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

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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
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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を、確率母関数を用いて定数的に導いた。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激に増大し、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、より低次の相関を交わり追い越してゆくことを見出した。続いて、高速中性子と発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：高速中性子と発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けことを証明し、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。
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Nomenclature

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

\( \nu \) The number of neutrons arising from a fission \(( \bar{\nu} \) 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 \
\( p_\nu \) Probability distribution of the number of neutrons from a spontaneous fission \
\( f_\nu \) Probability distribution of the number of neutrons from a fast induced fission \
\( f_\nu\) Probability distribution of the number of neutrons from a thermal induced fission \
\( \epsilon \) Efficiency of counter (counts per fission) \
\( \epsilon_n \) Efficiency of counter (counts per neutron) \
\( \alpha \) 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). \
\( \tau \) Mean time between fissions \
\( 1/(\alpha \tau) \) Total number of fissions by one source neutron \
\( \alpha_r \) Ratio of neutrons arising from (\( \alpha \), n) reaction to those from spontaneous fission \
\( \lambda \) 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 
\( F_1 \) Expected number of fast induced fission and spontaneous fission per unit time 
\( F_2 \) Expected number of thermal induced fission per unit time 
\( \nu_s \) First factorial moment (expected number of neutrons) of spontaneous fission 
\( \nu_{s1} \) First factorial moment (expected number of neutrons) of fast induced fission 
\( \nu_{s1} \) First factorial moment (expected number of neutrons) of thermal induced fission 
\( \nu_{s2} \) Second factorial moment (twice of expected neutron pairs) of spontaneous fission 
\( \nu_{s1} \) Second factorial moment (twice of expected neutron pairs) of fast induced fission 
\( \nu_{s2} \) Second factorial moment (twice of expected neutron pairs) of thermal induced fission 
\( \alpha_1 \) Rate of decay for neutrons arise from fast induced fission and spontaneous fission 
\( \alpha_2 \) Rate of decay for neutrons arise from thermal induced fission 
\( \tau_1 \) Mean time between fast induced fissions or spontaneous fissions 
\( \tau_2 \) 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。

<table>
<thead>
<tr>
<th>符号</th>
<th>解説</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\nu$</td>
<td>1回の核分裂で生じる中性子の数（$\bar{\nu}$ はその期待値）</td>
</tr>
<tr>
<td>$p$</td>
<td>1個の中性子が誘導核分裂反応を生じる事象の確率</td>
</tr>
<tr>
<td>$l$</td>
<td>1個の中性子が試料から漏れる事象の確率</td>
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<td>$P_m$</td>
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<tr>
<td>$G$</td>
<td>確率母関数</td>
</tr>
<tr>
<td>$p_{\nu}$</td>
<td>1回の自発核分裂で生じる中性子の数の確率分布</td>
</tr>
<tr>
<td>$f_{1\nu}$</td>
<td>1回の誘導核分裂（高速中性子源）で生じる中性子の数の確率分布</td>
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<tr>
<td>$f_{2\nu}$</td>
<td>1回の誘導核分裂（熱中性子源）で生じる中性子の数の確率分布</td>
</tr>
<tr>
<td>$\epsilon$</td>
<td>検出効率（counts per fission）</td>
</tr>
<tr>
<td>$\epsilon_n$</td>
<td>検出効率（counts per neutron）</td>
</tr>
<tr>
<td>$\alpha$</td>
<td>系又は試料中の即発中性子減衰定数</td>
</tr>
<tr>
<td>$\tau$</td>
<td>核分裂の平均的な時間間隔</td>
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<tr>
<td>$1/(\alpha \tau)$</td>
<td>1個の中性子から生じる核分裂の総数</td>
</tr>
<tr>
<td>$\alpha_r$</td>
<td>（$\alpha$, 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</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>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</td>
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<tr>
<td>$Y$</td>
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<td>$F_{1\nu}$</td>
<td>単位時間あたり誘導核分裂（高速中性子源）の数の期待値</td>
</tr>
<tr>
<td>$F_{2\nu}$</td>
<td>単位時間あたり誘導核分裂（熱中性子源）の数の期待値</td>
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<td>$v_{1\alpha}$</td>
<td>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</td>
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<td>$v_{11\alpha}$</td>
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<tr>
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</tr>
<tr>
<td>$v_{22\alpha}$</td>
<td>誘導核分裂（熱中性子起源）の二次階乗モーメントで、生じる中性子ベア数の期待値の二倍</td>
</tr>
<tr>
<td>$\alpha_1$</td>
<td>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</td>
</tr>
<tr>
<td>$\alpha_2$</td>
<td>誘導核分裂（熱中性子起源）の即発中性子減衰定数</td>
</tr>
<tr>
<td>$\tau_1$</td>
<td>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</td>
</tr>
<tr>
<td>$\tau_2$</td>
<td>誘導核分裂（熱中性子起源）の平均的な時間間隔</td>
</tr>
<tr>
<td>$S$</td>
<td>中性子の計数率 = 相関の有無を考えない単味（singlets）計数率 = Singles</td>
</tr>
<tr>
<td>$D$</td>
<td>中性子ベア（2個1組）の計数率 = 二重相関（doublets）計数率 = Doubles</td>
</tr>
<tr>
<td>$T$</td>
<td>中性子ベア（3個1組）の計数率 = 三重相関（triplets）計数率 = Triples</td>
</tr>
<tr>
<td>$D_1$</td>
<td>自発及び誘導核分裂（高速中性子による）による中性子ベアの計数率</td>
</tr>
<tr>
<td>$D_2$</td>
<td>誘導核分裂（熱中性子による）による中性子ベアの計数率</td>
</tr>
<tr>
<td>$f_d$</td>
<td>二重相関計数時のゲートの有効割合（double gate fraction）</td>
</tr>
<tr>
<td>$M_{TP}$</td>
<td>全増倍率</td>
</tr>
<tr>
<td>$M_L$</td>
<td>溼れ増倍率</td>
</tr>
</tbody>
</table>

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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\textsuperscript{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\textsuperscript{4}, its 2007 Addendum\textsuperscript{5} and Neutron Fluctuations\textsuperscript{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\textsuperscript{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\textsuperscript{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\textsuperscript{9} in 1985. Another viewpoint associated with $Y$-value was given by Croft\textsuperscript{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\textsuperscript{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 \((\alpha, n)\) reaction is a starting event. In this figure, expected number of spontaneous fission neutrons \(\nu_{s1}\) and expected number of induced fission neutrons \(\nu_{i1}\) are used where subscript ‘1’ indicates a singlet that means neutron is counted individually, whereas a doublet \((\nu_{s2}, \nu_{i2})\) 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 \((\alpha, n)\) reaction to the number from spontaneous fission \(\alpha_r\), leakage probability \(l\) and induced fission probability \(p\):

\[
\frac{M_{L} - 1}{\nu_{i1} - 1} = \frac{p}{1 - p \nu_{i1}} = p M_T, \quad \frac{M_{L} - 1}{\nu_{i1} - 1} = p, \quad \frac{M_{L} \nu_{i1} - 1}{\nu_{i1} - 1} = M_T
\]

are useful transformations.

Neutron leakage multiplication \(M_L\) and total multiplication \(M_T\) are defined as:

\[
M_L = l M_T = l \left[1 + p \nu_{i1} + (p \nu_{i1})^2 \ldots \right] = \frac{l}{1 - p \nu_{i1}} = \frac{1 - p - p_c}{1 - p \nu_{i1}} \approx \frac{1 - p}{1 - p \nu_{i1}}
\]

where \(p_c\) is neutron capture probability, ex. by \((n, \gamma)\) 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:

\[
G_h(z) = (1 - p) \sum_{n=1}^{\infty} z^n 1 + p \sum_{n=0}^{\infty} (G_h(z))^n P_{\nu_i} (\nu_i = n)
\]

where \(P_{\nu_i}\) 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 \(\nu_i\), 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’.

\[
G_h(z) = (1 - p) \sum_{n=1}^{\infty} z^n 1 + p \sum_{n=0}^{\infty} (G_h(z))^n P_{\nu_i} (\nu_i = n)
\]
The equation (2) is simplified to:

\[ G_h (z) = (1 - p) z + p G_i (G_h (z)) \]  

(3)

The \( k \)th derivatives are solved algebraically assisted by a computerb:

