously, the number of induced fissions by one source neutron, i.e. $1/(\alpha_1\,\tau_1)$ and $1/(\alpha_2\,\tau_2)$, are determined from $r = S_1\,\overline{\nu_{s1}}$ and equation (90) and (91). Consistency between equations (65), (75), (95) and (95) is proved from equation (97), (98) and (73):

$$Y_1 = \frac{1}{r}\,\epsilon_n M_L\left(\overline{F_1}\,\overline{\nu_{i2}^1} + S_1\,\overline{\nu_{s2}}\right) = \frac{1}{r}\,\frac{2}{\epsilon_n M_L}\,\frac{D_1}{f_d} = \frac{2}{S}\,\frac{D_1}{f_d} \tag{103}$$

$$Y_2 = \frac{1}{r}\,\epsilon_n M_L\,\overline{F_2}\,\overline{\nu_{i2}^2} = \frac{1}{r}\,\frac{2}{\epsilon_n M_L}\,\frac{D_2}{f_d} = \frac{2}{S}\,\frac{D_2}{f_d} \tag{104}$$

The ratio (D_1/D_2) is equal to the ratio of Y_1 to Y_2 (Y_1/Y_2).

Thus, Y_1 and Y_2 are obtained from $Y = (2\,D)/(S\,f_d)$ and (D_1/D_2) as:

$$Y_1 = Y\,\frac{1}{1 + D_2/D_1} \tag{105}$$

$$Y_2 = Y\,\frac{1}{1 + D_1/D_2} \tag{106}$$

Therefore, ϵ_1 and ϵ_2 are written also using Y_1 and Y_2:

$$\epsilon_1 = \alpha_1\,\tau_1\,\epsilon_n\,M_L = \alpha_1\,\tau_1\,Y_1\,\frac{S_1\,\overline{\nu_{s1}}}{\overline{F_1}\,\overline{\nu_{i2}^1} + S_1\,\overline{\nu_{s2}}} \tag{107}$$

$$\epsilon_2 = \alpha_2\,\tau_2\,\epsilon_n\,M_L = \alpha_2\,\tau_2\,Y_2\,\frac{S_1\,\overline{\nu_{s1}}}{\overline{F_2}\,\overline{\nu_{i2}^2}} \tag{108}$$

These equations satisfies the relation $\epsilon_1/\epsilon_2 = \overline{F_2}/\overline{F_1} = (\alpha_1\,\tau_1)/(\alpha_2\,\tau_2)$. It is not necessary that both ϵ_1 and ϵ_2 are < 1 (∵ counts/fission) whereas ϵ_n < 1 (∵ counts/neutron).

3.8 Proof of the hypothetical stochastic variable corresponding to *Y*-value

It became clear that the hypothetical stochastic variable $\dfrac{\mu_{ZZ}}{\overline{Z_1}}$ has good consistency with Y-value though it is unusual because Z $(|Z| \leq 1)$ is PGF variable and Z_1 is real discrete stochastic variables (non-negative integers). According to the definition of Y-value by Hoffmann (ref.8), Y is written as:

$$Y + 1 = \frac{\overline{(\lambda_d N_1 - \lambda_d \overline{N_1})^2}}{\lambda_d \overline{N_1}} = \frac{\text{variance of } S = \lambda_d N_1}{\text{mean of } S = \lambda_d N_1} \tag{109}$$

This equation means variance to mean of count rate which is one and $Y = 0$ for Poisson process. This equation is transformed to:

$$Y = \lambda_d\,\frac{\overline{N_1^2} - (\overline{N_1})^2}{\overline{N_1}} - 1 = \lambda_d\,\frac{\overline{N_1^2} - (\overline{N_1})^2 - \overline{N_1}/\lambda_d}{\overline{N_1}} \tag{110}$$

which is close to self-covariance to mean of N_1 but not same shown in the Appendix e). On the other hand, the hypothesis $\dfrac{\mu_{ZZ}}{\overline{Z_1}} = Y$ is written as:

$$Y = \frac{\mu_{ZZ}}{\overline{Z_1}} = \frac{2\,\lambda_d\,\mu_{XZ}\,t}{\lambda_d \overline{N_1} t} = \frac{2\,\mu_{XZ}}{\overline{N_1}} \tag{111}$$

Therefore, it is necessary to prove the following equation:

$$\mu_{XZ} = \frac{\lambda_d}{2}\left[\overline{N_1^2} - (\overline{N_1})^2 - \overline{N_1}/\lambda_d\right] \tag{112}$$

The proof is given as:

$$\overline{N_1^2} - (\overline{N_1})^2 - \overline{N_1}/\lambda_d = \frac{Y\overline{N_1}}{\lambda_d} = \frac{YS}{\lambda_d^2} \tag{113}$$

$$\left[\overline{N_1^2} - (\overline{N_1})^2 - \overline{N_1}/\lambda_d\right]\left(\frac{\omega_1\omega_2}{\lambda_2}\right)^2 = YS\left(\frac{\omega_1\omega_2}{\lambda_d\lambda_2}\right)^2 = YS\left(\frac{1}{\epsilon_n M_L}\right)^2 \tag{114}$$

$$YS\left(\frac{1}{\epsilon_n M_L}\right)^2 = \frac{2D}{f_d}\left(\frac{1}{\epsilon_n M_L}\right)^2 = \lambda_{1f}\overline{\nu_{i2}^1}\,\overline{N_1} + \lambda_{2f}\overline{\nu_{i2}^2}\,\overline{N_2} + S_1\overline{\nu_{s2}} \tag{115}$$

$$\mu_{XZ} = \frac{\lambda_d\,\lambda_2^2}{2\,\omega_1^2\,\omega_2^2}\left(\lambda_{1f}\overline{\nu_{i2}^1}\,\overline{N_1} + \lambda_{2f}\overline{\nu_{i2}^2}\,\overline{N_2} + S_1\overline{\nu_{s2}}\right) \quad \text{from equation (58)} \tag{116}$$

$$\therefore\ \mu_{XZ} = \frac{\lambda_d}{2}\left[\overline{N_1^2} - (\overline{N_1})^2 - \overline{N_1}/\lambda_d\right] \tag{117}$$

Equations (107) and (108) are simplified using $S = \lambda_d N_1$ as:

$$\epsilon_1 = \frac{S}{\overline{\overline{F_1}}} = \alpha_1\,\tau_1\,\epsilon_n\,M_L \tag{118}$$

$$\epsilon_2 = \frac{S}{\overline{\overline{F_2}}} = \alpha_2\,\tau_2\,\epsilon_n\,M_L \tag{119}$$

3.9 Evaluating decay constants by fast neutron and by thermal neutron

The decay constants by fast neutron α_1 and by thermal neutron α_2 are used as a form of product $\alpha_1\tau_1$ and $\alpha_2\tau_2$. However, Rossi-alpha combined distribution observed by DDSI has two decay components: 1) decay by a detector's moderator and by a sample with short time constant results from fast induced fissions; 2) decay by a detector's moderator and by thermalization inside/outside a sample with long time constant results from thermal induced fissions. Except for the effect of detector's moderator, the former reflects α_1 and the latter reflects α_2. The decay constants α_1 and α_2 are given by:

$$\alpha_1 = r\left(1 + \frac{S_1}{\overline{\overline{F_1}}}\right) \quad \because\ \tau_1 = \frac{1}{S_1 + \overline{\overline{F_1}}} \tag{120}$$

$$\alpha_2 = r \quad \because\ \tau_2 = \frac{1}{\overline{\overline{F_2}}} \tag{121}$$

$$\frac{1}{\alpha_2} = \frac{1}{\alpha_1}\left(1 + \frac{S_1}{\overline{\overline{F_1}}}\right) > \frac{1}{\alpha_1} \tag{122}$$

The time constant $1/\alpha_2$ results from thermal induced fissions is longer than the time constant $1/\alpha_1$ results from fast induced fissions. In the last subsection, we will find that the ratio of $1/\alpha_2$ to $1/\alpha_1$ varies from several to ten.

3.10 Procedure of the evaluation and calculation of doubles count rates by fast neutron and by thermal neutron

Procedure of evaluation:

1. Determine detector efficiency ϵ_n (counts/neutron) used for conventional NDAs and safeguards and doubles gate fraction f_d;
2. Select representative isotopes (ex. ^{238}U for fast induced fission, ^{235}U for thermal induced fission and ^{244}Cm for spontaneous fission) or a linear coupling of isotopes (ex. weighed ^{235}U, ^{239}Pu and ^{241}Pu for thermal induced fission);
3. Determine $\overline{\nu_{s1}}, \overline{\nu_{s2}}, \overline{\nu_{i1}^1}, \overline{\nu_{i2}^1}, \overline{\nu_{i1}^2}, \overline{\nu_{i2}^2}$;
4. Measure S, D_1/D_2 and M_L;
5. Evaluate $S_1 = S/(\overline{\nu_{s1}}\,\epsilon_n M_L)$ from equation (75);
6. Evaluate $\overline{F_1}$ and $\overline{F_2}$ from equations (101) and (102);
7. Confirm $\overline{F_1}$ and $\overline{F_2}$ satisfy equation (100);
8. Evaluate $1/(\alpha_1\tau_1)$ and $1/(\alpha_2\tau_2)$ from equations (90) and (91);
9. Evaluate Y_1 and Y_2 from equations (95) and (96);
10. Evaluate $1/\alpha_1$ and $1/\alpha_2$ from equations (120) and (121).

Procedure of D_1, D_2 calculation for confirmation:

1. Calculate doubles count rate by fast neutron D_1/f_d:

$$\frac{D_1}{f_d} = \frac{1}{2}\,\epsilon_n^2 M_L^2\,(\overline{F_1}\,\overline{\nu_{i2}^1} + S_1\overline{\nu_{s2}}) = \frac{S\,Y_1}{2} \tag{123}$$

2. Calculate doubles count rate by thermal neutron D_2/f_d:

$$\frac{D_2}{f_d} = \frac{1}{2}\,\epsilon_n^2 M_L^2\,\overline{F_2}\,\overline{\nu_{i2}^2} = \frac{S\,Y_2}{2} \tag{124}$$

3. Measure D and confirm $D/f_d = D_1/f_d + D_2/f_d$.

From the view point of safeguards, $\overline{F_1}$ and $\overline{F_2}$ are important because they are proportional to fissile weight. D_1 and D_2 are not so important though D is proportional to the weight of spontaneous fission isotopes in conventional NDAs used in the air. However, these procedures and calculations are absolutely basic, and it is necessary to compare the calculation to the results of DDSI measurement and/or simulations including, for example, effect of neutron absorbing elements such as ^{10}B.

3.11 Provisional calculations for UO_2 of 1~10 kgU containing small ^{244}Cm

U-Pu-Cm mixed oxide in water is supposed for provisional calculations because it will have to be measured and verified in the near future in Japan. According to the JAEA-Data/Code 2012-018[23], weight of ^{244}Cm, the strongest spontaneous fission nucleus: source neutron provider, was estimated to be 8.9 mg for 1 kgU (617 g ^{244}Cm/core and 69 tU/core). Spontaneous/fast induced/thermal induced fission nuclei are listed in Table 5-1～5-3. Weighed mean of spontaneous or fast induced fission neutron energy is 1.9～2.1 MeV and (n,f) cross-section for this neutron is 0.5～2 barns regardless of the isotopes.

Table 5-1 Spontaneous fission nuclei

Isotope	Fission yield [$s^{-1}g^{-1}$]	$\overline{\nu_{s1}}$	Neutron yield [$s^{-1}g^{-1}$]	Mass in core (ref.23) [g]	Relative strength
^{238}U	6.78E-3	1.99	1.35E-2	6.53E+7	0.01
^{238}Pu	1.17E+3	2.19	2.56E+3	7.44E+3	0.27
^{240}Pu	4.83E+2	2.15	1.04E+3	1.05E+5	1.54
^{242}Pu	8.07E+2	2.15	1.74E+3	2.02E+4	0.50
^{242}Cm	7.81E+6	2.54	1.98E+7	1.49E-1	0.04
^{244}Cm	4.11E+6	2.72	1.12E+7	6.17E+2	97.61
^{246}Cm	2.97E+6	2.93	8.70E+6	~ 2.54E-1†	0.03

† Estimated from ^{242}Cm/^{246}Cm ratio obtained by a burnup code due to lack of this value in ref.23.

Table 5-2 Fast induced fission nuclei

Isotope	at 2 MeV			Mass in core (ref.23) [g]	Relative strength
	Cross-section [barn]	$\overline{\nu_{i1}^{1}}$	Neutron yield [rel.]		
^{238}U	0.534E+0	2.59	1.38E+0	6.53E+7	92.64
^{235}U	1.29E+0	2.64	3.41E+0	1.11E+6	3.88
^{239}Pu	1.98E+0	3.16	6.26E+0	3.08E+5	1.98
^{236}U	0.819E+0	~2.6	2.13E+0	2.42E+5	0.53
^{240}Pu	1.75E+0	~3.2	5.60E+0	1.05E+5	0.60
^{241}Pu	1.68E+0	3.20	5.38E+0	3.62E+4	0.20
^{241}Am	1.89E+0	3.40	6.43E+0	2.65E+4	0.18

Table 5-3 Thermal induced fission nuclei

Isotope	at thermal			Mass in core (ref.23) [g]	Relative strength
	Cross-section [barn]	$\overline{\nu_{i1}^{2}}$	Neutron yield [rel.]		
^{235}U	5.85E+2	2.41	1.41E+3	1.11E+6	67.00
^{237}Np†	2.04E-2	2.63	5.37E-2	2.04E+4	0.00
^{238}Pu	1.70E+1	~2.85	4.85E+1	7.44E+3	0.02
^{239}Pu	7.48E+2	2.88	2.15E+3	3.08E+5	28.41
^{241}Pu	1.01E+3	2.92	2.95E+3	3.62E+4	4.57
^{241}Am†	3.14E+0	3.11	9.77E+0	2.65E+4	0.01

† For thermal neutron, ^{237}Np and ^{241}Am have (n,γ) cross-sections of 175 barns and 684 barns, respectively. Daughter nuclei ^{238}Np and ^{242m}Am have large (n,f) cross-sections of 2200 barns and 6400 barns, respectively, which are not included.

From these tables, it is almost clear that ^{244}Cm and ^{238}U are representatives of spontaneous fission and fast induced fission, respectively. For a representative of thermal induced fission, it is better to define an effective composition like conventional ^{240}Pu effective composition. However, ^{235}U is selected as a representative to simplify the provisional calculations in this study. The first and the second factorial moments of the representatives are shown in Table 6. These values were calculated from the distributions p_ν, f_ν^1, f_ν^2 consolidated/proposed by Zucker and Holden [12, 13, 14]:

Table 6 The first and the second factorial moments[h]

Spontaneous: ^{244}Cm		Fast induced: ^{238}U		Thermal induced: ^{235}U	
$\overline{\nu_{s1}}$	2.721	$\overline{\nu_{i1}^1}$	2.583	$\overline{\nu_{i1}^2}$	2.414
$\overline{\nu_{s2}}$	5.941	$\overline{\nu_{i2}^1}$	5.494	$\overline{\nu_{i2}^2}$	4.635

Also, S_1 and r are given as 3.66E+4 fissions/s and 9.96E+4 /s for 1 kgU containing 8.9 mg ^{244}Cm, respectively. Instead of measurement, D_1/D_2 is given in a stepwise manner from 2 to 200, and M_L is given to satisfy a rule that the number of source neutrons estimated inversely from $\overline{F_1}$, $\overline{F_2}$, cross-sections (fast and thermal), density of isotopes in oxide and sample thickness is equal to $r = S_1 \overline{\nu_{s1}}$. Here, $\overline{F_1}$ and $\overline{F_2}$ are calculated from M_L using equations (101) and (102), so the inverse estimation is done by setting a tentative M_L then seek a convergence (for example, by goal seek function in Excel®). The inversely estimated number of source neutrons is given as:

$$c = c_1 + c_2 = \frac{\overline{F_1}}{\sigma_1\, n_1\, x} + \frac{\overline{F_2}}{\sigma_2\, n_2\, x} \tag{125}$$

where c is inversely estimated number of source neutrons per unit time, n is density of isotopes in oxide, x is thickness of a sample, σ is cross-section and subscript 1 and 2 means fast and thermal, respectively.