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

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 \( h(u), f(u), H(u), M_{(p)i} \rightarrow M_{(p)k} \) and \( f_s(u) \) corresponds respectively to \( G_h (z), G_i (z), G_H (z), \overline{\nu_k} \) and \( G_s (z) \) are used.
The following relations are applied later to the above equations:

\[ G_i^{(k)}|_{z=1} = \nu_i(\nu_i - 1)(\nu_i - 2) \cdots (\nu_i - k + 1) = \nu_{ik} \]  
\[ \frac{1-p}{1-pG_i} = \frac{1-p}{1-p\nu_{i1}} = M_L \]  
\[ \frac{p}{1-pG_i} = \frac{p}{1-p\nu_{i1}} = M_{L\nu} \]

2.3 Probability generating function considering the source of a neutron

The starting event of Figure 1 is a spontaneous fission or a \((\alpha, n)\) reaction. The PGF for a neutron is rewritten using \(G_s(z)\) for spontaneous fission where \(P\nu_s\) is used instead of \(P\nu_i\) as:

\[ G_H(z) = \frac{\alpha r\nu_{s1}}{1 + \alpha r\nu_{s1}} G_h(z) + \frac{1}{1 + \alpha r\nu_{s1}} G_s(G_h(z)) \]

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. \(\alpha r\nu_{s1}/(1 + \alpha r\nu_{s1})\) and \(1/(1 + \alpha r\nu_{s1})\) means probabilities of a neutron from a \((\alpha, n)\) reaction or a neutron from a spontaneous fission, because \((\alpha, n)\) reaction occurs \(\alpha r\nu_{s1}\) times more frequently than spontaneous fission. Same as the equation (5), the next transformation is applied later to the above equation:

\[ G_s^{(k)}|_{z=1} = \nu_s(\nu_s - 1)(\nu_s - 2) \cdots (\nu_s - k + 1) = \nu_{sk} \]

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 \((1-p)^k(1 + \alpha r\nu_{s1})^{-1}(1-p\nu_{i1})^{1-2k}\) 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 \((M_L - 1)/(\nu_{i1} - 1)\) 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:

\[ P_m(m=k) = (1 + \alpha r\nu_{s1}) \frac{G_s^{(k)}|_{z=0}}{k!} \]

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 \( M^m_L / m! \) same as the conventional equations. Terms in the enclosure are collected by \( \nu_{s1}, \nu_{s2}, \nu_{s3} \cdots \) followed by collected by \( M_{Lv}, M^2_{Lv}, M^3_{Lv} \cdots \). Obtained probability mass distribution of multiple leakage neutrons are shown below. There are another expressions in the Appendix d):

\[
P_m (m=1) = M_L (1 + \alpha_r) \nu_{s1}
\]

\[
P_m (m=2) = \frac{M^2_L}{2} [\nu_{s2} + \nu_{s2} M_{Lv} (1 + \alpha_r) \nu_{s1}]
\]

\[
P_m (m=3) = \frac{M^3_L}{6} [\nu_{s3} + 3 \nu_{s2} M_{Lv} \nu_{s2} + (\nu_{s3} M_{Lv} + 3 \nu_{s2}^2 M^2_{Lv}) (1 + \alpha_r) \nu_{s1}]
\]

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

\[
S = F_p m_{\text{eff}} \epsilon_n (1 + \alpha_r) \nu_{s1} M_L
\]

\[
D = \frac{F_p m_{\text{eff}} \epsilon_n^2 f_d}{2} \left[ \frac{\nu_{s2} + \nu_{s2} (1 + \alpha_r) \nu_{s1} M_L - 1}{\nu_{s1} - 1} \right] M^2_L
\]

\[
T = \frac{F_p m_{\text{eff}} \epsilon_n^3 f_t}{6} \left\{ \frac{\nu_{s3} + M_{Lv} - 1}{\nu_{s1} - 1} \left[ (1 + \alpha_r) \nu_{s1} \nu_{s3} + 3 \nu_{s2} \nu_{s2} \right] + 3 (1 + \alpha_r) \nu_{s1} \left( \frac{M_L - 1}{\nu_{s1} - 1} \right)^2 \nu_{s2}^2 \right\} M^3_L
\]

where \( \nu_{s8}^{\text{def}} = \max m \sum_{\nu_{s1}=m}^{\nu_{s8}} \frac{\nu_{s1}}{m} \) \( P_v = \nu_{s1} (\nu_{s1} - 1) (\nu_{s1} - 2) \cdots (\nu_{s1} - m + 1) \) (17)

Real count rates are equal to the product of probability mass distribution of multiple leakage neutrons and \( F_p m_{\text{eff}} \epsilon_n^m (1/f_d/f_t/\cdots/f_7) \) where \( F_p \) is the number of spontaneous fissions per unit mass and time, \( m_{\text{eff}} \) is effective mass of spontaneous fission nuclei (\(^{238}\text{Pu}, ^{240}\text{Pu} \) and \(^{242}\text{Pu} \) ), \( \epsilon_n \) 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:

\[
P_m (m=4) = \frac{M^4_L}{24} [\nu_{s4} + 6 \nu_{s2} M_{Lv} \nu_{s3} + 4 \nu_{s3} M_{Lv} + 15 \nu_{s2}^2 M^2_{Lv}] \nu_{s2} + \left( \nu_{s4} M_{Lv} + 10 \nu_{s3} \nu_{s2} M^2_{Lv} + 15 \nu_{s2}^3 M^3_{Lv} \right) (1 + \alpha_r) \nu_{s1}]
\]
\[ P_m (m=5) = \frac{M_L^5}{120} \{ \bar{\nu}_{s5} + 10 \bar{\nu}_{12} M_{L\nu} \bar{\nu}_{s4} \]
\[ + \left( 10 \bar{\nu}_{13} M_{L\nu} + 45 \bar{\nu}_{12}^2 M_{L\nu}^2 \right) \bar{\nu}_{s3} \]
\[ + \left( 5 \bar{\nu}_{14} M_{L\nu} + 60 \bar{\nu}_{13} \bar{\nu}_{12} M_{L\nu}^2 + 105 \bar{\nu}_{12}^3 M_{L\nu}^3 \right) \bar{\nu}_{s2} \]
\[ + \left[ \bar{\nu}_{15} M_{L\nu} + \left( 15 \bar{\nu}_{14} \bar{\nu}_{12} + 10 \bar{\nu}_{13}^2 \right) M_{L\nu}^2 \right. \]
\[ \left. + 105 \bar{\nu}_{13} \bar{\nu}_{12}^2 M_{L\nu}^3 + 105 \bar{\nu}_{12}^4 M_{L\nu}^4 \right] (1 + \alpha_r) \bar{\nu}_{s1} \} \]

\[ P_m (m=6) = \frac{M_L^6}{720} \{ \bar{\nu}_{s6} + 15 \bar{\nu}_{12} M_{L\nu} \bar{\nu}_{s5} \]
\[ + \left( 20 \bar{\nu}_{13} M_{L\nu} + 105 \bar{\nu}_{12}^2 M_{L\nu}^2 \right) \bar{\nu}_{s4} \]
\[ + \left( 15 \bar{\nu}_{14} M_{L\nu} + 210 \bar{\nu}_{13} \bar{\nu}_{12} M_{L\nu}^2 + 420 \bar{\nu}_{12}^3 M_{L\nu}^3 \right) \bar{\nu}_{s3} \]
\[ + \left[ 6 \bar{\nu}_{15} M_{L\nu} + \left( 105 \bar{\nu}_{14} \bar{\nu}_{12} + 70 \bar{\nu}_{13}^2 \right) M_{L\nu}^2 \right. \]
\[ \left. + 840 \bar{\nu}_{13} \bar{\nu}_{12}^2 M_{L\nu}^3 + 945 \bar{\nu}_{12}^4 M_{L\nu}^4 \right] \bar{\nu}_{s2} \]
\[ + \left[ \bar{\nu}_{16} M_{L\nu} + \left( 21 \bar{\nu}_{15} \bar{\nu}_{12} + 35 \bar{\nu}_{14} \bar{\nu}_{13} \right) M_{L\nu}^2 \right. \]
\[ \left. + \left( 210 \bar{\nu}_{14} \bar{\nu}_{12}^2 + 280 \bar{\nu}_{13}^2 \bar{\nu}_{12} \right) M_{L\nu}^3 \right. \]
\[ \left. + 1260 \bar{\nu}_{13} \bar{\nu}_{12}^2 M_{L\nu}^4 + 945 \bar{\nu}_{12}^5 M_{L\nu}^5 \right] (1 + \alpha_r) \bar{\nu}_{s1} \} \]

\[ P_m (m=7) = \frac{M_L^7}{5040} \{ \bar{\nu}_{s7} + 21 \bar{\nu}_{12} M_{L\nu} \bar{\nu}_{s6} \]
\[ + \left( 35 \bar{\nu}_{13} M_{L\nu} + 210 \bar{\nu}_{12}^2 M_{L\nu}^2 \right) \bar{\nu}_{s5} \]
\[ + \left( 35 \bar{\nu}_{14} M_{L\nu} + 560 \bar{\nu}_{13} \bar{\nu}_{12} M_{L\nu}^2 + 1260 \bar{\nu}_{12}^3 M_{L\nu}^3 \right) \bar{\nu}_{s4} \]
\[ + \left[ 21 \bar{\nu}_{15} M_{L\nu} + \left( 420 \bar{\nu}_{14} \bar{\nu}_{12} + 280 \bar{\nu}_{13}^2 \right) M_{L\nu}^2 \right. \]
\[ \left. + 3780 \bar{\nu}_{13} \bar{\nu}_{12}^2 M_{L\nu}^3 + 4725 \bar{\nu}_{12}^4 M_{L\nu}^4 \right] \bar{\nu}_{s3} \]
\[ + \left[ 7 \bar{\nu}_{16} M_{L\nu} + \left( 168 \bar{\nu}_{15} \bar{\nu}_{12} + 280 \bar{\nu}_{14} \bar{\nu}_{13} \right) M_{L\nu}^2 \right. \]
\[ \left. + \left( 1890 \bar{\nu}_{14} \bar{\nu}_{12}^2 + 2520 \bar{\nu}_{13}^2 \bar{\nu}_{12} \right) M_{L\nu}^3 \right. \]
\[ \left. + 1260 \bar{\nu}_{13} \bar{\nu}_{12}^2 M_{L\nu}^4 + 10395 \bar{\nu}_{12}^5 M_{L\nu}^5 \right] \bar{\nu}_{s2} \]
\[ + \left[ \bar{\nu}_{17} M_{L\nu} + \left( 28 \bar{v}_{16} \bar{\nu}_{12} + 56 \bar{\nu}_{15} \bar{\nu}_{13} + 35 \bar{\nu}_{14} \bar{\nu}_{12} \right) M_{L\nu}^2 \right. \]
\[ \left. + \left( 378 \bar{\nu}_{15} \bar{\nu}_{12}^2 + 1260 \bar{\nu}_{14} \bar{\nu}_{13} \bar{\nu}_{12} + 280 \bar{\nu}_{13}^3 \right) M_{L\nu}^3 \right. \]
\[ \left. + \left( 3150 \bar{\nu}_{14} \bar{\nu}_{12} \bar{\nu}_{13}^2 + 6300 \bar{\nu}_{13}^2 \bar{\nu}_{12}^2 \right) M_{L\nu}^4 \right. \]
\[ \left. + 17325 \bar{\nu}_{13} \bar{\nu}_{12}^2 M_{L\nu}^5 + 10395 \bar{\nu}_{12}^6 M_{L\nu}^6 \right] (1 + \alpha_r) \bar{\nu}_{s1} \} \]
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.

**Source of singlets, doublets and triplets**

If means induced fission

**Leakage from a triplet**

For each one leak

For no leak (two cases)

For triplet:

Evaluation of the chain of sources

Evaluation of the chain of sources

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 \( F_p \), \( \epsilon_n \), \( f_d \), \( f_t \cdot f_7^\cdot \cdots f_7 \) and \( \alpha_r \) 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 \( \nu_{\text{sm}} \), \( ^{240}\text{Pu} \) and \( ^{239}\text{Pu} \) (2 MeV neutron) were selected because of their large contribution on neutron yield. The \( \nu_{\text{sm}} \) were calculated using \( P_\nu \) consolidated/proposed by Zucker and Holden\(^{12,13,14}\) in 1984-1986. They are shown in Table 1 and 2:

Table 1 \( P_\nu \) of \( ^{240}\text{Pu} \) and \( ^{239}\text{Pu} \) (2 MeV neutron)

<table>
<thead>
<tr>
<th>( \nu )</th>
<th>( ^{240}\text{Pu} )</th>
<th>( ^{239}\text{Pu} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.0632</td>
<td>0.0063</td>
</tr>
<tr>
<td>1</td>
<td>0.2320</td>
<td>0.0612</td>
</tr>
<tr>
<td>2</td>
<td>0.3333</td>
<td>0.2266</td>
</tr>
<tr>
<td>3</td>
<td>0.2528</td>
<td>0.3261</td>
</tr>
<tr>
<td>4</td>
<td>0.0986</td>
<td>0.2588</td>
</tr>
<tr>
<td>5</td>
<td>0.0180</td>
<td>0.0956</td>
</tr>
<tr>
<td>6</td>
<td>0.0020</td>
<td>0.0225</td>
</tr>
<tr>
<td>7</td>
<td>0.00006†</td>
<td>0.0026</td>
</tr>
</tbody>
</table>

†. Estimated by Gaussian fit of the distribution based on Terrell\(^{15}\) in 1957.

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

<table>
<thead>
<tr>
<th>( m )</th>
<th>( \nu_{\text{sm}} )</th>
<th>( \nu_{\text{im}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2.1537</td>
<td>3.1591</td>
</tr>
<tr>
<td>2</td>
<td>3.7889</td>
<td>8.2116</td>
</tr>
<tr>
<td>3</td>
<td>5.2155</td>
<td>17.1498</td>
</tr>
<tr>
<td>4</td>
<td>5.2965</td>
<td>27.9672</td>
</tr>
<tr>
<td>5</td>
<td>3.7510</td>
<td>34.2240</td>
</tr>
<tr>
<td>6</td>
<td>1.7423</td>
<td>29.3040</td>
</tr>
<tr>
<td>7</td>
<td>0.3024</td>
<td>13.1040</td>
</tr>
</tbody>
</table>

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

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

<table>
<thead>
<tr>
<th>( m )</th>
<th>( \eta_{\text{sm}} )</th>
<th>( \eta_{\text{im}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>2.1537</td>
<td>3.1591</td>
</tr>
<tr>
<td>2</td>
<td>1.8944</td>
<td>4.1058</td>
</tr>
<tr>
<td>3</td>
<td>0.8692</td>
<td>2.8583</td>
</tr>
<tr>
<td>4</td>
<td>0.2207</td>
<td>1.1653</td>
</tr>
<tr>
<td>5</td>
<td>0.0313</td>
<td>0.2852</td>
</tr>
<tr>
<td>6</td>
<td>0.0024</td>
<td>0.0407</td>
</tr>
<tr>
<td>7</td>
<td>0.0001</td>
<td>0.0026</td>
</tr>
</tbody>
</table>

Calculated \( S/D/T/Qr\)/\( Qt\)/\( Sx\)/\( Sp \) 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 \( M_L > 1.3 \) (\( \epsilon_n = 0.4 \), \( f_d = 0.67 \)).

\[ c. \text{erf} x = \frac{2}{\sqrt{\pi}} \int_0^x \exp (-z^2) \, dz = \frac{2}{\sqrt{\pi}} \int_0^{\sqrt{x^2}} \exp \left( -\frac{t^2}{2} \right) \, dt \] is applied to

\[ P_\nu = \sum_{m=0}^\nu P_m \sum_{m=0}^{\nu-1} P_m = \frac{1}{\sqrt{2\pi}} \int_{\nu-0.5}^{\nu+0.5} \exp \left( -\frac{t^2}{2} \right) \, dt \]

then we get

\[ P_\nu = \frac{1}{2} \left[ \text{erf} \left( \frac{\nu-0.5}{\sigma\sqrt{2}} \right) - \text{erf} \left( \frac{\nu-0.5}{\sigma\sqrt{2}} \right) \right] \]

where \( \nu \) and \( \sigma \) are mean and width of Gaussian distribution fit.