For n_1 and n_2, ^{238}U/U and ^{235}U/U are approximated to be 0.96 and 0.04 (from Table 5-2), and UO_2 density is 10.96 g cm^{-3} (natural U) given by a handbook of nuclear criticality safety[24], so n_1 and n_2 are 2.346×10^{22} cm^{-3} and 0.098×10^{22} cm^{-3}, respectively. For x, UO_2 volume containing 1 kgU is 103.5 cm^3 and x is 4.695 cm supposing a cubic. For the cross-section, σ_1 and σ_2 are 0.534 barns and 585 barns given by the ENDF web site[25] of National Nuclear Data Center.

h. An example of applying the concept of effective composition to estimate effective 1st/2nd moments:

Spontaneous: ^{244}Cm		Fast induced: ^{238}U and ^{235}U		Thermal induced: ^{235}U, ^{239}Pu, ^{241}Pu	
$\overline{\nu_{s1}}$	2.721	$\overline{\nu_{i1}^1}$	2.588	$\overline{\nu_{i1}^2}$	2.601
$\overline{\nu_{s2}}$	5.941	$\overline{\nu_{i2}^1}$	5.506	$\overline{\nu_{i2}^2}$	5.489

Results of UO_2 containing 1, 2, 5 and 10 kgU are shown in Table 7-1～7-4, assuming ϵ_n is 0.2 and changing D_1/D_2 from 2 to 200. Density of r is 962 s^{-1} cm^{-3} is same for all cases:

Table 7-1 UO_2 containing 1 kgU (103.5 cm^3, 8.9 mg ^{244}Cm)

D_1/D_2	–	200	100	50	20	10	5	2
S_1	[1/s]	36600 (= 4.11E+6 $s^{-1}g^{-1}$ × 8.9 mg)						
r	[1/s]	99589						
M_L	–	1.1583	1.1647	1.1774	1.2156	1.2791	1.4056	1.7812
$\overline{F_1}$	[1/s]	5852	5846	5834	5799	5740	5623	5275
$\overline{F_2}$	[1/s]	269	538	1077	2689	5372	10716	26583
$\overline{F_1}\,\overline{\nu_1^1}$	[1/s]	15115	15100	15069	14978	14826	14524	13626
$\overline{F_2}\,\overline{\nu_1^2}$	[1/s]	650	1300	2599	6492	12967	25867	64171
$\overline{F_1}\,\overline{\nu_1^1}+\overline{F_2}\,\overline{\nu_1^2}$	[1/s]	15765	16400	17668	21470	27794	40391	77797
$\lambda_1\,\overline{N_1}$	[1/s]	115354	115988	117257	121059	127382	139980	177386
$1/(\alpha_1\,\tau_1)$	–	0.0588	0.0587	0.0586	0.0582	0.0576	0.0565	0.0530
$1/(\alpha_2\,\tau_2)$	–	0.0027	0.0054	0.0108	0.0270	0.0539	0.1076	0.2669
Y_1	–	0.581	0.584	0.590	0.609	0.640	0.701	0.881
Y_2	–	0.003	0.006	0.012	0.030	0.064	0.140	0.441
$1/\alpha_1$	[s]	1.38E-06	1.38E-06	1.38E-06	1.37E-06	1.36E-06	1.34E-06	1.26E-06
$1/\alpha_2$	[s]	1.00E-05	1.00E-05	1.00E-05	1.00E-05	1.00E-05	1.00E-05	1.00E-05
α_1/α_2	–	7.255	7.261	7.273	7.312	7.376	7.509	7.938

Table 7-2 UO_2 containing 2 kgU (207.0 cm^3, 17.8 mg ^{244}Cm)

D_1/D_2	–	200	100	50	20	10	5	2
S_1	[1/s]	73200						
r	[1/s]	199177						
M_L	–	1.1980	1.2046	1.2178	1.2572	1.3228	1.4536	1.8418
$\overline{F_1}$	[1/s]	14748	14736	14712	14639	14517	14275	13557
$\overline{F_2}$	[1/s]	557	1113	2225	5559	11103	22149	54947
$\overline{F_1}\,\overline{\nu_1^1}$	[1/s]	38095	38063	38000	37811	37498	36873	35017
$\overline{F_2}\,\overline{\nu_1^2}$	[1/s]	1343	2687	5372	13419	26803	53468	132643
$\overline{F_1}\,\overline{\nu_1^1}+\overline{F_2}\,\overline{\nu_1^2}$	[1/s]	39438	40750	43372	51231	64301	90341	167659
$\lambda_1\,\overline{N_1}$	[1/s]	238615	239927	242549	250408	263478	289518	366837
$1/(\alpha_1\,\tau_1)$	–	0.0740	0.0740	0.0739	0.0735	0.0729	0.0717	0.0681
$1/(\alpha_2\,\tau_2)$	–	0.0028	0.0056	0.0112	0.0279	0.0557	0.1112	0.2759
Y_1	–	0.621	0.624	0.631	0.651	0.684	0.749	0.942
Y_2	–	0.003	0.006	0.013	0.033	0.068	0.150	0.471
$1/\alpha_1$	[s]	8.42E-07	8.41E-07	8.40E-07	8.37E-07	8.31E-07	8.19E-07	7.85E-07
$1/\alpha_2$	[s]	5.02E-06	5.02E-06	5.02E-06	5.02E-06	5.02E-06	5.02E-06	5.02E-06
α_1/α_2	–	5.963	5.967	5.976	6.000	6.042	6.128	6.400

Table 7-3 UO_2 containing 5 kgU (517.5 cm^3, 44.5 mg ^{244}Cm)

D_1/D_2	–	200	100	50	20	10	5	2
S_1	[1/s]	183000						
r	[1/s]	497943						
M_L	–	1.2668	1.2737	1.2876	1.3293	1.3986	1.5367	1.9466
$\overline{F_1}$	[1/s]	50050	50018	49954	49761	49440	48801	46904
$\overline{F_2}$	[1/s]	1469	2939	5876	14677	29317	58482	145080
$\overline{F_1}\,\overline{\nu_1^1}$	[1/s]	129279	129196	129030	128532	127704	126053	121153
$\overline{F_2}\,\overline{\nu_1^2}$	[1/s]	3547	7094	14183	35431	70770	141175	350224
$\overline{F_1}\,\overline{\nu_1^1}+\overline{F_2}\,\overline{\nu_1^2}$	[1/s]	132827	136290	143214	163963	198474	267228	471376
$\lambda_1\,\overline{N_1}$	[1/s]	630770	634233	641157	661906	696417	765171	969319
$1/(\alpha_1\,\tau_1)$	–	0.1005	0.1004	0.1003	0.0999	0.0993	0.0980	0.0942
$1/(\alpha_2\,\tau_2)$	–	0.0030	0.0059	0.0118	0.0295	0.0589	0.1174	0.2914
Y_1	–	0.693	0.697	0.704	0.726	0.763	0.837	1.052
Y_2	–	0.003	0.007	0.014	0.036	0.076	0.167	0.526
$1/\alpha_1$	[s]	4.31E-07	4.31E-07	4.31E-07	4.29E-07	4.27E-07	4.23E-07	4.10E-07
$1/\alpha_2$	[s]	2.01E-06	2.01E-06	2.01E-06	2.01E-06	2.01E-06	2.01E-06	2.01E-06
α_1/α_2	–	4.656	4.659	4.663	4.678	4.701	4.750	4.902

Table 7-4 UO_2 containing 10 kgU (1035.1 cm^3, 89 mg ^{244}Cm)

D_1/D_2	–	200	100	50	20	10	5	2
S_1	[1/s]	366000						
r	[1/s]	995886						
M_L	–	1.3346	1.3420	1.3566	1.4005	1.4734	1.6187	2.0502
$\overline{F_1}$	[1/s]	126132	126064	125929	125523	124848	123503	119509
$\overline{F_2}$	[1/s]	3093	6186	12368	30896	61711	123104	305393
$\overline{F_1}\,\overline{\nu_1^1}$	[1/s]	325798	325623	325273	324225	322481	319007	308692
$\overline{F_2}\,\overline{\nu_1^2}$	[1/s]	7467	14932	29856	74582	148971	297172	737218
$\overline{F_1}\,\overline{\nu_1^1}+\overline{F_2}\,\overline{\nu_1^2}$	[1/s]	333265	340555	355129	398807	471452	616179	1045909
$\lambda_1\,\overline{N_1}$	[1/s]	1329151	1336441	1351015	1394693	1467338	1612065	2041795
$1/(\alpha_1\,\tau_1)$	–	0.1267	0.1266	0.1264	0.1260	0.1254	0.1240	0.1200
$1/(\alpha_2\,\tau_2)$	–	0.0031	0.0062	0.0124	0.0310	0.0620	0.1236	0.3067
Y_1	–	0.769	0.773	0.781	0.806	0.846	0.927	1.166
Y_2	–	0.004	0.008	0.016	0.040	0.085	0.185	0.583
$1/\alpha_1$	[s]	2.57E-07	2.57E-07	2.57E-07	2.56E-07	2.55E-07	2.53E-07	2.47E-07
$1/\alpha_2$	[s]	1.00E-06	1.00E-06	1.00E-06	1.00E-06	1.00E-06	1.00E-06	1.00E-06
α_1/α_2	–	3.902	3.903	3.906	3.916	3.932	3.964	4.063

The time constant $1/\alpha_2$ is several to ten times longer than $1/\alpha_1$, which is consistent to Rossi-alpha combined distribution though affected by the moderator of detector.

Regarding the time constant $1/\alpha_2$, the time constant and the ratio α_1/α_2 become larger when a sample becomes smaller, which is important to apply fitting curves independently to Rossi-alpha combined distribution.

At last, calculated count rates are shown in Table 8-1~8-4 and Figure 8-1~8-2, assuming f_d (doubles gate fraction) is 0.68 (both for D_1 gate and for D_2 gate):

Table 8-1 UO_2 containing 1 kgU (8.9 mg ^{244}Cm)

D_1/D_2	–	200	100	50	20	10	5	2
S	[1/s]	23071	23198	23451	24212	25476	27996	35477
D_1	[1/s]	4554.2	4603.8	4703.9	5009.9	5539.8	6672.4	10632.6
D_2	[1/s]	22.8	46.0	94.1	250.5	554.0	1334.5	5316.3
D_1/D_2 result	–	200.0	100.0	50.0	20.0	10.0	5.0	2.0
D	[1/s]	4577	4650	4798	5260	6094	8007	15949
Y	–	0.583	0.590	0.602	0.639	0.704	0.841	1.322

Table 8-2 UO_2 containing 2 kgU (17.8 mg ^{244}Cm)

D_1/D_2	–	200	100	50	20	10	5	2
S	[1/s]	47723	47985	48510	50082	52696	57904	73367
D_1	[1/s]	10070.0	10179.7	10400.7	11076.9	12247.6	14749.9	23498.0
D_2	[1/s]	50.3	101.8	208.0	553.8	1224.8	2950.0	11749.0
D_1/D_2 result	–	200.0	100.0	50.0	20.0	10.0	5.0	2.0
D	[1/s]	10120	10281	10609	11631	13472	17700	35247
Y	–	0.624	0.630	0.643	0.683	0.752	0.899	1.413

Table 8-3 UO_2 containing 5 kgU (44.5 mg ^{244}Cm)

D_1/D_2	–	200	100	50	20	10	5	2
S	[1/s]	126154	126847	128231	132381	139283	153034	193864
D_1	[1/s]	29727.3	30050.7	30702.4	32696.3	36147.8	43524.9	69310.8
D_2	[1/s]	148.6	300.5	614.0	1634.8	3614.8	8705.0	34655.4
D_1/D_2 result	–	200.0	100.0	50.0	20.0	10.0	5.0	2.0
D	[1/s]	29876	30351	31316	34331	39763	52230	103966
Y	–	0.697	0.704	0.718	0.763	0.840	1.004	1.577

Table 8-4 UO_2 containing 10 kgU (89 mg ^{244}Cm)

D_1/D_2	–	200	100	50	20	10	5	2
S	[1/s]	265830	267288	270203	278939	293468	322413	408359
D_1	[1/s]	69462.9	70217.8	71739.0	76393.1	84449.0	101666.1	161838.6
D_2	[1/s]	347.3	702.2	1434.8	3819.7	8444.9	20333.2	80919.3
D_1/D_2 result	–	200.0	100.0	50.0	20.0	10.0	5.0	2.0
D	[1/s]	69810	70920	73174	80213	92894	121999	242758
Y	–	0.772	0.780	0.797	0.846	0.931	1.113	1.748