The width is given as 1.08 \pm 0.01 by Terrell (ref.15) for all nuclei except for \( ^{252}\text{Cf} \), 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 \( ^{240}\text{Pu} \) are 2.092 and 1.150, respectively. This mean is not equal to \( \nu_{\text{sm}} \) (2.1537 for \( ^{239}\text{Pu} \)).
Table 4 $S/D/T/Q_r/Q_t/S_x/S_p$ for one gram spontaneous fission source

($\epsilon_n = 0.4$, $\alpha_r = 0.7$, $f_d = 0.67$)

<table>
<thead>
<tr>
<th>$M_L$</th>
<th>$S$ ($m = 1$)</th>
<th>$D$ ($m = 2$)</th>
<th>$T$ ($m = 3$)</th>
<th>$Q_r$ ($m = 4$)</th>
<th>$Q_t$ ($m = 5$)</th>
<th>$S_x$ ($m = 6$)</th>
<th>$S_p$ ($m = 7$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.0</td>
<td>694</td>
<td>96.3</td>
<td>11.8</td>
<td>0.806</td>
<td>0.0306</td>
<td>0.0064</td>
<td>0.000004</td>
</tr>
<tr>
<td>1.1</td>
<td>764</td>
<td>159</td>
<td>42.4</td>
<td>12.5</td>
<td>4.23</td>
<td>1.54</td>
<td>0.603</td>
</tr>
<tr>
<td>1.2</td>
<td>833</td>
<td>241</td>
<td>102</td>
<td>51.7</td>
<td>29.5</td>
<td>18.1</td>
<td>11.7</td>
</tr>
<tr>
<td>1.3</td>
<td>902</td>
<td>342</td>
<td>205</td>
<td>150</td>
<td>124</td>
<td>109</td>
<td>101</td>
</tr>
<tr>
<td>1.4</td>
<td>972</td>
<td>466</td>
<td>371</td>
<td>363</td>
<td>399</td>
<td>470</td>
<td>579</td>
</tr>
<tr>
<td>1.5</td>
<td>1,041</td>
<td>615</td>
<td>621</td>
<td>776</td>
<td>1,087</td>
<td>1,632</td>
<td>2,563</td>
</tr>
<tr>
<td>1.6</td>
<td>1,111</td>
<td>790</td>
<td>984</td>
<td>1,519</td>
<td>2,628</td>
<td>4,871</td>
<td>9,450</td>
</tr>
</tbody>
</table>

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 $M_L$. 

- 9 -
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 $\epsilon_n$ and $f_d$ ($f_1 \cdots f_7$ are supposed to be $f_d^n$), 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 $D/T/Q_r/Q_t/S_x/S_p$ ($\epsilon_n = 0.4, \alpha_r = 0.7, f_d = 0.67$)

Figure 4b Crossing of $D/T/Q_r/Q_t/S_x/S_p$ ($\epsilon_n = 0.4, \alpha_r = 0.7, f_d = 0.85$)
In contrast, $\alpha_r$ 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\textsuperscript{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 $m_{\text{eff}} = 40\sim 50$ g is equivalent to $M_L = 1.4$ at the crossing point. Quantitative analysis is possible\textsuperscript{15} but it is not so familiar.

\begin{figure}[htp]
\centering
\includegraphics[width=0.5\textwidth]{figure5a.png}
\caption{Crossing of $D/T/Q_r$ (reprinted from Fig.1 of LA-UR-97-2716)\textsuperscript{16}.}
\end{figure}

\begin{figure}[htp]
\centering
\includegraphics[width=0.5\textwidth]{figure5b.png}
\caption{Crossing of $D/T/Q_r/Q_t/S_x/S_p$ ($\epsilon_n = 0.5$, $\alpha_r = 0$, $f_d = 0.67$)}
\end{figure}

d. The ratio of fast fission cross section to total cross section is estimated to be 0.2\sim 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\sim 0.602. By the way, mean free path of 2 MeV neutron is $\sim 2.77$ cm. From $P_E$ and the path, radius of Pu metal sphere is estimated to be 1.56\sim 1.80 cm. Estimated weight is 315\sim 483 g and estimated $m_{\text{eff}}$ is 30\sim 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 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 and published 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 $\gamma$-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 where interarrival or waiting times of transition of an event distributes exponentially. 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 \( \lambda \) is probabilities of various reactions in the flow for a neutron for a unit time, \( \nu \) is the number of neutrons from a fission, \( p_\nu, f_1^\nu, f_2^\nu \) are probability distributions (spontaneous, induced by fast and by thermal, respectively) and \( S_1 \) is the number of spontaneous fissions for a unit time.

\[
\begin{align*}
\lambda_{1a} &\quad \text{Absorbed/Transmute/Escape} \\
\lambda_{T2} &\quad \text{Detector} \\
\lambda_d &\quad \text{Absorbed/Transmute/Escape} \\
\end{align*}
\]

\[
\begin{align*}
\lambda_{1f} &\quad \text{Induced fission} \\
\lambda_2 &\quad \text{Fast neutron} \\
\lambda_{2f} &\quad \text{Detected neutron} \\
\end{align*}
\]

\[
\begin{align*}
\frac{(\lambda_{1a}+\lambda_{T2}+\lambda_d+\lambda_{1f})N_1+(\lambda_{2a}+\lambda_{T1}+\lambda_{2f})N_2+S_1}{ \text{P}(N_1,N_2,Z_1,t) \quad \text{Steady state through fluctuations} \quad \text{P}(N_1,N_2,Z_1,t+\text{dt})} \\
\end{align*}
\]

- Absorption of a fast neutron
- Thermal neutron \( \rightarrow \) Fast neutron
- Fast neutron \( \rightarrow \) Detected neutron
- Absorption of a thermal neutron
- Fast neutron \( \rightarrow \) Thermal neutron
- Induced fission by a fast neutron
- Induced fission by a thermal neutron
- Spontaneous fission

---

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):

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

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 \((N_1 + 1)P(N_1 + 1, N_2, Z_1, t), \ldots\) are given as:

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

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

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

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

\[
Z \frac{\partial G}{\partial X} = \sum_{N_1} \sum_{N_2} \sum_{Z_1} X^{N_1} Y^{N_2} Z^{Z_1} (N_1 + 1) P(N_1 + 1, N_2, Z_1 - 1, t) \tag{29}
\]
\[ X^\nu \frac{\partial G}{\partial Y} = \sum_{N_1} \sum_{N_2} \sum_{Z_1} X^{N_1} Y^{N_2} Z^{Z_1} (N_2 + 1) P(N_1 - \nu, N_2 + 1, Z_1, t) \]  
\[ X^\nu \frac{\partial G}{\partial X} = \sum_{N_1} \sum_{N_2} \sum_{Z_1} X^{N_1} Y^{N_2} Z^{Z_1} (N_1 + 1 - \nu) P(N_1 + 1 - \nu, N_2 + 1, Z_1, t) \]  
\[ \sum_\nu f_\nu^{2} X^\nu \frac{\partial G}{\partial Y} = \sum_{N_1} \sum_{N_2} \sum_{Z_1} X^{N_1} Y^{N_2} Z^{Z_1} (N_2 + 1) f_\nu^{2} P(N_1 - \nu, N_2 + 1, Z_1, t) \]  
\[ \sum_\nu f_\nu^{1} X^\nu \frac{\partial G}{\partial X} = \sum_{N_1} \sum_{N_2} \sum_{Z_1} X^{N_1} Y^{N_2} Z^{Z_1} (N_1 + 1 - \nu) f_\nu^{1} P(N_1 + 1 - \nu, N_2, Z_1, t) \]  
\[ \sum_\nu p_\nu X^\nu G = \sum_{N_1} \sum_{N_2} \sum_{Z_1} X^{N_1} Y^{N_2} Z^{Z_1} p_\nu P(N_1 - \nu, N_2, Z_1, t) \]

### 3.3 Solving the equation using probability generating function

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

\[ \frac{\partial G}{\partial t} = (\lambda_1 a + \lambda T_2 Y + \lambda d Z + \lambda_1 f \sum_\nu f_\nu^{1} X^\nu) \frac{\partial G}{\partial X} \]
\[ + (\lambda_2 a + \lambda T_1 X + \lambda_2 f \sum_\nu f_\nu^{2} X^\nu) \frac{\partial G}{\partial Y} + S_1 \sum_\nu p_\nu X^\nu G \]
\[ - (\lambda_1 a + \lambda T_2 + \lambda d + \lambda_1 f) X \frac{\partial G}{\partial X} - (\lambda_2 a + \lambda T_1 + \lambda_2 f) Y \frac{\partial G}{\partial Y} - S_1 G \]

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

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

\[ \frac{\partial G}{\partial X} \bigg|_{X=1} = N_1 \quad \text{Expected number of fast neutron} \]

\[ \frac{\partial G}{\partial Y} \bigg|_{Y=1} = N_2 \quad \text{Expected number of thermal neutron} \]

\[ \frac{d}{dX} \sum_\nu f_\nu^{1} X^\nu \bigg|_{X=1} = \nu^{1}_{11} \quad \text{1st factorial moment of induced fission by fast neutron} \]

\[ \frac{d}{dX} \sum_\nu f_\nu^{2} X^\nu \bigg|_{X=1} = \nu^{2}_{11} \quad \text{1st factorial moment of induced fission by thermal neutron} \]

\[ \frac{d}{dX} \sum_\nu p_\nu X^\nu \bigg|_{X=1} = \nu^{s1} \quad \text{1st factorial moment of spontaneous fission} \]
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:

$$\overline{N}_1 = \frac{\lambda_2 S_1 \bar{\nu}_{s1}}{\lambda_1 \lambda_2 - \lambda_1 f \lambda_2 \bar{\nu}_{i1}^2 - \lambda_{T1} \lambda_{T2}}$$  \hfill (42)

$$\overline{N}_2 = \frac{\lambda_{T2} S_1 \bar{\nu}_{s1}}{\lambda_1 \lambda_2 - \lambda_1 f \lambda_2 \bar{\nu}_{i1}^2 - \lambda_{T1} \lambda_{T2}}$$  \hfill (43)

$$\overline{Z}_1 = \lambda_d \overline{N}_1 t$$  \hfill (44)

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

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):

$$\frac{d^2}{dX^2} \sum_{\nu} f_\nu^1 X^\nu \bigg|_{X=1} = \bar{\nu}_{i1}^1 (\nu_{i1}^1 - 1) = \bar{\nu}_{i2}^1$$  \hfill (45)

2nd factorial moment of induced fission by fast neutron

$$\frac{d^2}{dX^2} \sum_{\nu} f_\nu^2 X^\nu \bigg|_{X=1} = \bar{\nu}_{i2}^2 (\nu_{i2}^2 - 1) = \bar{\nu}_{i2}^2$$  \hfill (46)

2nd factorial moment of induced fission by thermal neutron

$$\frac{d^2}{dX^2} \sum_{\nu} \rho_\nu X^\nu \bigg|_{X=1} = \bar{\nu}_{s2} (\nu_{s2} - 1) = \bar{\nu}_{s2}$$  \hfill (47)

2nd factorial moment of spontaneous fission

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 $\mu_{XX}, \mu_{YY}, \mu_{ZZ}, \mu_{XY}, \mu_{XZ}, \mu_{YZ}$, 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:

$$\frac{\partial}{\partial t} \mu_{XX} = 2 (\lambda_1 + \lambda_1 f \bar{\nu}_{i1}^1) \mu_{XX} + 2 (\lambda_{T1} + \lambda_2 f \bar{\nu}_{i1}^2) \mu_{XY} + \lambda_1 f \bar{\nu}_{i2}^1 \overline{N}_1 + \lambda_2 f \bar{\nu}_{i2}^2 \overline{N}_2 + S_1 \bar{\nu}_{s2}$$  \hfill (48)

$$\frac{\partial}{\partial t} \mu_{YY} = -2 \lambda_2 \mu_{YY} + 2 \lambda_{T2} \mu_{XY}$$  \hfill (49)

$$\frac{\partial}{\partial t} \mu_{ZZ} = 2 \lambda_d \mu_{XZ}$$  \hfill (50)
\[
\frac{\partial}{\partial t} \mu_{XY} = -\lambda_1 - \lambda_2 + \lambda_1 f \nu_1^T \mu_{XY} + \lambda_{T2} \mu_{XX} + (\lambda_{T1} + \lambda_2 f \nu_1^T) \mu_{YY}
\]  
\[
\frac{\partial}{\partial t} \mu_{XZ} = -\lambda_1 + \lambda_1 f \nu_1^T \mu_{XZ} + \lambda_d \mu_{XX} + (\lambda_{T1} + \lambda_2 f \nu_1^T) \mu_{YZ}
\]  
\[
\frac{\partial}{\partial t} \mu_{YZ} = -\lambda_2 \mu_{YZ} + \lambda_d \mu_{XY} + \lambda_{T2} \mu_{XZ}
\]

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:

\[
\mu_{XX} = \frac{(\omega_1 \omega_2 + \lambda_2^2)(\lambda_1 f \nu_1^T N_1 + \lambda_2 f \nu_2^T N_2 + S_1 \nu_{s2})}{2(\lambda_1 + \lambda_2 - \lambda_1 f \nu_1^T) \omega_1 \omega_2}
\]  
\[
\mu_{YY} = \frac{\lambda_2^2}{\omega_1 \omega_2 + \lambda_2^2} \mu_{XX}
\]  
\[
\mu_{XY} = \frac{\lambda_2 \lambda_{T2}}{\omega_1 \omega_2 + \lambda_2^2} \mu_{XX}
\]  
\[
\omega_1 \omega_2 = \lambda_1 \lambda_2 - \lambda_1 f \nu_1^T - \lambda_2 f \nu_2^T - \lambda_{T1} \lambda_{T2}
\]

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 \( \lambda_1 f \rightarrow 0 \) because induced fission by fast neutron is not considered in ref.19, \( \lambda_{T1} \rightarrow 0 \) because probability of thermal to fast is little (only by delayed fission) and \( \lambda_{T2} \rightarrow \lambda_R \) because \( \lambda_R \) is used instead of \( \lambda_{T2} \). The results below are obtained:

\[
\mu_{XZ} = \frac{\lambda_d (\lambda_{T1} + \lambda_2 f \nu_1^T)}{\omega_1 \omega_2} \mu_{XY} + \frac{\lambda_d \lambda_2}{\omega_1 \omega_2} \mu_{XX}
\]  
\[
= \frac{\lambda_d \lambda_2^2}{2 \omega_1^2 \omega_2} (\lambda_1 f \nu_1^T N_1 + \lambda_2 f \nu_2^T N_2 + S_1 \nu_{s2})
\]
\[
\mu_{ZZ} = \frac{\lambda_2^2}{\omega_1^2 \omega_2} (\lambda_1 f \nu_1^T N_1 + \lambda_2 f \nu_2^T N_2 + S_1 \nu_{s2}) t
\]

Here, a measure is introduced to eliminate \( t \):

\[
\frac{\mu_{ZZ}}{Z_1} = \frac{\lambda_d \lambda_2^2}{\omega_1^2 \omega_2} (\lambda_1 f \nu_1^T N_1 + \lambda_2 f \nu_2^T N_2 + S_1 \nu_{s2} N_1)
\]

The measure is modified using \( \epsilon = \lambda_d / \lambda_1 f \) (counts per fast fission) as:

\[
\frac{\mu_{ZZ}}{Z_1} = \epsilon \left( \frac{\lambda_2 f}{\omega_1 \omega_2} \right)^2 (\nu_1^T + \frac{\lambda_2 f}{\lambda_1 f} N_1 \nu_2^T \nu_{s2} + \frac{S_1}{N_1} \nu_{s2})
\]

It should be noted that the measure is proportional to the linear coupling of \( \nu_1^T, \nu_2^T \) and \( \nu_{s2} \), 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 in 1949 is defined as:

\[
\frac{\bar{c}^2 - (\bar{c})^2}{\bar{c}} \overset{\text{def}}{=} 1 + Y
\]

(62)

\[
Y = \frac{\epsilon (\nu^2 - \nu)}{(\alpha \tau)^2} = \frac{\epsilon \nu (\nu - 1)}{(\alpha \tau)^2} = \epsilon \left( \frac{1}{\alpha \tau} \right)^2 \nu_2^2
\]

(63)

where \(c\) is count rate, \(\tau\) is mean time between fissions, \(\epsilon\) is efficiency (counts/fission), \(\nu_2\) is 2nd factorial moment of induced fission and \(\alpha\) 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 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:

\[
\frac{\mu_{ZZ}}{Z_1} = Y
\]

(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):

\[
D = \frac{1}{2} f_d S Y
\]

(65)

\[
D \frac{f_d}{f_d} = \frac{F \epsilon^2 (\nu^2 - \nu)}{2 \alpha^2 \tau^2} \left( 1 - \frac{1 - e^{-\alpha t}}{\alpha t} \right)
\]

(66)

\[
S = F \epsilon \frac{Z_1}{t} = \lambda_d \bar{N}_1
\]

(67)

\[
Y = \frac{\epsilon (\nu^2 - \nu)}{(\alpha \tau)^2} \left( 1 - \frac{1 - e^{-\alpha t}}{\alpha t} \right)
\]

(68)

For reference, ‘variance to mean’ in ref.19 and ref.20 is equal to variance of \(Z\) (not \(\mu_{ZZ}\) but \(\sigma_{ZZ}^2\)) 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 \( \left( 1 - \frac{1 - e^{-\alpha t}}{\alpha t} \right) \) 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 \( \nu/(\alpha \tau) \) 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:

\[
\frac{D_f}{f_d} = \frac{\mu_{Z'Z}}{2t} = \frac{1}{2} \left[ \frac{\lambda_2 \lambda_d^2}{\omega_1^2 \omega_2^2} \left( \frac{N_1}{N_2} \right)^2 \left( F_1 \nu_{i1}^2 + F_2 \nu_{i2}^2 + S_1 \nu_{s2} \right) \right]
\]