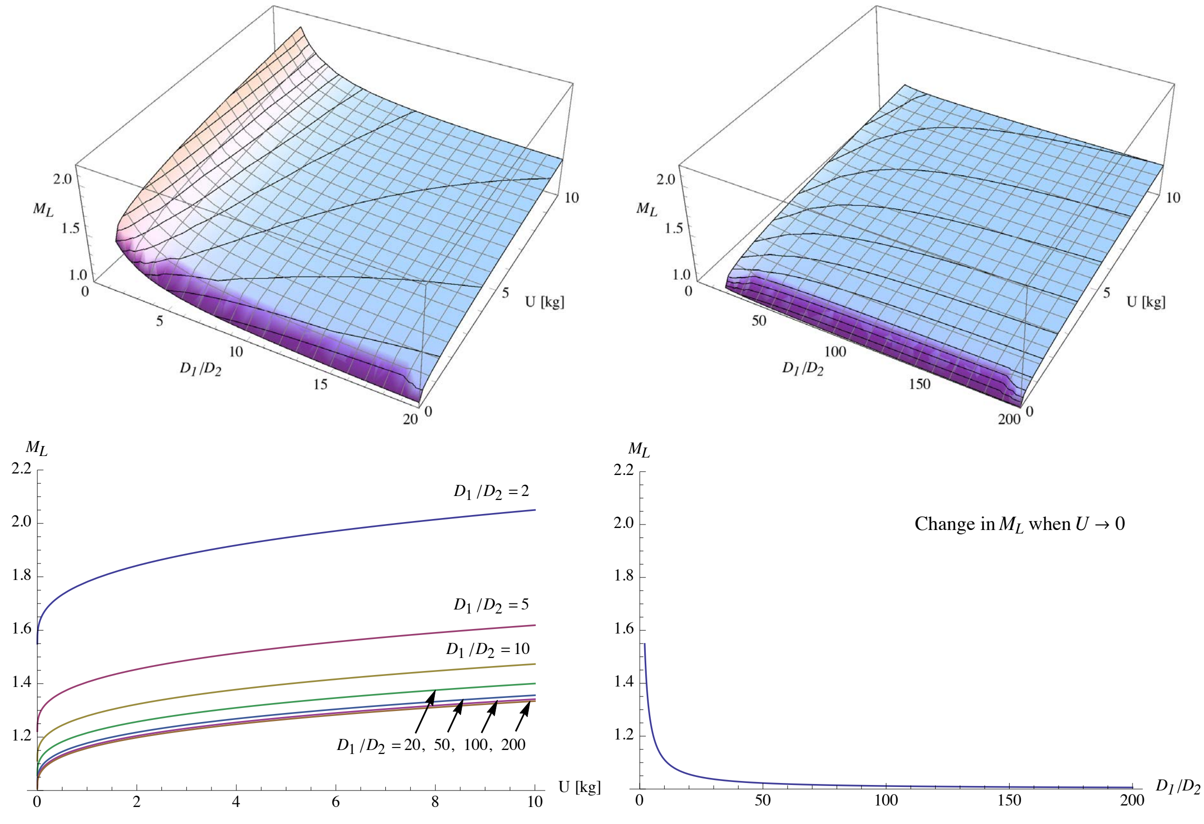


Figure 8-1 Change in M_L in response to D_1/D_2 and U weight

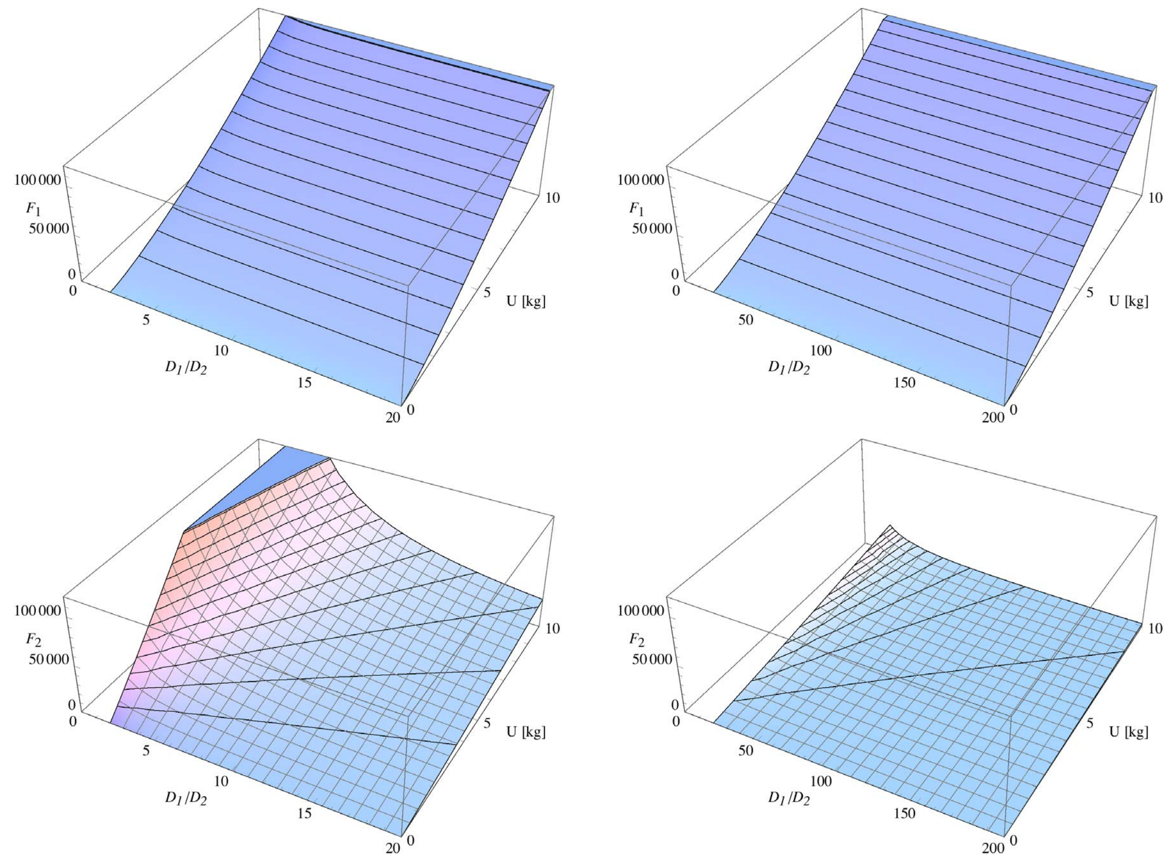


Figure 8-2 Changes in $\overline{F_1}$ and $\overline{F_2}$ in response to D_1/D_2 and U weight

As a result of provisional calculation, it was confirmed that no inconsistency in equations nor outlier in calculations exists. Changes in M_L, $\overline{F_1}$ and $\overline{F_2}$ are shown in Figure 8 in response to D_1/D_2 and U weight that is proportional to ^{244}Cm weight (as a neutron source). There is a trend that M_L and $\overline{F_2}$ change at small D_1/D_2, affected strongly by an astonishing number of thermal fission neutrons, which is not normal. Except for the area, the changes are almost moderate. On the other hand, $\overline{F_1}$ is almost flat because r is proportional to U or ^{244}Cm weight and $1/(\alpha_1\,\tau_1)$ is almost independent from D_1/D_2 shown in Table 7-1~7-4.

From the view point of materials accountancy and safeguards, ^{235}U, ^{239}Pu and ^{241}Pu are especially focused on, however $\overline{F_2}$ obtained by the procedure in 3.10 is a linear coupling of ^{235}U, ^{239}Pu and ^{241}Pu contributions. Each contribution is proportional to the product of cross-section and atomic-density because neutron flux and sample volume/thickness are common. Effect of absorption resulting from (n,γ) reaction by ^{238}U, (n,α) reaction by ^{10}B and so on are also coupled linearly but negatively to the contributions, i.e.:

$$\overline{F_2} \;\propto\; \left[\sigma^{\text{thm.}}_{(\text{n,f})}\frac{W}{M}\right]^{^{235}\text{U}} + \left[\sigma^{\text{thm.}}_{(\text{n,f})}\frac{W}{M}\right]^{^{239}\text{Pu}} + \left[\sigma^{\text{thm.}}_{(\text{n,f})}\frac{W}{M}\right]^{^{241}\text{Pu}} - \left[\sigma^{\text{thm.}}_{(\text{n},\gamma)}\frac{W}{M}\right]^{^{238}\text{U}} - \left[\sigma^{\text{thm.}}_{(\text{n},\alpha)}\frac{W}{M}\right]^{^{10}\text{B}} \tag{126}$$

where W is weight, M is molar mass and 'thm.' is thermal neutron energy (0.0253 eV). Equation (126) has minimum terms, thus examinations for absorption by other isotopes (ex. ^{240}Pu, ^{242}Pu) with reference to Appendix g) would be necessary. The proportionality factor of equation (126) consists of the number of source neutrons per unit time, sample density and thickness, which is similar to equation (125). Anyway, $\overline{F_2}$ is focused on for safeguards application instead of D_2.

In addition, it should be noted that neutrons having energy of scattering region (between fast and thermal) have not been considered in this study, because energy loss of neutron in a sample is small (from mean free path of fast neutron and sample size) until it leaks to water, which results in almost of all neutrons are either fast or thermal. Another point to remember is that non-uniformity of thermal neutrons in a sample (from mean free path of thermal neutron and sample size) has not been considered. This point will have to be investigated in detail because thermal neutrons moderated in water do not invade deeply into a sample. The solution would be to limit sample thickness around one centimeter (almost same as a pellet). In this case, an appropriate sample size would be 103.5 cm^2 × 1 cm for UO_2 containing 1 kg U. As previously mentioned, the time constant $1/\alpha_2$ and the ratio α_1/α_2 becomes larger when a sample becomes smaller, which makes easy to fit curves independently to Rossi-alpha combined distribution. So, it would be reasonable that a large sample is divided into several pieces to satisfy ~1 cm thickness. The last point to remember is that the number of neutrons from (α,n) reactions correspond to ^{244}Cm spontaneous fission is estimated[26] to be 706 s^{-1} for 1 kgU (8.9 mg ^{244}Cm), which is <1% of $r = S_1\,\overline{\nu_{s1}}$.

4. Conclusions

This study belongs to the theory of neutron branching process and fluctuations. The well-known applications of the theory were reactor noise analysis or subcriticality assay so far. This time, it was confirmed that solving the theory using probability generating function was effective and available to non destructive assay practically realized in the near future for safeguards.

The first example is to derive formulae of multiplicity distribution up to septuplet. Its principle was reported by K. Böhnel in 1985, but such a high-order expansion was the first case due to its increasing complexity. In this study, basic characteristics of the high-order correlation was investigated and it was found that high-order correlations increase rapidly, cross and leave lower-order correlations behind, when leakage multiplication is > 1.3 which depends on detector efficiency and counter setting. It is sure that counting technique for high-order correlations has not been established because of large uncertainty of counting statistics due to thermalization and small leakage multiplication of a sample. However, it could be possible if fast neutron detecting/counting technique could be established without moderator and a sample could have larger leakage multiplication within criticality control. Therefore, the first example is a strategic move in the future.

The second example is to derive formulae for fission rates and doubles count rates by fast neutron and by thermal neutron in their coexisting system. Its principle was reported by I. Pázsit and L. Pál in 2012, but such a physical interpretation, i.e. associating their stochastic variables with practical doubles count rate and leakage multiplication, is the first case. In this study, it was found that from Rossi-alpha combined distribution and measured ratio of each area obtained by Differential Die-Away Self-Interrogation (DDSI) and conventional assay data, it is possible to estimate: the number of induced fissions per unit time by fast neutron and by thermal neutron; the number of induced fissions (< 1) by one source neutron; and individual doubles count rates. During the research, a hypothesis introduced in their report was proved to be true. Provisional calculations were done for UO_2 of 1~10 kgU containing ~ 0.009 wt% ^{244}Cm. It should be noted that the fission rate by thermal neutron $\overline{F_2}$ is focused on instead of doubles count rate D_2 for safeguards application.

Acknowledgements

I would like to appreciate what all staffs of Technology Development and Promotion Section, Nuclear Material Control Section and Conversion Technology Section have done to complete the report. Also, I would like to appreciate communications with Prof. Imre Pázsit regarding the Section 3.

References

1 TANAKA H., NAKAMURA H. and HOSOMA T., Design of Geometrical Detector Arrangement for Extensive Holdup Measurement, IAEA-SM-367/14/08, Proc. IAEA Symposium 2001.

2 NAKAMURA H., TANAKA H., HOSOMA T., ICHIGE K., TAKAHASHI Y. and TANAKA I., Instant Measurement of Plutonium Amount in a Bag-Out Solid Waste, 22, Proc. of INMM Japan Chapter Annual Meeting, 2001 (in Japanese).

3 NAKAMURA H., BEDDINGFIELD D.H., MONTOYA J.S., NAKAMICHI H., MUKAI Y. and KURITA T., Implementation of Dynamic Cross-Talk Correction (DCTC) for MOX Holdup Assay Measurements Among Multiple Gloveboxes, a136_1, Proc. INMM 2012.

4 Passive Nondestructive Assay of Nuclear Materials, REILLY D., ENSSLIN N. and SMITH, Jr. H. Ed., NUREG/CR-5550, LA-UR-90-732, 1991, 700p.

5 Passive Nondestructive Assay of Nuclear Materials 2007 Addendum, REILLY D. Ed., 2007.

6 PÁZSIT I. and L. Pál, Neutron Fluctuations, Elsevier, 2008, 340p.

7 HOSOMA T., Essentials of Neutron Multiplicity Counting Mathematics − An example of U-Pu Mixed Dioxide −, JAEA-Research 2015-009 (in Japanese), 162p.

8 HOFFMANN F., Chapter 9 of Statistical Aspects of Pile Theory, The Science and Engineering of Nuclear Power Volume II, Addison-Wesley Press, Inc., 1949, pp.103-119.

9 HAGE W. and CIFARELLI D.M., On the Factorial Moments of the Neutron Multiplicity Distribution of Fission Cascades, Nuclear Instruments and Methods in Physics Research A 236, 1985, pp.165-177.

10 CROFT S., FAVALLI A., HAUCK D.K., HENZLOVA D. and SANTI P.A., Feynman variance-to-mean in the context of passive neutron coincidence counting, Nuclear Instruments and Methods in Physics Research A 686, 2012, pp.136-144.

11 BÖHNEL K., The Effect of Multiplication on the Quantitative Determination of Spontaneously Fissioning Isotopes by Neutron Correlation Analysis, Nuclear Science and Engineering 90, 1985, pp.75-82.

12 HOLDEN N.E. and ZUCKER M.S., A Reevaluation of the Average Prompt Neutron Emission Multiplicity (Nubar) Values from Fission of Uranium and Transuranium Nuclides, BNL-NCS-35513, 1984.

13 HOLDEN N.E. and ZUCKER M.S., Prompt Neutron Multiplicities for the Transplutoniurn Nuclides, BNL-NCS-36379, 1985.

14 ZUCKER M.S. and HOLDEN N.E., Energy Dependence of the Neutron Multiplicity $P\nu$ in Fast Neutron Induced Fission of 235,238U and 239Pu, BNL-38491, 1986.

15 TERRELL J., Distributions of Fission Neutron Numbers, Physical Review 108(3), 1957, pp.783-789.

16 ENSSLIN N., GAVRON A., HARKER W.C., KRICK M.S., LANGNER D.G., MILLER M.C., PICKRELL M.M., Expected Precision for Neutron Multiplicity Assay using Higher Order Moments, LA-UR-97-2716, 1997.

17 CHRTON W.S. and HUNPHREY M.A., JNMM, vol.40, no.3, 2012, pp.12-17.

18 ANDERSON J., PÁL L. and PÁZSIT I., On the Feynman-alpha Formula for Fast Neutrons, Proc. ESARDA 2011.

19 ANDERSON J., PÁL L., PÁZSIT I., CHERNIKOVA D. and POZZI S.A., Derivation and Quantitative Analysis of the Differential Self-interrogation Feynman-alpha Method, Eur. Phys. J. Plus, vol.127, no.21, 2012.

20 ANDERSON J., CHERNIKOVA D., PÁZSIT I., PÁL L. and POZZI S.A., Two-point Theory for the Differential Self-interrogation Feynman-alpha Method, Eur. Phys. J. Plus, vol.127, no.90, 2012.
21 GALLAGER R.G., Chapter 7 of Markov processes with countable-state spaces, Stochastic Processes, Cambridge University Press, 2013, 536p.
22 FEYNMAN R.P., HOFFMANN F. and SERBER R., Dispersion of the Neutron Emission in U-235 Fission, J. Nuclear Energy, vol.3, 1956, pp.64-69.
23 NISHIHARA K., IWAMOTO H. and SUYAMA K., Estimation of Fuel Compositions in Fukushima-Daiichi Nuclear Power Plant, JAEA-Data/Code 2012-018 (in Japanese), 190p.
24 WORKING GROUP ON NUCLEAR CRITICALITY SAFETY DATA, Nuclear Criticality Safety Handbook ver.2, JAERI 1340, 1999 (in Japanese), 189p.
25 NATIONAL NUCLEAR DATA CENTER in Brookhaven National Laboratory, Evaluated Nuclear Data File (ENDF), http://www.nndc.bnl.gov/exfor/endf00.jsp, accessed on October 20, 2016.
26 MATSUNOBU H., OKU T., IIJIMA S., NAITO Y., MASUKAWA F. and NAKASIMA R., Data Book for Calculating Neutron Yields from (α,n) Reaction and Spontaneous Fission, JAERI 1324, 1992 (in Japanese), 260p.

Appendices (Informative)

a) Formulation of coupled pair, accidental pair and accidentals (≠ pair)

Coupled pair, accidental pair and accidentals (≠pair) were illustrated well in Figure a1.