(69)

where the physical meaning of \( \lambda_2 \) is given by Figure 6 as a probability of input thermal neutron based on \( \bar{N}_2 \):

\[
\lambda_2 \bar{N}_2 = \lambda_1 \bar{N}_1 \rightarrow \lambda_2 = \frac{N_1}{N_2} \lambda_1 \lambda_2
\]

(70)

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

\[
\frac{D_f}{f_d} = \frac{1}{2} \frac{\lambda_d^2}{\omega_1^2} \frac{\lambda_2}{\omega_2^2} \left( \frac{N_1}{N_2} \right)^2 \left( F_1 \nu_{i1}^2 + F_2 \nu_{i2}^2 + S_1 \nu_{s2} \right)
\]

(71)

Square of [Efficiency (counts/neutron) × leakage mulplication] Sum of fission rate × 2nd factorial moment that means twice of the number of neutron pairs

also:

\[
S = \lambda_d \frac{\lambda_2}{\omega_1} \frac{\bar{N}_1}{\bar{N}_2} \bar{S}_1 \nu_{s1} = \left[ S \neq \lambda_d \frac{\lambda_2}{\omega_1} \frac{\bar{N}_1}{\bar{N}_2} \left( F_1 \nu_{i1}^2 + F_2 \nu_{i2}^2 + S_1 \nu_{s2} \right) \right]
\]

(72)

and:

\[
\frac{\lambda_d \lambda_2}{\omega_1 \omega_2} = \lambda_d \frac{\lambda_2}{\omega_1} \frac{\bar{N}_1}{\bar{N}_2} = \epsilon_n \epsilon_M
\]

(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 \( \nu_{i1}^2, \nu_{i1}^2 \) and \( \nu_{s1} \), 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:

\[
\frac{D}{f_d} = \frac{1}{2} \epsilon_n^2 M_L^2 \left( S_1 \nu_{s1} + \overline{F}_1 \nu_{i1} + \overline{F}_2 \nu_{i2}^2 \right) \tag{74}
\]

\[
S = \epsilon_n M_L S_1 \nu_{s1} \tag{75}
\]

To separate \( \epsilon_n \) and \( M_L \), physical meaning of \( \epsilon_n \) is considered, and \( \epsilon_n \) is given as:

\[
\epsilon_n = \frac{\lambda_d}{\lambda_{1f}} \frac{\lambda_d \lambda_2}{\lambda_1 \lambda_2 - \lambda_{T1} \lambda_{T2}} \tag{76}
\]

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

\[
\epsilon_n = \frac{\lambda_d}{\lambda_1} \tag{77}
\]

\[
M_L = \frac{\lambda_1 \lambda_2}{\omega_1 \omega_2} = 1 + \frac{\lambda_2 \lambda_{1f} \nu_{i1}^2 + \lambda_{T2} \lambda_{2f} \nu_{i2}^2}{\omega_1 \omega_2} \geq 1 \tag{78}
\]

The important equation below regarding \( M_L \) is obtained:

\[
M_L = \frac{1}{S_1 \nu_{s1}} \left( S_1 \nu_{s1} + \overline{F}_1 \nu_{i1} + \overline{F}_2 \nu_{i1}^2 \right) \tag{79}
\]

\[
\therefore \omega_1 \omega_2 = \frac{\lambda_{T2} S_1 \nu_{s1}}{N_2} = \frac{\lambda_2 S_1 \nu_{s1}}{N_1} \tag{80}
\]

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 \( \alpha \) in \( Y \)-value. From the similarity between equation (61) and (63), equations below are derived for induced fission by fast neutron:

\[
\frac{1}{\alpha \tau} = \frac{\lambda_2 \lambda_{1f}}{\omega_1 \omega_2} \tag{81}
\]

\[
S = \epsilon \frac{1}{\alpha \tau} S_1 \nu_{s1} \rightarrow \frac{1}{\alpha \tau} = \frac{\epsilon_n}{\epsilon} M_L \tag{82}
\]

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 $\nu_{i1}$, $\nu_{i2}$, and $\nu_{s1}$. 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. $N_1$ and $N_2$ are at steady state as a result of leakage, so $F_1$ and $F_2$ 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 $N_1$ and on $N_2$ are defined:

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

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

$$\mu ZZ Z_1^{-1} = \epsilon_1 \left( \frac{\lambda_2 \lambda_{1f}}{\omega_1 \omega_2} \right)^2 \left( \frac{\nu_{i2}}{F_1} + \frac{S_1}{F_1} \nu_{s2} \right) + \epsilon_2 \left( \frac{\lambda_2 \lambda_{2f}}{\omega_1 \omega_2} \frac{N_2}{N_1} \right)^2 \frac{\nu_{i2}}{N_1} \quad \text{for } Y_1 \text{ based on } N_1 \tag{85}$$

$$\mu ZZ Z_2^{-1} = \frac{1}{\alpha_1 \tau_1} \quad \text{for } Y_1 \tag{86}$$

$$\mu ZZ Z_2^{-1} = \frac{\omega_1 \omega_2}{\lambda_{1f} \lambda_2} \quad \text{for } Y_1 \tag{87}$$

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 $\lambda T_1 \to 0$:

$$\alpha_1 \tau_1 = \frac{1}{F_1} \left[ \frac{\lambda_1}{\lambda_{1f}} F_1 - F_1 \nu_{i1} - F_2 \nu_{11}^2 \right]$$

(88)

$$\alpha_2 \tau_2 = \frac{1}{F_2} \left[ \frac{\lambda_1}{\lambda_{1f}} F_1 - F_1 \nu_{i1} - F_2 \nu_{11}^2 \right]$$

(89)

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

$$F_1 = \frac{r}{\alpha_1 \tau_1}$$

(90)

$$F_2 = \frac{r}{\alpha_2 \tau_2}$$

(91)

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

(92)

$$r = \frac{\lambda_1}{\lambda_{1f}} \frac{F_1 - F_1 \nu_{i1}^1 - F_2 \nu_{i1}^2}{\nu_{i1}^2} = \lambda_1 N_1 - \left( F_1 \nu_{i1}^1 + F_2 \nu_{i1}^2 \right)$$

(93)

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

(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 $F_1$ is ‘net’ number of fast induced fissions in the system affected by thermal induced fission and leakage shown in Figure 6. $F_1$ 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 $r$:

$$Y_1 = \frac{1}{r} \epsilon_n M_L \left( F_1 \nu_{i2}^1 + S_1 \nu_{i2}^2 \right) = \frac{1}{r} \epsilon_1 \frac{1}{\alpha_1 \tau_1} \left( F_1 \nu_{i2}^1 + S_1 \nu_{i2}^2 \right)$$

(95)

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

(96)

Equation (95) and (96) satisfies the relation $\frac{\mu ZZ}{Z_1} = Y = Y_1 + Y_2$. These equations are useful to obtain $\epsilon_1$ and $\epsilon_2$ 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 $\lambda(s)$. 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 $\lambda(s)$.

$$\frac{D_1}{f_d} = \frac{1}{2} \frac{\lambda^2 \omega^2}{\omega^2_1 \omega^2_2} (\overline{F_1 \nu_{i2}} + S_1 \nu_{s2})$$  \hspace{1cm} (97)

$$\frac{D_2}{f_d} = \frac{1}{2} \frac{\lambda^2 \omega^2}{\omega^2_1 \omega^2_2} \overline{F_2 \nu_{i2}}$$  \hspace{1cm} (98)

$$\frac{D_1}{D_2} = \frac{\overline{F_1 \nu_{i2}}}{\overline{F_2 \nu_{i2}}} + \frac{S_1 \nu_{s2}}{\overline{F_2 \nu_{i2}}}$$  \hspace{1cm} (99)

From equation (92):

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

Therefore, the number of induced fissions $F_1$ and $F_2$ 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:

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

$$\overline{F_2} = S_1 \frac{\nu_{i2}^2 \nu_{s1} (M_L - 1) + \nu_{i1}^2 \nu_{s2}}{(D_1/D_2) \nu_{i1}^2 \nu_{i2}^2 + \nu_{i1}^2 \nu_{s2}}$$  \hspace{1cm} (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 $r = S_1 \nu_{s1}$ and equation (90) and (91). Consistency between equations (65), (75), (95) and (95) is proved from equation (97), (98) and (73):

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

$$Y_2 = \frac{1}{r} \epsilon_n M_L \overline{F_2 \nu_{i2}} = \frac{1}{r} \frac{2}{\epsilon_n M_L} \frac{D_2}{f_d} = \frac{2}{S} \frac{D_2}{f_d}$$  \hspace{1cm} (104)

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

- 24 -
Thus, $Y_1$ and $Y_2$ are obtained from $Y = (2D)/(S_f d)$ and $(D_1/D_2)$ as:

$$Y_1 = Y \frac{1}{1 + D_2/D_1} \quad (105)$$

$$Y_2 = Y \frac{1}{1 + D_1/D_2} \quad (106)$$

Therefore, $\epsilon_1$ and $\epsilon_2$ are written also using $Y_1$ and $Y_2$:

$$\epsilon_1 = \alpha_1 \tau_1 \epsilon_n \bar{M}_L = \alpha_1 \tau_1 Y_1 \frac{S_1 \nu s_1}{F_1 \nu s_1^2 + S_1 \nu s_2} \quad (107)$$

$$\epsilon_2 = \alpha_2 \tau_2 \epsilon_n \bar{M}_L = \alpha_2 \tau_2 Y_2 \frac{S_1 \nu s_1}{F_2 \nu s_2^2} \quad (108)$$

These equations satisfies the relation $\epsilon_1/\epsilon_2 = F_2/F_1 = (\alpha_1 \tau_1)/(\alpha_2 \tau_2)$. It is not necessary that both $\epsilon_1$ and $\epsilon_2$ are $< 1$ (counts/fission) whereas $\epsilon_n < 1$ (counts/neutron).

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

It became clear that the hypothetical stochastic variable $\frac{\mu_{ZZ}}{Z_1}$ 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:

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

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

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

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 $\frac{\mu_{ZZ}}{Z_1} = Y$ is written as:

$$Y = \frac{\mu_{ZZ}}{Z_1} = \frac{2 \lambda_d \mu_{XZ} t}{\lambda_d N_1 t} = \frac{2 \mu_{XZ}}{N_1} \quad (111)$$

Therefore, it is necessary to prove the following equation:

$$\mu_{XZ} = \frac{\lambda_d}{2} \left[ N_1^2 - (N_1)^2 - N_1/\lambda_d \right] \quad (112)$$
The proof is given as:

\[ \frac{N_1^2 - (N_1)^2}{\lambda_d} - \frac{N_1}{\lambda_d} = Y \frac{N_1}{\lambda_d} = \frac{YS}{\lambda_d^2} \]  

(113)

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

(114)

\[ YS \left( \frac{1}{\epsilon_n M_L} \right)^2 = \frac{2D}{f_d} \left( \frac{1}{\epsilon_n M_L} \right)^2 = \lambda_1 \nu_{i1} + \lambda_2 \nu_{i2} N_2 + S_1 v_{s2} \]  

(115)

\[ \mu_{XZ} = \frac{\lambda_d \lambda_2^2}{2 \omega_1^2 \omega_2} \left( \lambda_1 \nu_{i1} N_1 + \lambda_2 \nu_{i2} N_2 + S_1 v_{s2} \right) \]  

from equation (58)

(116)

\[ \therefore \mu_{XZ} = \frac{\lambda_d}{2} \left[ \frac{N_1^2}{\lambda_d} - \frac{(N_1)^2}{\lambda_d} \right] \]  

(117)

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

\[ \epsilon_1 = \frac{S}{F_1} = \alpha_1 \tau_1 \epsilon_n M_L \]  

(118)

\[ \epsilon_2 = \frac{S}{F_2} = \alpha_2 \tau_2 \epsilon_n M_L \]  

(119)

### 3.9 Evaluating decay constants by fast neutron and by thermal neutron

The decay constants by fast neutron \( \alpha_1 \) and by thermal neutron \( \alpha_2 \) 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 \( \alpha_1 \) and the latter reflects \( \alpha_2 \). The decay constants \( \alpha_1 \) and \( \alpha_2 \) are given by:

\[ \alpha_1 = r \left( 1 + \frac{S_1}{F_1} \right) \therefore \tau_1 = \frac{1}{S_1 + F_1} \]  

(120)

\[ \alpha_2 = r \therefore \tau_2 = \frac{1}{F_2} \]  

(121)

\[ \frac{1}{\alpha_2} = \frac{1}{\alpha_1} \left( 1 + \frac{S_1}{F_1} \right) > \frac{1}{\alpha_1} \]  

(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 \( \epsilon_n \) (counts/neutron) used for conventional NDAs and safeguards and doubles gate fraction \( f_d \);

2. Select representative isotopes (ex. \( \text{nat}^{238}\text{U} \) for fast induced fission, \( \text{nat}^{235}\text{U} \) for thermal induced fission and \( \text{nat}^{244}\text{Cm} \) for spontaneous fission) or a linear coupling of isotopes (ex. weighed \( \text{nat}^{235}\text{U}, \text{nat}^{239}\text{Pu} \) and \( \text{nat}^{241}\text{Pu} \) for thermal induced fission);

3. Determine \( \nu_{s1}, \nu_{s2}, \nu_{i1}^1, \nu_{i1}^2, \nu_{i2}^1, \nu_{i2}^2 \);

4. Measure \( S, D_1/D_2 \) and \( M_L \);

5. Evaluate \( S_1 = S/(\epsilon_n M_L) \) from equation (75);

6. Evaluate \( F_1 \) and \( F_2 \) from equations (101) and (102);

7. Confirm \( F_1 \) and \( F_2 \) 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 \( D_1/f_d \):

\[
\frac{D_1}{f_d} = \frac{1}{2} \epsilon_n^2 M_L^2 (F_1 \nu_{i1}^2 + S_1 \nu_{s2}) = \frac{SY_1}{2} \tag{123}
\]

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

\[
\frac{D_2}{f_d} = \frac{1}{2} \epsilon_n^2 M_L^2 F_2 \nu_{i2}^2 = \frac{SY_2}{2} \tag{124}
\]

3. Measure \( D \) and confirm \( D/f_d = D_1/f_d + D_2/f_d \).

From the viewpoint of safeguards, \( F_1 \) and \( F_2 \) 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 \( ^{10}\text{B} \).
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>
<thead>
<tr>
<th>Isotope</th>
<th>Fission yield [s$^{-1}$g$^{-1}$]</th>
<th>$\nu_{s1}$</th>
<th>Neutron yield [s$^{-1}$g$^{-1}$]</th>
<th>Mass in core (ref.23) [g]</th>
<th>Relative strength</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{238}$U</td>
<td>6.78E-3</td>
<td>1.99</td>
<td>1.35E-2</td>
<td>6.53E+7</td>
<td>0.01</td>
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<tr>
<td>$^{239}$Pu</td>
<td>1.17E+3</td>
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<td>2.56E+3</td>
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<td>0.27</td>
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<tr>
<td>$^{240}$Pu</td>
<td>4.83E+2</td>
<td>2.15</td>
<td>1.04E+3</td>
<td>1.05E+5</td>
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<tr>
<td>$^{242}$Pu</td>
<td>8.07E+2</td>
<td>2.15</td>
<td>1.74E+3</td>
<td>2.02E+4</td>
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</tr>
<tr>
<td>$^{242}$Cm</td>
<td>7.81E+6</td>
<td>2.54</td>
<td>1.98E+7</td>
<td>1.49E-1</td>
<td>0.04</td>
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<tr>
<td>$^{244}$Cm</td>
<td>4.11E+6</td>
<td>2.72</td>
<td>1.12E+7</td>
<td>6.17E+2</td>
<td>97.61</td>
</tr>
<tr>
<td>$^{246}$Cm</td>
<td>2.97E+6</td>
<td>2.93</td>
<td>8.70E+6</td>
<td>~2.54E-1$^+$</td>
<td>0.03</td>
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</table>

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

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Cross-section [barn]</th>
<th>at 2 MeV</th>
<th>Neutron yield [rel.]</th>
<th>Mass in core (ref.23) [g]</th>
<th>Relative strength</th>
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<tr>
<td>$^{238}$U</td>
<td>0.534E+0</td>
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<td>$^{239}$U</td>
<td>1.29E+0</td>
<td>2.64</td>
<td>3.41E+0</td>
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<td>$^{239}$Pu</td>
<td>1.98E+0</td>
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<td>6.26E+0</td>
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<td>$^{240}$U</td>
<td>0.819E+0</td>
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<tr>
<td>$^{241}$Am</td>
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<td>3.40</td>
<td>6.43E+0</td>
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<table>
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<th>Isotope</th>
<th>Cross-section [barn]</th>
<th>at thermal</th>
<th>Neutron yield [rel.]</th>
<th>Mass in core (ref.23) [g]</th>
<th>Relative strength</th>
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<td>1.41E+3</td>
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<td>$^{237}$Np$^+$</td>
<td>2.04E-2</td>
<td>2.63</td>
<td>5.37E-2</td>
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<tr>
<td>$^{239}$Pu</td>
<td>1.70E+1</td>
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<td>7.48E+2</td>
<td>2.88</td>
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<td>28.41</td>
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<tr>
<td>$^{241}$Pu</td>
<td>1.01E+3</td>
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<td>2.95E+3</td>
<td>3.62E+4</td>
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<tr>
<td>$^{241}$Am$^+$</td>
<td>3.14E+0</td>
<td>3.11</td>
<td>9.77E+0</td>
<td>2.65E+4</td>
<td>0.01</td>
</tr>
</tbody>
</table>