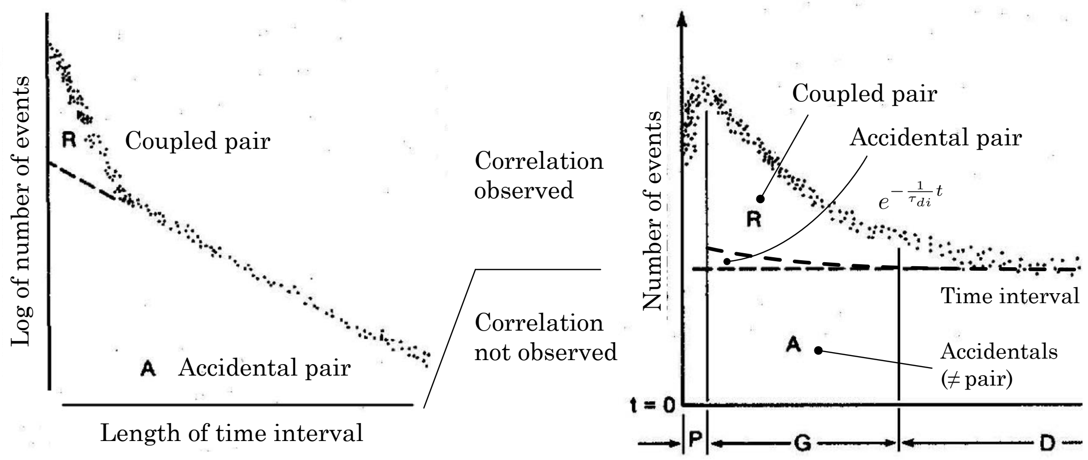


Figure a1 Coupled pair, accidental pair and accidentals (≠ pair) in Rossi-alpha distribution [reprinted from Fig. 16.2 and Fig. 16.3 (break curve added) of LA-UR-90-732] [4]

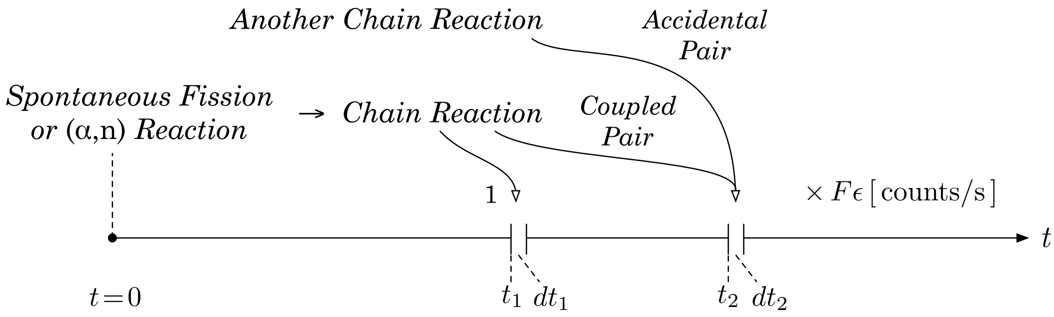
Be sure that both x-axes are not arrival time but interarrival time of renewal process. Coupled pair and accidental pair are formulated by F. Hoffmann in 1949 as:

$$\left\{\begin{array}{l}\text{Expected number of}\\ \text{correlated neutrons}\\ \text{in interval } t\end{array}\right\} = \int_{t_2=0}^{t_2=t}\int_{t_1=0}^{t_1=t_2}\left\{\overbrace{F\epsilon}^{at\,t_1}\times\overbrace{\left[\underbrace{F\epsilon}_{Accidental}+\underbrace{\frac{\epsilon\,\overline{\nu(\nu-1)}}{2\,\alpha\,\tau^2}\,\exp^{-\alpha(t_2-t_1)}}_{Coupled}\right]}^{at\,t_2}\right\}dt_1 dt_2$$

$$= F^2\epsilon^2\int_{t_2=0}^{t_2=t}\left[\,t_1\,\right]_0^{t_2}\,dt_2 + F\epsilon^2\,\frac{\overline{\nu(\nu-1)}}{2\,\alpha\,\tau^2}\int_{t_2=0}^{t_2=t}\exp^{-\alpha t_2}\left[\frac{1}{\alpha}\exp^{\alpha t_1}\right]_0^{t_2}dt_2$$

$$= \frac{F^2\epsilon^2\,t^2}{2} + F\epsilon^2\,\frac{\overline{\nu(\nu-1)}}{2\,\alpha^2\,\tau^2}\left[\,t-\frac{1}{\alpha}\left(1-\exp^{-\alpha t}\right)\right]$$

$$= \frac{F^2\epsilon^2\,t^2}{2} + F\epsilon^2\,\frac{\overline{\nu(\nu-1)}}{2\,\alpha^2\,\tau^2}\,t\left(1-\frac{1-\exp^{-\alpha t}}{\alpha t}\right)$$



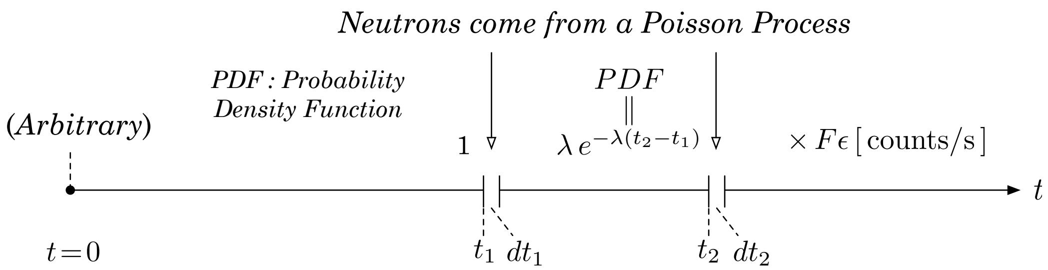
On the other hand, accidentals ($\neq$ pair) is formulated as:

$$\left\{ \begin{array}{l} \text{Expected number of} \\ \text{uncorrelated neutrons} \\ \text{in interval } \; t \end{array} \right\} = \int_{t_2=0}^{t_2=t} \int_{t_1=0}^{t_1=t_2} \left[\overbrace{F\epsilon}^{at\, t_1} \times \overbrace{F\epsilon\, \lambda \exp^{-\lambda(t_2-t_1)}}^{at\, t_2} \right] dt_1 dt_2$$

$$= F^2\epsilon^2 \int_{t_2=0}^{t_2=t} \left(1 - \exp^{-\lambda t_2}\right) dt_2$$

$$= F^2\epsilon^2\, t - F^2\epsilon^2 \frac{1}{\lambda} \left(1 - \exp^{-\lambda t}\right)$$

$$= F^2\epsilon^2\, t \left(1 - \frac{1 - \exp^{-\lambda t}}{\lambda\, t}\right)$$



where F is the number of fissions per unit time, ϵ is efficiency of counter (counts per fission), α is rate of decay regarding prompt neutrons, τ is mean time between fissions and λ is '*rate*' of Poisson process. The number of correlated neutrons $N_c(t)$ and uncorrelated neutrons $N_u(t)$ and their asymptotic terms (Figure a2) are:

$$N_c(t) = \overbrace{\frac{F^2\epsilon^2\, t^2}{2}}^{\text{accidental pair}} \overbrace{+ F\epsilon^2 \frac{\overline{\nu(\nu-1)}}{2\,\alpha^2\,\tau^2}\, t \left(1 - \frac{1 - \exp^{-\alpha t}}{\alpha t}\right)}^{\text{coupled pair}} \tag{a1}$$

$$N_u(t) = F^2\epsilon^2\, t \left(1 - \frac{1 - \exp^{-\lambda t}}{\lambda\, t}\right) \tag{a2}$$

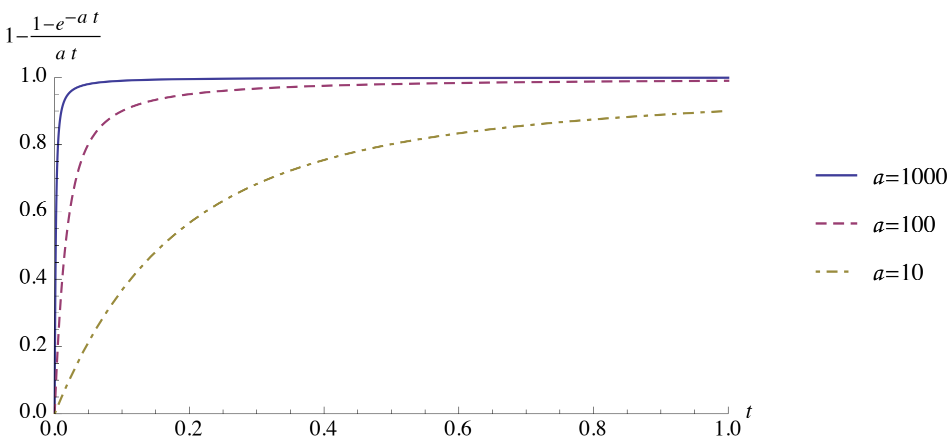


Figure a2 Curve of the asymptotic term of $N_c(t)$ and $N_u(t)$ close to $t=0$.

The first derivatives of coupled pair of correlated neutrons $N_c^{\text{coupled}}(t)$ and uncorrelated neutrons $N_u(t)$ are constants:

$$\frac{d\,N_c^{\text{coupled}}(t)}{dt} = F\epsilon^2\,\frac{\overline{\nu(\nu-1)}}{2\,\alpha^2\,\tau^2} = F\times\frac{\overline{\nu(\nu-1)}}{2}\times\left(\frac{\epsilon}{\alpha\,\tau}\right)^2 \tag{a3}$$

$$= F_p\,m_{\mathit{eff}}\times\overline{\nu_2}\times\left(\frac{\epsilon_n\,p_l\,\overline{\nu}}{\alpha\,\tau}\right)^2 \qquad \because\ \epsilon=\epsilon_n\,p_l\,\overline{\nu}$$

$$= F_p\,m_{\mathit{eff}}\times\overline{\nu_2}\times\epsilon_n^2\times M_L^2 \quad \because\ M_L = p_l M_T = p_l\left(\frac{\overline{\nu}}{\alpha\,\tau}\right)$$

$$= D/f_d$$

$$\frac{dN_u(t)}{dt} = F^2\epsilon^2 = S^2 \text{ or } T^2 \tag{a4}$$

where F_p is the number of spontaneous fissions per unit mass and time, m_{eff} is effective mass of spontaneous fission nuclei, ϵ_n is counting efficiency (counts per neutron), p_l is probability of neutron leak, f_d is characteristic value depend on counter setting, and T is total count rate (not triples). Obtained equations are equal to the conventional ones.

On the other hand, accidental pair of correlated neutrons $N_c^{\text{accidental}}(t)$ is quadratic function of t, where t is given as the gate opening time (gate width) of coincidence counter. The physical meaning is two neutrons arrive within t according to Poisson process. The probability mass function in given below and $e^{-\lambda t}$ is normalization factor. So, the probability of two neutrons arrive within t is given as $(\lambda t)^2/2$ which is same as $N_c^{\text{accidental}}(t)$.

$$P_{\lambda t}(n) = \frac{(\lambda t)^n\,e^{-\lambda t}}{n\,!} \tag{a5}$$

Rossi-alpha distribution has two layers having different time-axis: the first floor has normal arrival time and the second floor has interarrival time in renewal process as shown in Figure a3.

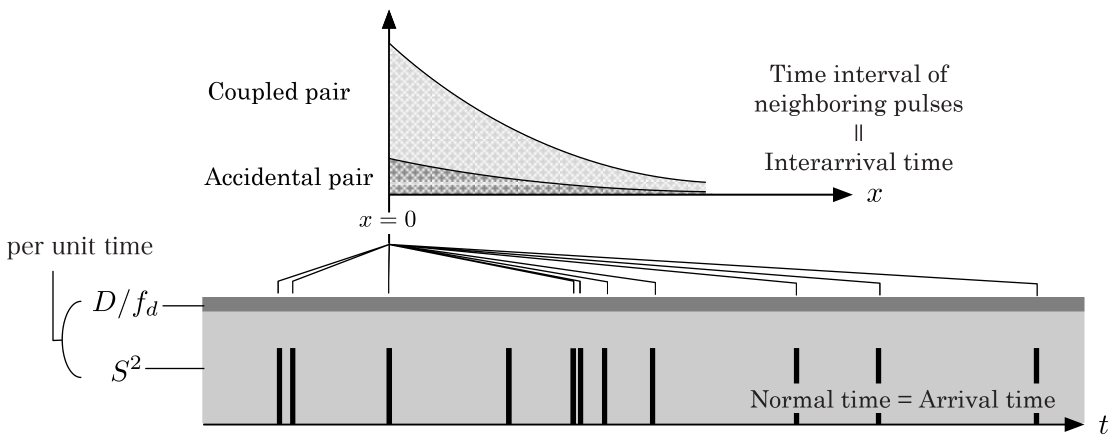


Figure a3 Two layers of Rossi-alpha distribution

b) Definition and derivations of probability generating function

Probability generating function G_X for discrete stochastic variable X (non-negative integer) is defined as a function of continuous variable $|z| \le 1$:

$$G_X(z) \stackrel{\text{def}}{=} \overline{z^X} = \sum_{n=0}^{\infty} z^n P_X(X{=}n) \tag{b1}$$

where P_X is probability mass distribution of X, thus $\sum_{n=0}^{\infty} P_X(X{=}n) = 1$.

The k^{th} derivatives of G_X regarding z are expressed as:

$$\begin{aligned} G_X(z) &= \sum_{n=0}^{\infty} z^n P_X(X{=}n) = P_X(X{=}0) + \sum_{n=1}^{\infty} z^n P_X(X{=}n) \\ G_X'(z) &= \sum_{n=1}^{\infty} n\, z^{n-1} P_X(X{=}n) = 1\, P_X(X{=}1) + \sum_{n=2}^{\infty} n\, z^{n-1} P_X(X{=}n) \\ G_X''(z) &= \sum_{n=2}^{\infty} n(n-1)\, z^{n-2} P_X(X{=}n) = 2\, P_X(X{=}2) + \sum_{n=3}^{\infty} n(n-1)\, z^{n-2} P_X(X{=}n) \end{aligned} \tag{b2}$$

Therefore, P_X is expressed using the k^{th} derivative of G_X at $z=0$ as:

$$G_X^{(k)}|_{z=0} = k!\, P_X(X{=}k) \quad \rightarrow \quad P_X(X{=}k) = \frac{G_X^{(k)}|_{z=0}}{k!} \tag{b3}$$

Also, the k^{th} factorial moment is expressed as the k^{th} derivative of G_X at $z=1$ as:

$$G_X^{(k)}|_{z=1} = \sum_{n=k}^{\infty} n(n-1)\cdots(n-k+1)\, P_X(X{=}n) = \overline{X(X-1)\cdots(X-k+1)} \tag{b4}$$

These two equations are very useful in the context of solving equations. In addition, G_X is expressed using the k^{th} factorial moments. The proof is given by binomial expansion:

$$G_X(z) = \sum_{k=0}^{\infty} \frac{M_{P_X}^{(k)}}{k!} (z-1)^k \qquad \text{where } M_{P_X}^{(k)} \stackrel{\text{def}}{=} G_X^{(k)}|_{z=1} \tag{b5}$$

For independent discrete stochastic variables $X_1, X_2, \cdots X_N$, the composite G_X is given as a product of each G_X as:

$$G_{X_1+X_2+\cdots X_N}(z) = G_{X_1}(z)\, G_{X_2}(z) \cdots G_{X_N}(z) \tag{b6}$$

Especially for $S = X_1 + X_2 + \cdots + X_N, X_i = \{0,1\}$, G_S is given as:

$$G_S(z) = [\,G_X(z)\,]^N = \overline{[\,G_X(z)\,]^N} = G_N\,(G_X(z)) \tag{b7}$$

This equation is also very important and useful in the body.

c) Example of the solving process by *Mathematica®* in the section 2.2 and 2.3

```
z[u_] := (1 - p) u + p y[h[u]]
dh1 = Flatten[Solve[h^(1)[u] == z^(1)[u], h^(1)[u]]];
dh2 = Simplify[Flatten[Solve[h^(2)[u] == z^(2)[u], h^(2)[u]] /. dh1]];
dh3 = Simplify[Flatten[Solve[h^(3)[u] == z^(3)[u], h^(3)[u]] /. dh1 /. dh2]];
dh4 = Simplify[Flatten[Solve[h^(4)[u] == z^(4)[u], h^(4)[u]] /. dh1 /. dh2 /. dh3]];
dh5 = Simplify[Flatten[Solve[h^(5)[u] == z^(5)[u], h^(5)[u]] /. dh1 /. dh2 /. dh3 /. dh4]];
dh6 = Simplify[Flatten[Solve[h^(6)[u] == z^(6)[u], h^(6)[u]] /. dh1 /. dh2 /. dh3 /. dh4 /. dh5]];
dh7 = Simplify[Flatten[Solve[h^(7)[u] == z^(7)[u], h^(7)[u]] /. dh1 /. dh2 /. dh3 /. dh4 /. dh5 /. dh6]];

H[u_] := (ar ns1)/(1 + ar ns1) h[u] + 1/(1 + ar ns1) x[h[u]]
dH1 = Simplify[H^(1)[u] /. dh1];
dH2 = Simplify[H^(2)[u] /. dh1 /. dh2];
dH3 = Simplify[H^(3)[u] /. dh1 /. dh2 /. dh3];
dH4 = Simplify[H^(4)[u] /. dh1 /. dh2 /. dh3 /. dh4];
dH5 = Simplify[H^(5)[u] /. dh1 /. dh2 /. dh3 /. dh4 /. dh5];
dH6 = Simplify[H^(6)[u] /. dh1 /. dh2 /. dh3 /. dh4 /. dh5 /. dh6];
dH7 = Simplify[H^(7)[u] /. dh1 /. dh2 /. dh3 /. dh4 /. dh5 /. dh6 /. dh7];
```

d) Another expression of probability mass distribution

Another expression of probability mass distribution of multiple leakage neutrons is possible. Instead of $\overline{\nu_{s/i\,m}}$ (factorial moment), $\overline{\eta_{s/i\,m}}$ that means directly the number of multiple neutron pairs is defined as:

$$\overline{\eta_{s/i\,m}} \overset{\text{def}}{=} \sum_{\nu_{s/i}=m}^{\max} \binom{\nu_{s/i}}{m} P_\nu = \frac{\overline{\nu_{s/i}\,(\nu_{s/i}-1)\,(\nu_{s/i}-2)\cdots(\nu_{s/i}-m+1)}}{m\,!} \quad \text{and } M_{L\eta} = M_{L\nu} \qquad \text{(d1)}$$

Probability mass distribution of multiple leakage neutrons using $\overline{\eta_{s/i\,m}}$ is given as:

$$P_m\,(m{=}1) = M_L\,(1+\alpha_r)\,\overline{\eta_{s1}} \qquad \text{(d2)}$$

$$P_m\,(m{=}2) = M_L^2\,\left[\overline{\eta_{s2}} + \overline{\eta_{i2}}\,M_{L\eta}\,(1+\alpha_r)\,\overline{\eta_{s1}}\,\right] \qquad \text{(d3)}$$

$$P_m\,(m{=}3) = M_L^3\,\left[\overline{\eta_{s3}} + 2\,\overline{\eta_{i2}}\,M_{L\eta}\,\overline{\eta_{s2}} + \left(\overline{\eta_{i3}}\,M_{L\eta} + 2\,\overline{\eta_{i2}}^2\,M_{L\eta}^2\right)\,(1+\alpha_r)\,\overline{\eta_{s1}}\,\right] \qquad \text{(d4)}$$

$$\begin{aligned} P_m\,(m{=}4) = M_L^4\,\Big[&\overline{\eta_{s4}} + 3\,\overline{\eta_{i2}}\,M_{L\eta}\,\overline{\eta_{s3}} \\ &+ \left(2\,\overline{\eta_{i3}}\,M_{L\eta} + 5\,\overline{\eta_{i2}}^2\,M_{L\eta}^2\right)\,\overline{\eta_{s2}} \\ &+ \left(\overline{\eta_{i4}}\,M_{L\eta} + 5\,\overline{\eta_{i3}}\,\overline{\eta_{i2}}\,M_{L\eta}^2 + 5\,\overline{\eta_{i2}}^3\,M_{L\eta}^3\right)\,(1+\alpha_r)\,\overline{\eta_{s1}}\,\Big] \end{aligned} \qquad \text{(d5)}$$

$$
\begin{aligned}
P_m\,(m{=}5) = M_L^5\ \{\,&\overline{\eta_{s5}} + 4\,\overline{\eta_{i2}}\,M_{L\eta}\,\overline{\eta_{s4}} \\
&+ \left(3\,\overline{\eta_{i3}}\,M_{L\eta} + 9\,\overline{\eta_{i2}}^2\,M_{L\eta}^2\right)\overline{\eta_{s3}} \\
&+ \left(2\,\overline{\eta_{i4}}\,M_{L\eta} + 12\,\overline{\eta_{i3}}\,\overline{\eta_{i2}}\,M_{L\eta}^2 + 14\,\overline{\eta_{i2}}^3\,M_{L\eta}^3\right)\overline{\eta_{s2}} \\
&+ \Big[\,\overline{\eta_{i5}}\,M_{L\eta} + \left(6\,\overline{\eta_{i4}}\,\overline{\eta_{i2}} + 3\,\overline{\eta_{i3}}^2\right)M_{L\eta}^2 \\
&\qquad + 21\,\overline{\eta_{i3}}\,\overline{\eta_{i2}}^2\,M_{L\eta}^3 + 14\,\overline{\eta_{i2}}^4\,M_{L\eta}^4\Big]\,(1+\alpha_r)\,\overline{\eta_{s1}}\,\}
\end{aligned}
\tag{d6}
$$

$$
\begin{aligned}
P_m\,(m{=}6) = M_L^6\ \{\,&\overline{\eta_{s6}} + 5\,\overline{\eta_{i2}}\,M_{L\eta}\,\overline{\eta_{s5}} \\
&+ \left(4\,\overline{\eta_{i3}}\,M_{L\eta} + 14\,\overline{\eta_{i2}}^2\,M_{L\eta}^2\right)\overline{\eta_{s4}} \\
&+ \left(3\,\overline{\eta_{i4}}\,M_{L\eta} + 21\,\overline{\eta_{i3}}\,\overline{\eta_{i2}}\,M_{L\eta}^2 + 28\,\overline{\eta_{i2}}^3\,M_{L\eta}^3\right)\overline{\eta_{s3}} \\
&+ \Big[\,2\,\overline{\eta_{i5}}\,M_{L\eta} + \left(14\,\overline{\eta_{i4}}\,\overline{\eta_{i2}} + 7\,\overline{\eta_{i3}}^2\right)M_{L\eta}^2 \\
&\qquad + 56\,\overline{\eta_{i3}}\,\overline{\eta_{i2}}^2\,M_{L\eta}^3 + 42\,\overline{\eta_{i2}}^4\,M_{L\eta}^4\Big]\,\overline{\eta_{s2}} \\
&+ \Big[\,\overline{\eta_{i6}}\,M_{L\eta} + (7\,\overline{\eta_{i5}}\,\overline{\eta_{i2}} + 7\,\overline{\eta_{i4}}\,\overline{\eta_{i3}})\,M_{L\eta}^2 \\
&\qquad + \left(28\,\overline{\eta_{i4}}\,\overline{\eta_{i2}}^2 + 28\,\overline{\eta_{i3}}^2\,\overline{\eta_{i2}}\right)M_{L\eta}^3 \\
&\qquad\quad + 84\,\overline{\eta_{i3}}\,\overline{\eta_{i2}}^3\,M_{L\eta}^4 + 42\,\overline{\eta_{i2}}^5\,M_{L\eta}^5\Big]\,(1+\alpha_r)\,\overline{\eta_{s1}}\,\}
\end{aligned}
\tag{d7}
$$

$$
\begin{aligned}
P_m\,(m{=}7) = M_L^7\ \{\,&\overline{\eta_{s7}} + 6\,\overline{\eta_{i2}}\,M_{L\eta}\,\overline{\eta_{s6}} \\
&+ \left(5\,\overline{\eta_{i3}}\,M_{L\eta} + 20\,\overline{\eta_{i2}}^2\,M_{L\eta}^2\right)\overline{\eta_{s5}} \\
&+ \left(4\,\overline{\eta_{i4}}\,M_{L\eta} + 32\,\overline{\eta_{i3}}\,\overline{\eta_{i2}}\,M_{L\eta}^2 + 48\,\overline{\eta_{i2}}^3\,M_{L\eta}^3\right)\overline{\eta_{s4}} \\
&+ \Big[\,3\,\overline{\eta_{i5}}\,M_{L\eta} + \left(24\,\overline{\eta_{i4}}\,\overline{\eta_{i2}} + 12\,\overline{\eta_{i3}}^2\right)M_{L\eta}^2 \\
&\qquad + 108\,\overline{\eta_{i3}}\,\overline{\eta_{i2}}^2\,M_{L\eta}^3 + 90\,\overline{\eta_{i2}}^4\,M_{L\eta}^4\Big]\,\overline{\eta_{s3}} \\
&+ \Big[\,2\,\overline{\eta_{i6}}\,M_{L\eta} + (16\,\overline{\eta_{i5}}\,\overline{\eta_{i2}} + 16\,\overline{\eta_{i4}}\,\overline{\eta_{i3}})\,M_{L\eta}^2 \\
&\qquad + \left(72\,\overline{\eta_{i4}}\,\overline{\eta_{i2}}^2 + 72\,\overline{\eta_{i3}}^2\,\overline{\eta_{i2}}\right)M_{L\eta}^3 \\
&\qquad\quad + 240\,\overline{\eta_{i3}}\,\overline{\eta_{i2}}^3\,M_{L\eta}^4 + 132\,\overline{\eta_{i2}}^5\,M_{L\eta}^5\Big]\,\overline{\eta_{s2}} \\
&+ \Big[\,\overline{\eta_{i7}}\,M_{L\eta} + \left(8\,\overline{\eta_{i6}}\,\overline{\eta_{i2}} + 8\,\overline{\eta_{i5}}\,\overline{\eta_{i3}} + 4\,\overline{\eta_{i4}}^2\right)M_{L\eta}^2 \\
&\qquad + \left(36\,\overline{\eta_{i5}}\,\overline{\eta_{i2}}^2 + 72\,\overline{\eta_{i4}}\,\overline{\eta_{i3}}\,\overline{\eta_{i2}} + 12\,\overline{\eta_{i3}}^3\right)M_{L\eta}^3 \\
&\qquad + \left(120\,\overline{\eta_{i4}}\,\overline{\eta_{i2}}^3 + 180\,\overline{\eta_{i3}}^2\,\overline{\eta_{i2}}^2\right)M_{L\eta}^4 \\
&\qquad\quad + 330\,\overline{\eta_{i3}}\,\overline{\eta_{i2}}^4\,M_{L\eta}^5 + 132\,\overline{\eta_{i2}}^6\,M_{L\eta}^6\Big]\,(1+\alpha_r)\,\overline{\eta_{s1}}\,\}
\end{aligned}
\tag{d8}
$$

Compared to the expression using $\overline{\nu_{s/i\,m}}$, it is not necessary to divide by $m!$, coefficients become smaller and physical meaning becomes natural. Singles, Doubles, Triples, Quadruples, Quintuples, Sextuples, Septuples are given in equation (22) in the body.

e) Joint probability generating function and relation to covariance

Joint probability generating function G_{XY} for discrete stochastic variables X and Y (non-negative integers) is defined as a function of continuous variable $|z_1| \leq 1$ and $|z_2| \leq 1$:

$$G_{XY}\,(z_1, z_2) \overset{\text{def}}{=} \overline{z_1^X\, z_2^Y} = \sum_{n=0}^{\infty}\sum_{m=0}^{\infty} z_1^n\, z_2^m\, P_{XY}\,(X{=}n, Y{=}m) \tag{e1}$$

From equation (a4):

$$\overline{X} = \left.\frac{\partial\, G_{XY}\,(z_1, z_2)}{\partial\, z_1}\right|_{z_1=1} \tag{e2}$$

$$\overline{Y} = \left.\frac{\partial\, G_{XY}\,(z_1, z_2)}{\partial\, z_2}\right|_{z_2=1} \tag{e3}$$

and:

$$\overline{X(X-1)} = \left.\frac{\partial^2\, G_{XY}\,(z_1, z_2)}{\partial z_1^2}\right|_{z_1=1} \tag{e4}$$

$$\overline{XY} = \left.\frac{\partial^2\, G_{XY}\,(z_1, z_2)}{\partial z_1\, \partial z_2}\right|_{z_1=z_2=1} \tag{e5}$$

Self-covariance (not self-correlation nor autocorrelation) of X is defined as:

$$\mathrm{Cov}(X, X) = \overline{X(X{-}1)} - (\overline{X})^2 = \overline{X^2} - \overline{X} - (\overline{X})^2 = \underbrace{\overline{(X{-}\overline{X})^2}}_{\text{variance}} - \underbrace{\overline{X}}_{\text{mean}} \tag{e6}$$

Thus:

$$\mathrm{Var}(X) = \mathrm{Cov}(X, X) + \mu(X) \tag{e7}$$

$$\mathrm{Var}(X) = M''_{P_X} + M'_{P_X} - \left(M'_{P_X}\right)^2 \tag{e8}$$

Normal covariance of X and Y is defined as:

$$\mathrm{Cov}(X, Y) = \overline{XY} - \overline{X}\,\overline{Y} = \underbrace{\overline{(X{-}\overline{X})(Y{-}\overline{Y})}}_{\text{covariance}} \tag{e9}$$

Difference between equation (e6) and equation (e9) comes from the degree of freedom. Therefore,

$\left.\dfrac{\partial^2 G}{\partial X^2}\right|_{X=1} = \overline{N_1(N_1{-}1)}$ is not self-covariance of N_1 and

$\left.\dfrac{\partial^2 G}{\partial X \partial Y}\right|_{X=Y=1} = \overline{N_1 N_2}$ is not covariance of N_1 and N_2.

f) Various expressions of leakage multiplication

Conventional expression is shown below where l is probability of an event where a neutron leaks from a sample:

$$M_L = l\, M_T = l\, [1 + p\, \overline{\nu_{i1}} + (p\, \overline{\nu_{i1}})^2 \ldots] = \frac{l}{1 - p\, \overline{\nu_{i1}}} = \frac{1 - p - p_c}{1 - p\, \overline{\nu_{i1}}} \approx \frac{1 - p}{1 - p\, \overline{\nu_{i1}}} \tag{f1}$$

Another expression derived in the body is:

$$M_L = \frac{1}{S_1\, \overline{\nu_{s1}}} \left(S_1\, \overline{\nu_{s1}} + \overline{F_1}\, \overline{\nu_{i1}^1} + \overline{F_2}\, \overline{\nu_{i1}^2} \right) \tag{f2}$$

An additional different expression is possible using l_s which is prompt neutron lifespan introduced in the primary part of reactor physics:

$$\frac{1}{n} \frac{\mathrm{d}n}{\mathrm{d}t} = \frac{k_{eff}\ (1 - \beta_{eff}) - 1}{l_s} \approx \frac{k_{eff} - 1}{l_s} \quad \text{if delayed neutron is sufficiently small} \tag{f3}$$

$$\rho \overset{\text{def}}{=} \frac{k_{eff} - 1}{k_{eff}} \tag{f4}$$

$$\Lambda \overset{\text{def}}{=} \frac{l_s}{k_{eff}} \tag{f5}$$

$$\alpha = \frac{\beta_{eff} - \rho}{\Lambda} \approx \frac{-\rho}{\Lambda} = \frac{1 - k_{eff}}{l_s} \quad \text{if delayed neutron is sufficiently small} \tag{f6}$$

where n is the number of neutrons in arbitrary generation, β_{eff} is the ratio of the number of delayed neutrons to the one of total neutrons, k_{eff} is effective multiplication factor, ρ is reactivity and Λ is prompt neutron generation time. There equations satisfy the relation below that is the definition of α:

$$n(t) = n(0)\, e^{-\alpha t} \tag{f7}$$

On the other hand, the following equation was introduced by Hoffmann (Reference 8):

$$\frac{\overline{\nu}}{\alpha \tau} = \frac{1}{1 - p\, \overline{\nu}} = \frac{1}{1 - k_{eff}} = M_T \tag{f8}$$

Therefore:

$$M_L = l\, M_T = \frac{l\, \overline{\nu}}{\alpha\, \tau} = \frac{l}{\alpha\, l_s} \qquad \because \frac{\tau}{l_s} = \overline{\nu} \tag{f9}$$

This equation is consistent with Table 7-1 ~ 7-4, because M_L and l are close to one and l_s is the order of 10^{-6}.

Regarding equation (f9), physical meaning of $\overline{\nu}/(\alpha\tau) = M_T$ is clear because $1/(\alpha\tau)$ is the number of induced fissions by one source neutron and $\overline{\nu}/(\alpha\tau)$ is the number of neutrons induced by one source neutron. Physical meaning of $\tau = \overline{\nu}\, l_s$ is also clear because τ is mean time between fissions and l_s is neutron lifespan.

Secondary additional different expression is possible using ϵ_n, ϵ_1 and ϵ_2:

$$M_L = \frac{\epsilon_1}{\epsilon_n}\frac{1}{\alpha_1\,\tau_1} = \frac{\epsilon_2}{\epsilon_n}\frac{1}{\alpha_2\,\tau_2} \qquad \text{(f10)}$$

This equation satisfy equation (83) and (84) in the body. Furthermore, M_L is written as a sum of M_L^1 (contribution of fast neutron fission) and M_L^2 (contribution of thermal neutron fission):

$$M_L^1 = \frac{1}{S_1\,\overline{\nu_{s1}}}\left(S_1\,\overline{\nu_{s1}} + \overline{F_1}\,\overline{\nu_{i1}^1}\right) = 1 + \frac{\overline{\nu_{i1}^1}}{\alpha_1\,\tau_1} \qquad \text{(f11)}$$

$$M_L^2 = \frac{1}{S_1\,\overline{\nu_{s1}}}\,\overline{F_2}\,\overline{\nu_{i1}^2} = \frac{\overline{\nu_{i1}^2}}{\alpha_2\,\tau_2} \qquad \text{(f12)}$$

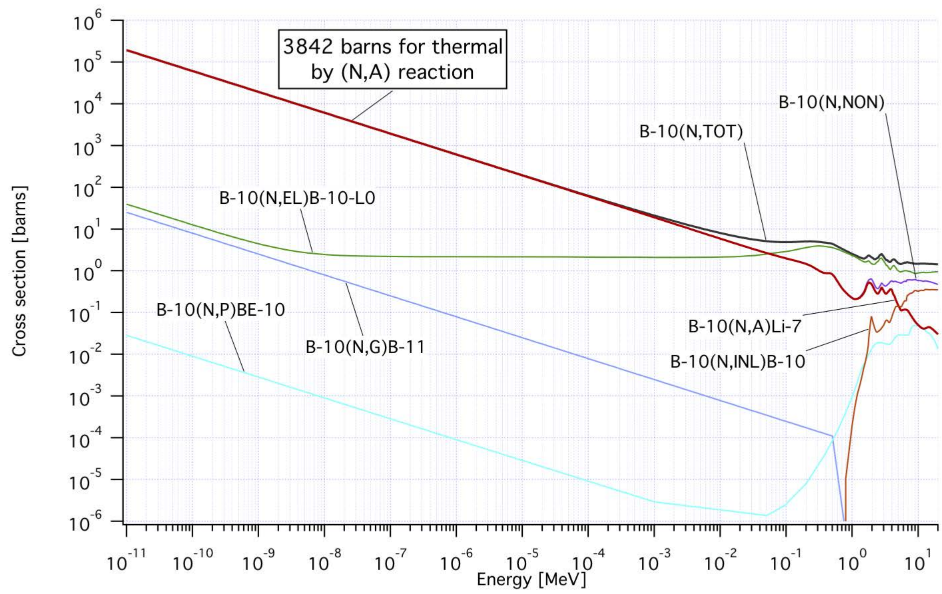
These equations are derived for future references.

g) Cross sections to be referred in equation (126)

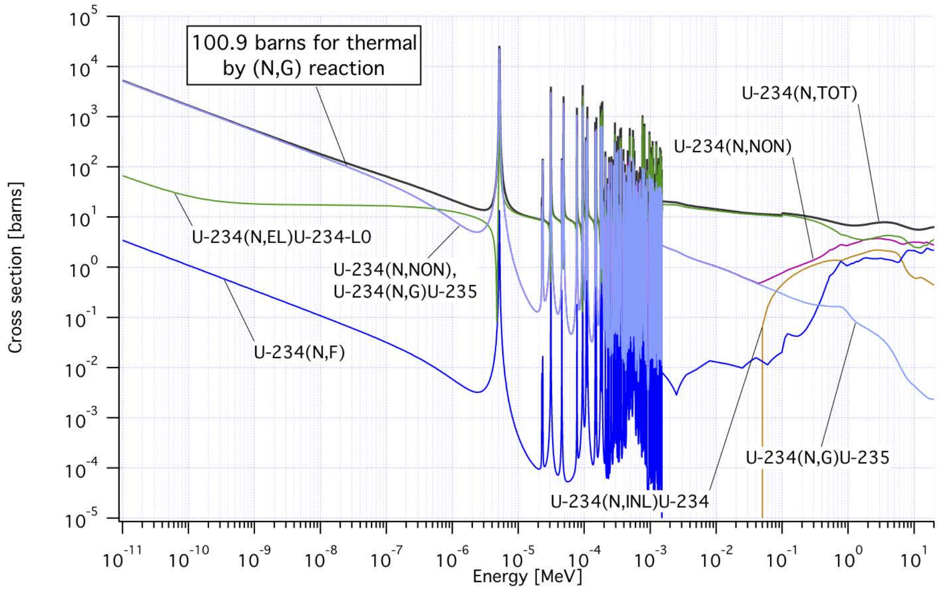
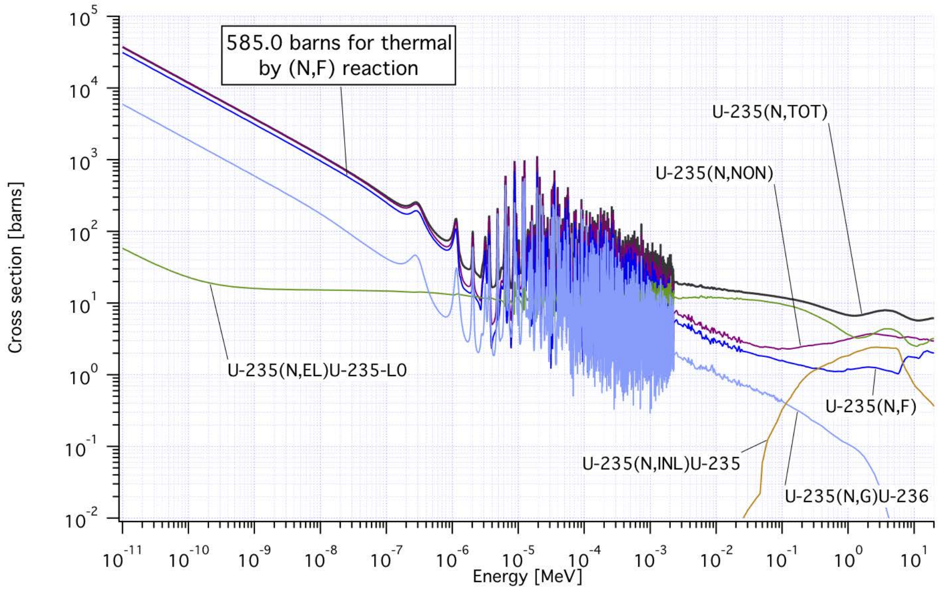
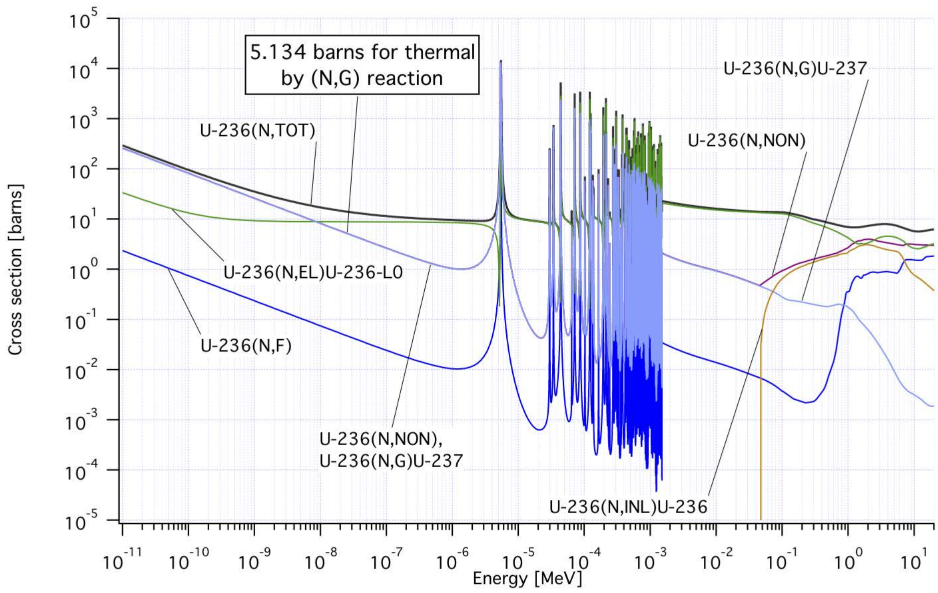
・Abbreviations A, G, P, F, TOT, EL, NON and INL used in reactions are α, γ, proton, fission, total, elastic, nonelastic (≡TOT−EL), inelastic (≡NON−absorptions), respectively;

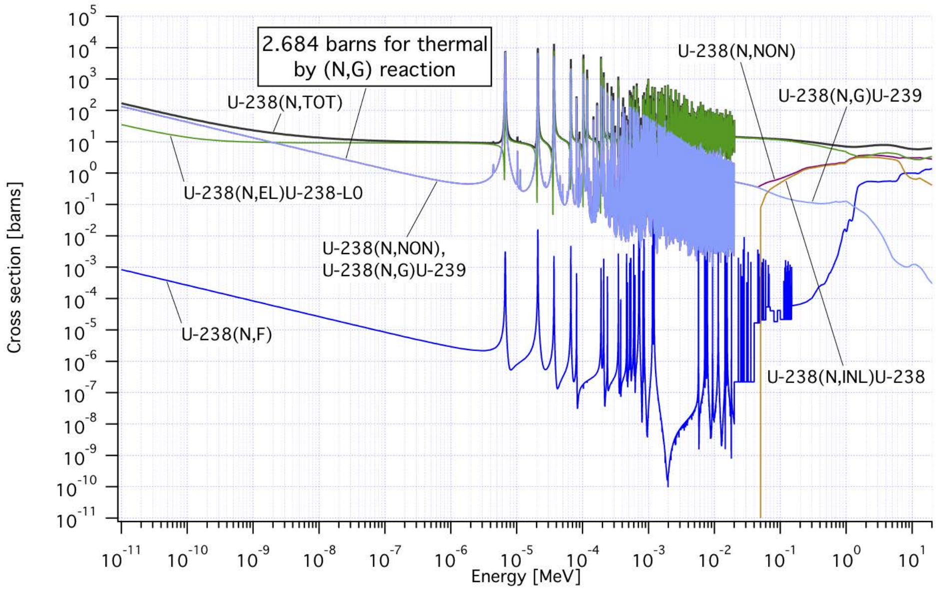
・^{16}O is eliminated due to small (n,γ) cross section (0.00019 barns for thermal neutron);

・^{237}Np (n,γ) ⇒ ^{238}Np, ^{241}Am (n,γ) ⇒ ^{242m}Am or ^{243}Am (via ^{242}Am) are added for safeguards.

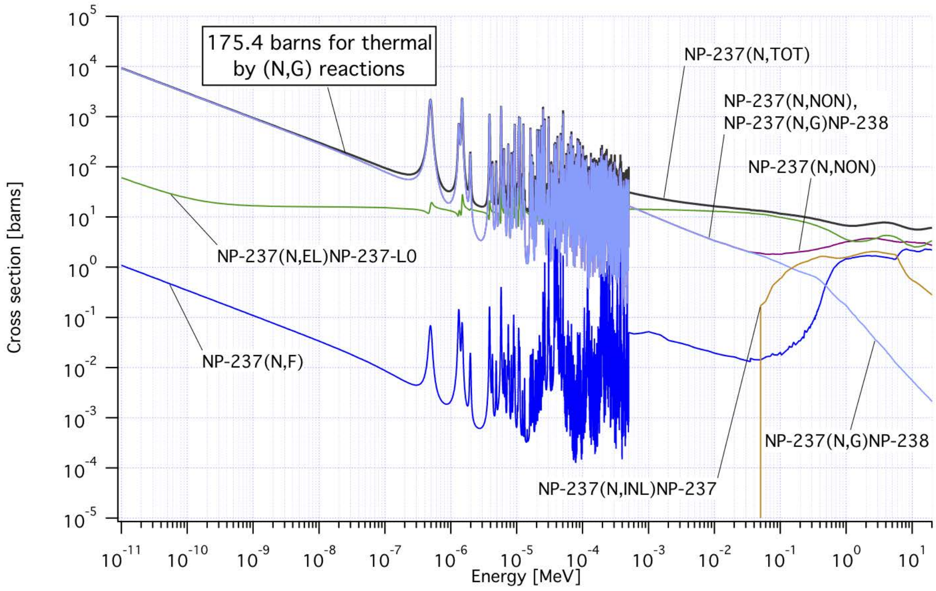


^{10}B

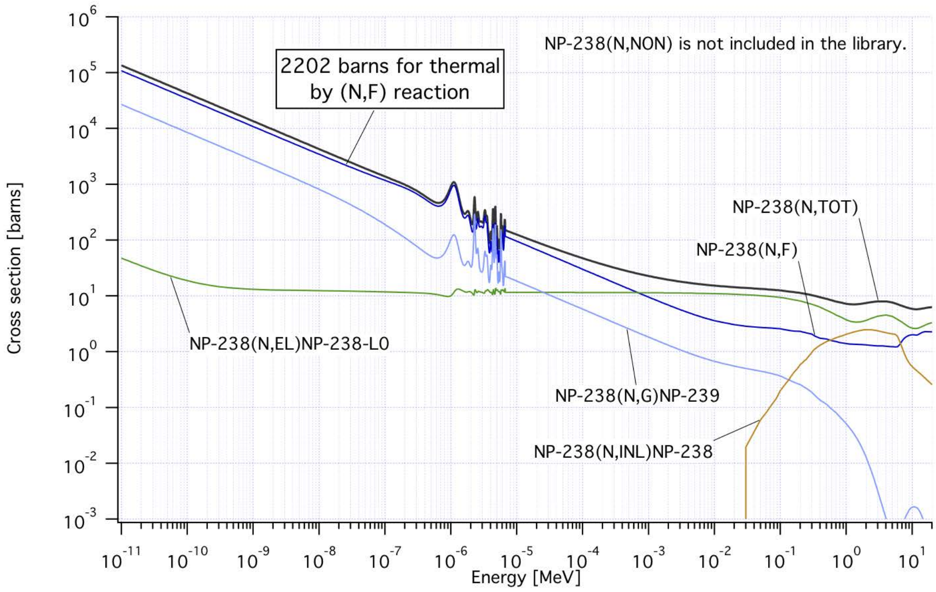
^{234}U	
^{235}U	
^{236}U	



^{238}U

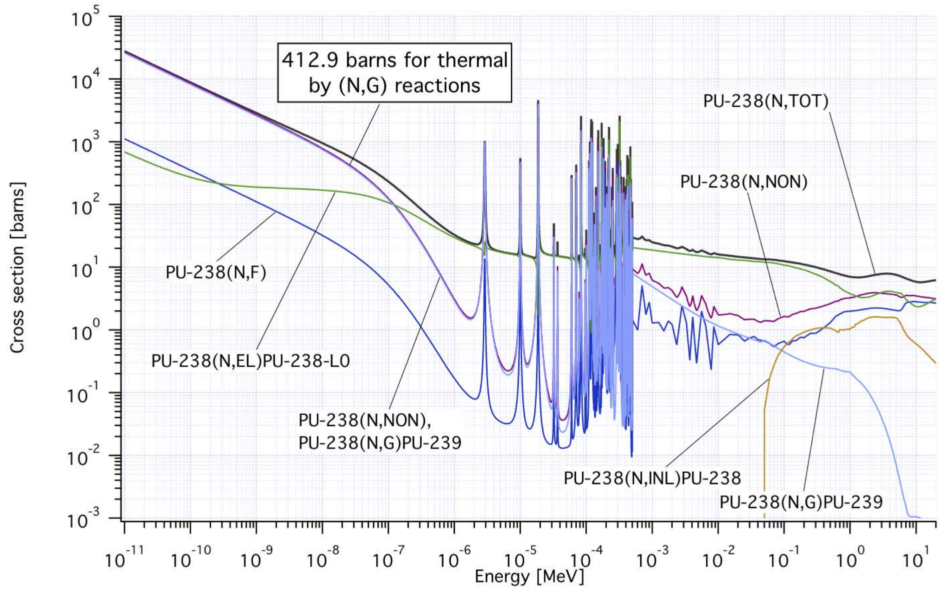


^{237}Np

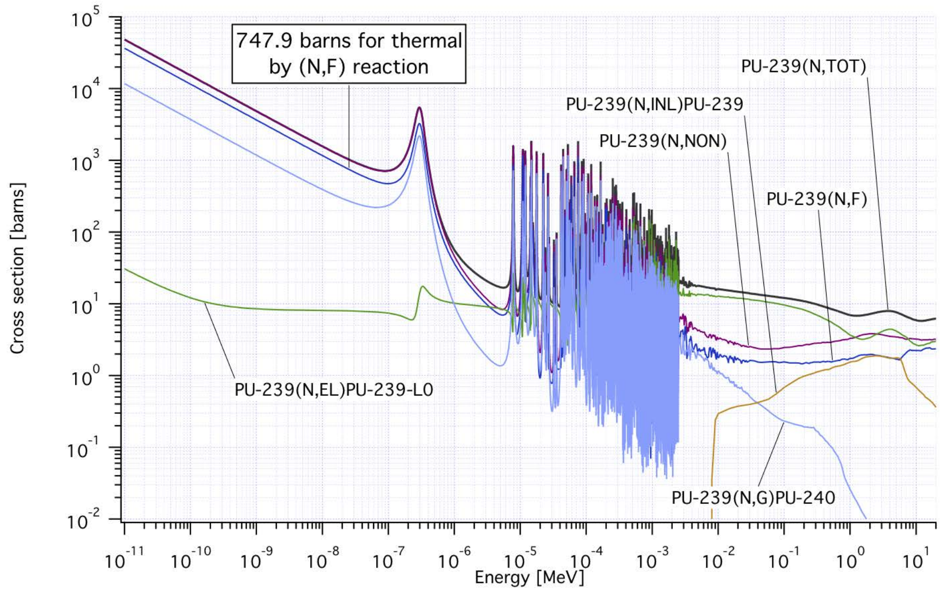


^{238}Np

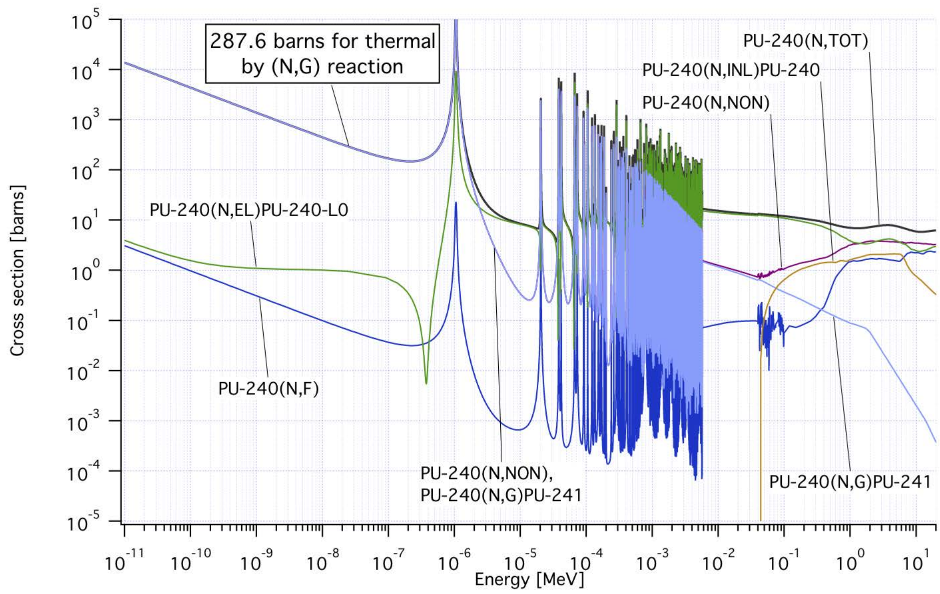
^{238}Pu

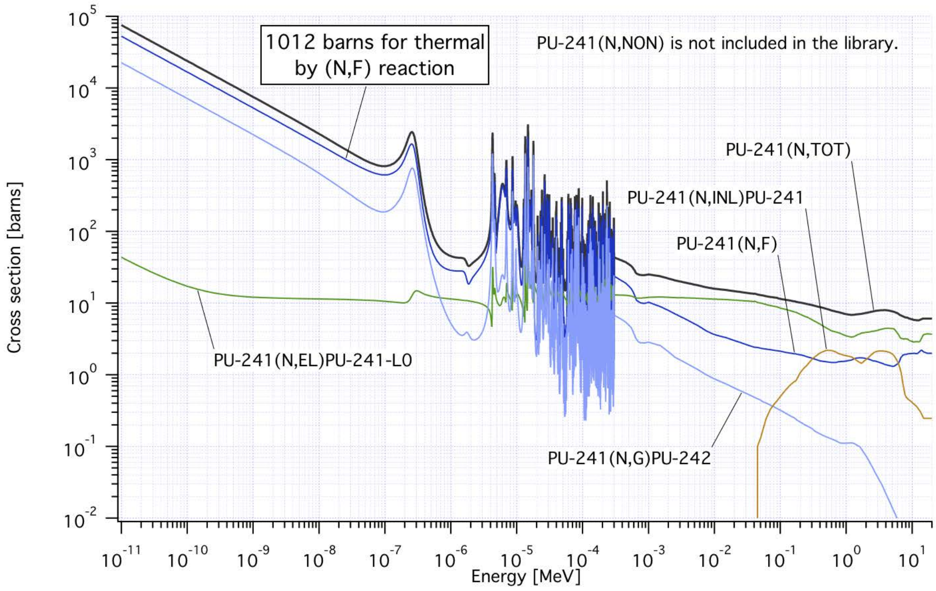


^{239}Pu

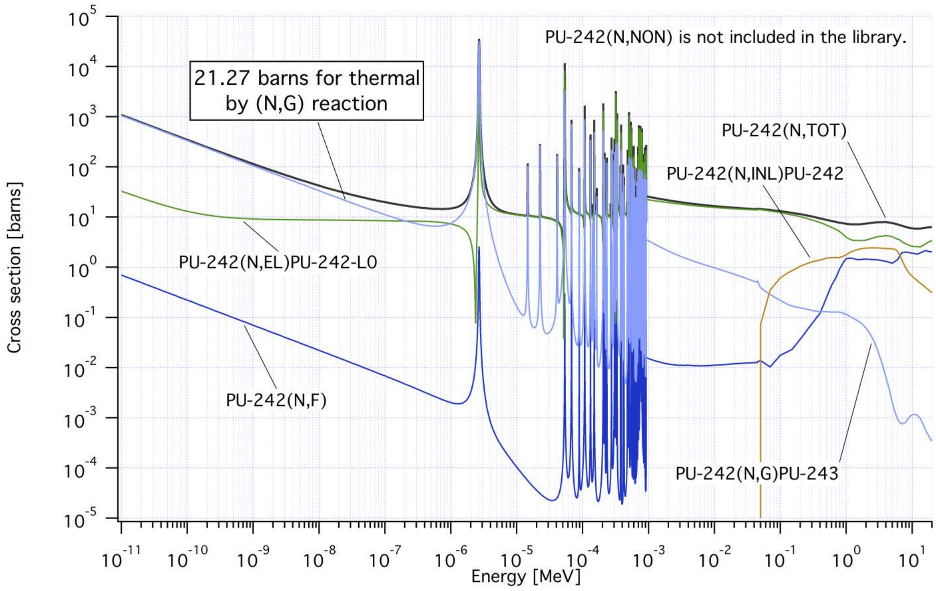


^{240}Pu

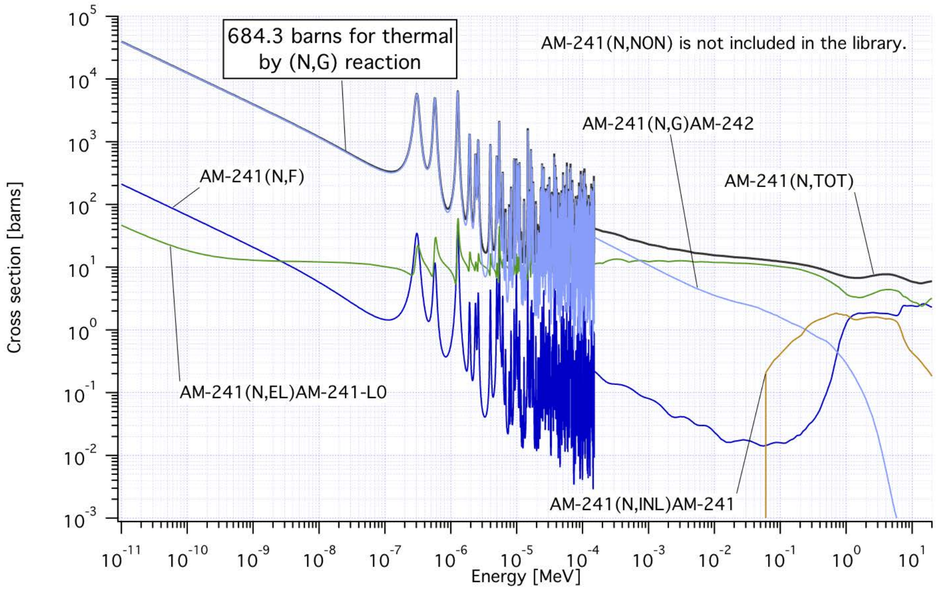




^{241}Pu

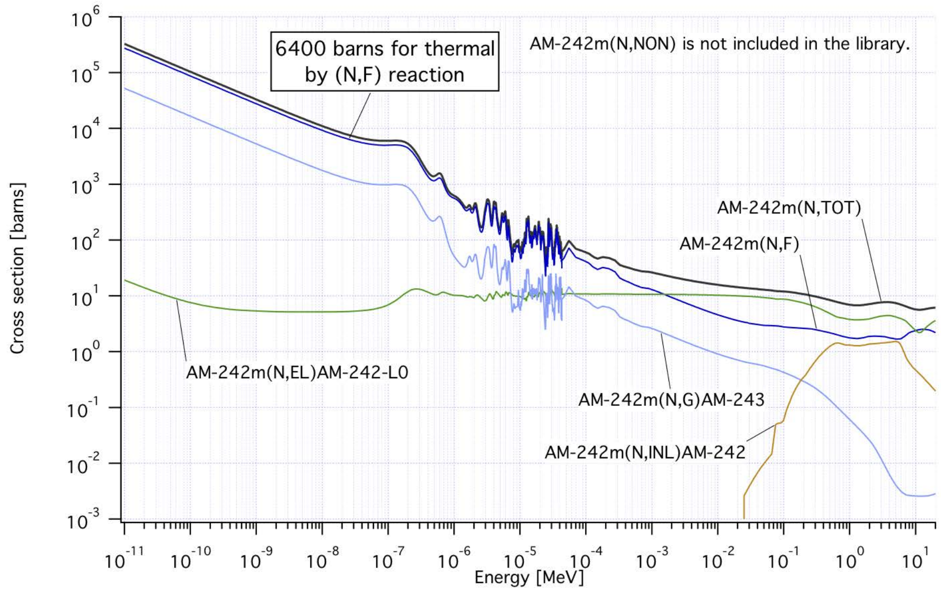


^{242}Pu

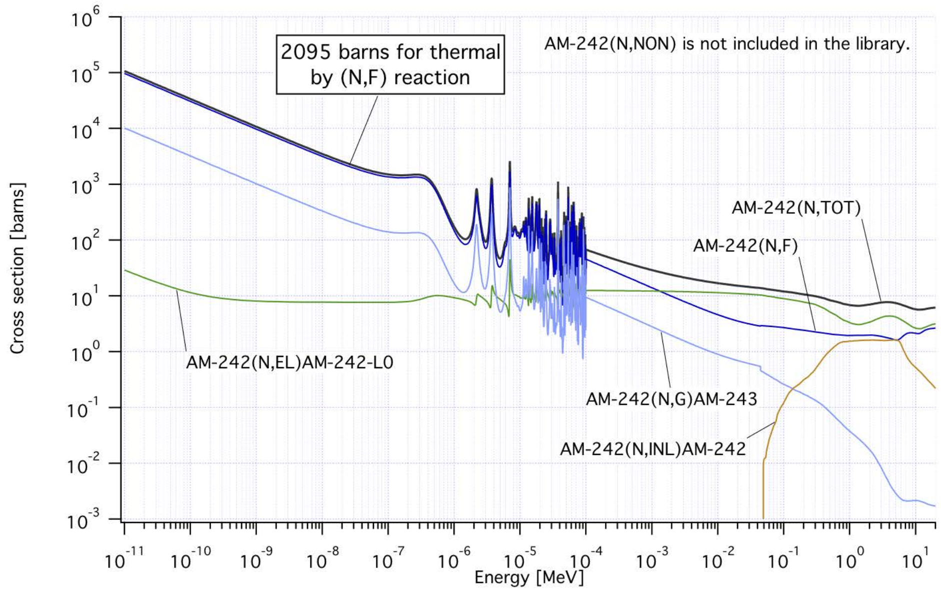


^{241}Am

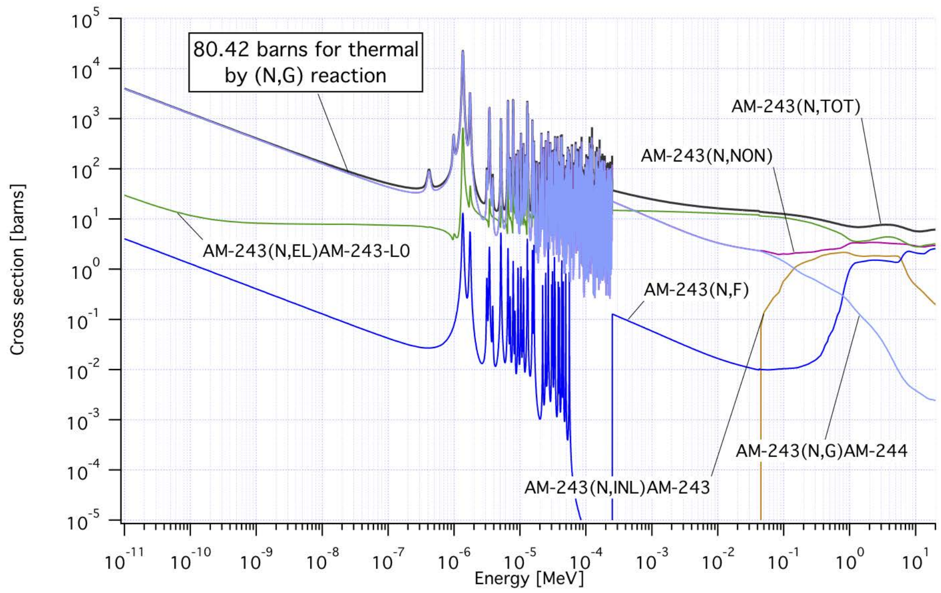
^{242m}Am

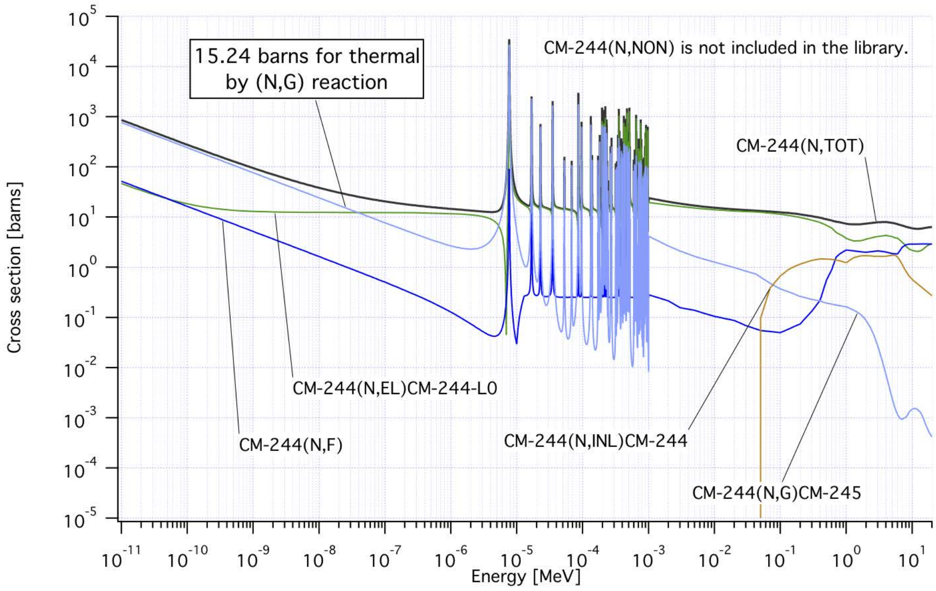


^{242}Am



^{243}Am




15.24 barns for thermal by (N,G) reaction
CM-244(N,NON) is not included in the library.
CM-244(N,TOT)
CM-244(N,EL)CM-244-L0
CM-244(N,F)
CM-244(N,INL)CM-244
CM-244(N,G)CM-245
Cross section [barns]
Energy [MeV]

^{244}Cm

This is a blank page.

国際単位系（SI）

表１．SI 基本単位

基本量	SI 基本単位	
	名称	記号
長さ	メートル	m
質量	キログラム	kg
時間	秒	s
電流	アンペア	A
熱力学温度	ケルビン	K
物質量	モル	mol
光度	カンデラ	cd

表２．基本単位を用いて表されるSI組立単位の例

組立量	SI 組立単位	
	名称	記号
面積	平方メートル	m^2
体積	立方メートル	m^3
速さ，速度	メートル毎秒	m/s
加速度	メートル毎秒毎秒	m/s^2
波数	毎メートル	m^{-1}
密度，質量密度	キログラム毎立方メートル	kg/m^3
面積密度	キログラム毎平方メートル	kg/m^2
比体積	立方メートル毎キログラム	m^3/kg
電流密度	アンペア毎平方メートル	A/m^2
磁界の強さ	アンペア毎メートル	A/m
量濃度 (a)，濃度	モル毎立方メートル	mol/m^3
質量濃度	キログラム毎立方メートル	kg/m^3
輝度	カンデラ毎平方メートル	cd/m^2
屈折率 (b)	（数字の） 1	1
比透磁率 (b)	（数字の） 1	1

（a） 量濃度（amount concentration）は臨床化学の分野では物質濃度（substance concentration）ともよばれる。
（b） これらは無次元量あるいは次元１をもつ量であるが、そのことを表す単位記号である数字の１は通常は表記しない。

表３．固有の名称と記号で表されるSI組立単位

組立量	SI 組立単位			
	名称	記号	他のSI単位による表し方	SI基本単位による表し方
平面角	ラジアン (b)	rad	1 (b)	m/m
立体角	ステラジアン (b)	sr (c)	1 (b)	m^2/m^2
周波数	ヘルツ (d)	Hz		s^{-1}
力	ニュートン	N		$m\ kg\ s^{-2}$
圧力，応力	パスカル	Pa	N/m^2	$m^{-1}\ kg\ s^{-2}$
エネルギー，仕事，熱量	ジュール	J	N m	$m^2\ kg\ s^{-2}$
仕事率，工率，放射束	ワット	W	J/s	$m^2\ kg\ s^{-3}$
電荷，電気量	クーロン	C		s A
電位差（電圧），起電力	ボルト	V	W/A	$m^2\ kg\ s^{-3}\ A^{-1}$
静電容量	ファラド	F	C/V	$m^{-2}\ kg^{-1}\ s^4\ A^2$
電気抵抗	オーム	Ω	V/A	$m^2\ kg\ s^{-3}\ A^{-2}$
コンダクタンス	ジーメンス	S	A/V	$m^{-2}\ kg^{-1}\ s^3\ A^2$
磁束	ウエーバ	Wb	Vs	$m^2\ kg\ s^{-2}\ A^{-1}$
磁束密度	テスラ	T	Wb/m^2	$kg\ s^{-2}\ A^{-1}$
インダクタンス	ヘンリー	H	Wb/A	$m^2\ kg\ s^{-2}\ A^{-2}$
セルシウス温度	セルシウス度 (e)	℃		K
光束	ルーメン	lm	cd sr (c)	cd
照度	ルクス	lx	lm/m^2	$m^{-2}\ cd$
放射性核種の放射能 (f)	ベクレル (d)	Bq		s^{-1}
吸収線量, 比エネルギー分与, カーマ	グレイ	Gy	J/kg	$m^2\ s^{-2}$
線量当量, 周辺線量当量, 方向性線量当量, 個人線量当量	シーベルト (g)	Sv	J/kg	$m^2\ s^{-2}$
酸素活性	カタール	kat		$s^{-1}\ mol$

(a)SI接頭語は固有の名称と記号を持つ組立単位と組み合わせても使用できる。しかし接頭語を付した単位はもはやコヒーレントではない。
(b)ラジアンとステラジアンは数字の１に対する単位の特別な名称で、量についての情報をつたえるために使われる。実際には、使用する時には記号rad及びsrが用いられるが、習慣として組立単位としての記号である数字の１は明示されない。
(c)測光学ではステラジアンという名称と記号srを単位の表し方の中に、そのまま維持している。
(d)ヘルツは周期現象についてのみ、ベクレルは放射性核種の統計的過程についてのみ使用される。
(e)セルシウス度はケルビンの特別な名称で、セルシウス温度を表すために使用される。セルシウス度とケルビンの単位の大きさは同一である。したがって、温度差や温度間隔を表す数値はどちらの単位で表しても同じである。
(f)放射性核種の放射能（activity referred to a radionuclide）は、しばしば誤った用語で”radioactivity”と記される。
(g)単位シーベルト（PV,2002,70,205）についてはCIPM勧告2（CI-2002）を参照。

表４．単位の中に固有の名称と記号を含むSI組立単位の例

組立量	SI 組立単位		
	名称	記号	SI 基本単位による表し方
粘度	パスカル秒	Pa s	$m^{-1}\ kg\ s^{-1}$
力のモーメント	ニュートンメートル	N m	$m^2\ kg\ s^{-2}$
表面張力	ニュートン毎メートル	N/m	$kg\ s^{-2}$
角速度	ラジアン毎秒	rad/s	$m\ m^{-1}\ s^{-1}=s^{-1}$
角加速度	ラジアン毎秒毎秒	rad/s^2	$m\ m^{-1}\ s^{-2}=s^{-2}$
熱流密度，放射照度	ワット毎平方メートル	W/m^2	$kg\ s^{-3}$
熱容量，エントロピー	ジュール毎ケルビン	J/K	$m^2\ kg\ s^{-2}\ K^{-1}$
比熱容量，比エントロピー	ジュール毎キログラム毎ケルビン	J/(kg K)	$m^2\ s^{-2}\ K^{-1}$
比エネルギー	ジュール毎キログラム	J/kg	$m^2\ s^{-2}$
熱伝導率	ワット毎メートル毎ケルビン	W/(m K)	$m\ kg\ s^{-3}\ K^{-1}$
体積エネルギー	ジュール毎立方メートル	J/m^3	$m^{-1}\ kg\ s^{-2}$
電界の強さ	ボルト毎メートル	V/m	$m\ kg\ s^{-3}\ A^{-1}$
電荷密度	クーロン毎立方メートル	C/m^3	$m^{-3}\ s\ A$
表面電荷	クーロン毎平方メートル	C/m^2	$m^{-2}\ s\ A$
電束密度，電気変位	クーロン毎平方メートル	C/m^2	$m^{-2}\ s\ A$
誘電率	ファラド毎メートル	F/m	$m^{-3}\ kg^{-1}\ s^4\ A^2$
透磁率	ヘンリー毎メートル	H/m	$m\ kg\ s^{-2}\ A^{-2}$
モルエネルギー	ジュール毎モル	J/mol	$m^2\ kg\ s^{-2}\ mol^{-1}$
モルエントロピー, モル熱容量	ジュール毎モル毎ケルビン	J/(mol K)	$m^2\ kg\ s^{-2}\ K^{-1}\ mol^{-1}$
照射線量（Ｘ線及びγ線）	クーロン毎キログラム	C/kg	$kg^{-1}\ s\ A$
吸収線量率	グレイ毎秒	Gy/s	$m^2\ s^{-3}$
放射強度	ワット毎ステラジアン	W/sr	$m^4\ m^{-2}\ kg\ s^{-3}=m^2\ kg\ s^{-3}$
放射輝度	ワット毎平方メートル毎ステラジアン	$W/(m^2\ sr)$	$m^2\ m^{-2}\ kg\ s^{-3}=kg\ s^{-3}$
酵素活性濃度	カタール毎立方メートル	kat/m^3	$m^{-3}\ s^{-1}\ mol$

表５．SI 接頭語

乗数	名称	記号	乗数	名称	記号
10^{24}	ヨタ	Y	10^{-1}	デシ	d
10^{21}	ゼタ	Z	10^{-2}	センチ	c
10^{18}	エクサ	E	10^{-3}	ミリ	m
10^{15}	ペタ	P	10^{-6}	マイクロ	µ
10^{12}	テラ	T	10^{-9}	ナノ	n
10^{9}	ギガ	G	10^{-12}	ピコ	p
10^{6}	メガ	M	10^{-15}	フェムト	f
10^{3}	キロ	k	10^{-18}	アト	a
10^{2}	ヘクト	h	10^{-21}	ゼプト	z
10^{1}	デカ	da	10^{-24}	ヨクト	y

表６．SIに属さないが、SIと併用される単位

名称	記号	SI 単位による値
分	min	1 min=60 s
時	h	1 h =60 min=3600 s
日	d	1 d=24 h=86 400 s
度	°	1°=(π/180) rad
分	’	1’=(1/60)°=(π/10 800) rad
秒	”	1”=(1/60)’=(π/648 000) rad
ヘクタール	ha	$1\ ha=1\ hm^2=10^4 m^2$
リットル	L，l	$1\ L=1\ l=1\ dm^3=10^3 cm^3=10^{-3} m^3$
トン	t	$1\ t=10^3\ kg$

表７．SIに属さないが、SIと併用される単位で、SI単位で表される数値が実験的に得られるもの

名称	記号	SI 単位で表される数値
電子ボルト	eV	$1\ eV=1.602\ 176\ 53(14)\times10^{-19} J$
ダルトン	Da	$1\ Da=1.660\ 538\ 86(28)\times10^{-27} kg$
統一原子質量単位	u	1 u=1 Da
天文単位	ua	$1\ ua=1.495\ 978\ 706\ 91(6)\times10^{11} m$

表８．SIに属さないが、SIと併用されるその他の単位

名称	記号	SI 単位で表される数値
バール	bar	$1\ bar=0.1 MPa=100\ kPa=10^5 Pa$
水銀柱ミリメートル	mmHg	1 mmHg≈133.322Pa
オングストローム	Å	$1\ Å=0.1nm=100pm=10^{-10} m$
海里	M	1 M=1852m
バーン	b	$1\ b=100fm^2=(10^{-12}cm)^2=10^{-28}m^2$
ノット	kn	1 kn=(1852/3600)m/s
ネーパ	Np	SI単位との数値的な関係は、対数量の定義に依存。
ベル	B	
デシベル	dB	

表９．固有の名称をもつCGS組立単位

名称	記号	SI 単位で表される数値
エルグ	erg	$1\ erg=10^{-7}\ J$
ダイン	dyn	$1\ dyn=10^{-5} N$
ポアズ	P	$1\ P=1\ dyn\ s\ cm^{-2}=0.1Pa\ s$
ストークス	St	$1\ St=1cm^2\ s^{-1}=10^{-4} m^2\ s^{-1}$
スチルブ	sb	$1\ sb=1cd\ cm^{-2}=10^4 cd\ m^{-2}$
フォト	ph	$1\ ph=1cd\ sr\ cm^{-2}=10^4 lx$
ガル	Gal	$1\ Gal=1cm\ s^{-2}=10^{-2} ms^{-2}$
マクスウエル	Mx	$1\ Mx=1G\ cm^2=10^{-8} Wb$
ガウス	G	$1\ G=1Mx\ cm^{-2}=10^{-4} T$
エルステッド (a)	Oe	$1\ Oe \triangleq (10^3/4\pi) A\ m^{-1}$

（a） ３元系のCGS単位系とSIでは直接比較できないため、等号「 ≙ 」は対応関係を示すものである。

表10．SIに属さないその他の単位の例

名称	記号	SI 単位で表される数値
キュリー	Ci	$1\ Ci=3.7\times10^{10} Bq$
レントゲン	R	$1\ R=2.58\times10^{-4} C/kg$
ラド	rad	$1\ rad=1cGy=10^{-2} Gy$
レム	rem	$1\ rem=1\ cSv=10^{-2} Sv$
ガンマ	γ	$1\ \gamma=1\ nT=10^{-9} T$
フェルミ		1 フェルミ$=1\ fm=10^{-15} m$
メートル系カラット		1 メートル系カラット = 0.2 g = 2×10^{-4}kg
トル	Torr	1 Torr = (101 325/760) Pa
標準大気圧	atm	1 atm = 101 325 Pa
カロリー	cal	1 cal=4.1858J（「15℃」カロリー），4.1868J（「IT」カロリー），4.184J（「熱化学」カロリー）
ミクロン	µ	$1\ \mu=1\mu m=10^{-6} m$

（第8版，2006年）