$^+$ For thermal neutron, $^{237}$Np and $^{241}$Am have (n,$\gamma$) cross-sections of 175 barns and 684 barns, respectively. Daughter nuclei $^{238}$Np and $^{242}$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, $^{238}$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 $p_\nu$, $f_{\nu 1}$, $f_{\nu 2}$ consolidated/proposed by Zucker and Holden$^{12, 13, 14}$:

| \begin{tabular}{c|c|c|c|c} 
Spontaneous: $^{244}$Cm & Fast induced: $^{238}$U & Thermal induced: $^{235}$U \\ \hline $\nu_{s1}$ & $\nu_{i1}$ & $\nu_{i1}$ & $\nu_{i1}$ & 2.414 \\ $\nu_{s2}$ & 5.941 & $\nu_{i2}$ & 5.494 & 4.635 \\
\end{tabular} |

Also, $S_1$ and $r$ are given as $3.66E+4$ fissions/s and $9.96E+4$ /s for 1kgU 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 $F_1, F_2$, cross-sections (fast and thermal), density of isotopes in oxide and sample thickness is equal to $r = S_1 \nu_{s1}$. 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:

$$c = c_1 + c_2 = \frac{\overline{F}_1}{\sigma_1 n_1 x} + \frac{\overline{F}_2}{\sigma_2 n_2 x}$$

(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, $\sigma$ 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 $2.346\times10^{22}$ cm$^{-3}$ and 0.098$\times10^{22}$ 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, $\sigma_1$ and $\sigma_2$ 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:
Results of UO₂ containing 1, 2, 5 and 10 kgU are shown in Table 7-1~7-4, assuming $\epsilon_n$ 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  UO₂ containing 1 kgU (103.5 cm³, 8.9 mg $^{244}\text{Cm}$)

<table>
<thead>
<tr>
<th>$D_1/D_2$</th>
<th>–</th>
<th>200</th>
<th>100</th>
<th>50</th>
<th>20</th>
<th>10</th>
<th>5</th>
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<tbody>
<tr>
<td>$S_1$ [1/s]</td>
<td>36600 ($= 4.11E+6$ s⁻¹ g⁻¹ × 8.9 mg)</td>
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<tr>
<td>$r$ [1/s]</td>
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</tr>
<tr>
<td>$M_L$</td>
<td>–</td>
<td>1.1583</td>
<td>1.1647</td>
<td>1.1774</td>
<td>1.2156</td>
<td>1.2791</td>
<td>1.4056</td>
<td>1.7812</td>
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<tr>
<td>$F_1$ [1/s]</td>
<td>5852</td>
<td>5846</td>
<td>5834</td>
<td>5799</td>
<td>5740</td>
<td>5623</td>
<td>5275</td>
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<tr>
<td>$F_2$ [1/s]</td>
<td>269</td>
<td>538</td>
<td>1077</td>
<td>2689</td>
<td>5372</td>
<td>10716</td>
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<tr>
<td>$F_1 \nu_1$ [1/s]</td>
<td>15115</td>
<td>15100</td>
<td>15069</td>
<td>14978</td>
<td>14826</td>
<td>14524</td>
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<tr>
<td>$F_2 \nu_2$ [1/s]</td>
<td>650</td>
<td>1300</td>
<td>2599</td>
<td>6492</td>
<td>12967</td>
<td>25863</td>
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<tr>
<td>$F_1 \nu_1 + F_2 \nu_2$ [1/s]</td>
<td>15765</td>
<td>16400</td>
<td>17668</td>
<td>21470</td>
<td>27794</td>
<td>40391</td>
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<tr>
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<td>–</td>
<td>115354</td>
<td>115988</td>
<td>117257</td>
<td>121059</td>
<td>127382</td>
<td>139980</td>
<td>177386</td>
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<td>$1/(\alpha_1 \tau_1)$</td>
<td>–</td>
<td>0.0588</td>
<td>0.0587</td>
<td>0.0586</td>
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<td>0.0530</td>
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<tr>
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<td>0.0054</td>
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<td>0.0270</td>
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<td>0.1076</td>
<td>0.2669</td>
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<tr>
<td>$Y_1$</td>
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<td>0.581</td>
<td>0.584</td>
<td>0.590</td>
<td>0.609</td>
<td>0.640</td>
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<td>0.030</td>
<td>0.064</td>
<td>0.140</td>
<td>0.441</td>
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<td>$1/\alpha_1$ [s]</td>
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<td>1.38E-06</td>
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<td>1.38E-06</td>
<td>1.38E-06</td>
<td>1.38E-06</td>
<td>1.38E-06</td>
<td>1.38E-06</td>
</tr>
<tr>
<td>$1/\alpha_2$ [s]</td>
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<td>1.00E-05</td>
<td>1.00E-05</td>
<td>1.00E-05</td>
<td>1.00E-05</td>
<td>1.00E-05</td>
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<tr>
<td>$\alpha_1/\alpha_2$</td>
<td>–</td>
<td>7.255</td>
<td>7.261</td>
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<td>7.312</td>
<td>7.376</td>
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### Table 7-2  UO₂ containing 2 kgU (207.0 cm³, 17.8 mg $^{244}\text{Cm}$)

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<th>–</th>
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<td>$S_1$ [1/s]</td>
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<tr>
<td>$r$ [1/s]</td>
<td>199177</td>
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<tr>
<td>$M_L$</td>
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<td>1.2046</td>
<td>1.2178</td>
<td>1.2572</td>
<td>1.3228</td>
<td>1.4536</td>
<td>1.8418</td>
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<tr>
<td>$F_1$ [1/s]</td>
<td>14748</td>
<td>14736</td>
<td>14712</td>
<td>14639</td>
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<td>557</td>
<td>1113</td>
<td>2225</td>
<td>5559</td>
<td>11103</td>
<td>22149</td>
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<tr>
<td>$F_1 \nu_1$ [1/s]</td>
<td>38095</td>
<td>38063</td>
<td>38000</td>
<td>37811</td>
<td>37498</td>
<td>36873</td>
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<td>$F_2 \nu_2$ [1/s]</td>
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<td>2687</td>
<td>5372</td>
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<td>26803</td>
<td>53468</td>
<td>132643</td>
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<tr>
<td>$F_1 \nu_1 + F_2 \nu_2$ [1/s]</td>
<td>39438</td>
<td>40750</td>
<td>43372</td>
<td>51231</td>
<td>64301</td>
<td>90341</td>
<td>167659</td>
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<tr>
<td>$\lambda_1 N_1$ [1/s]</td>
<td>–</td>
<td>238615</td>
<td>239927</td>
<td>242549</td>
<td>250408</td>
<td>263478</td>
<td>289518</td>
<td>366837</td>
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<tr>
<td>$1/(\alpha_1 \tau_1)$</td>
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<td>0.0739</td>
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<tr>
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<td>0.2669</td>
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<tr>
<td>$Y_1$</td>
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<td>0.621</td>
<td>0.624</td>
<td>0.631</td>
<td>0.651</td>
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<tr>
<td>$Y_2$</td>
<td>–</td>
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<td>0.006</td>
<td>0.013</td>
<td>0.030</td>
<td>0.064</td>
<td>0.140</td>
<td>0.441</td>
</tr>
<tr>
<td>$1/\alpha_1$ [s]</td>
<td>–</td>
<td>8.42E-07</td>
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<td>$1/\alpha_2$ [s]</td>
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<td>5.02E-06</td>
<td>5.02E-06</td>
<td>5.02E-06</td>
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<tr>
<td>$\alpha_1/\alpha_2$</td>
<td>–</td>
<td>5.963</td>
<td>5.967</td>
<td>5.976</td>
<td>6.000</td>
<td>6.042</td>
<td>6.128</td>
<td>6.400</td>
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Table 7-3  UO₂ containing 5 kgU (517.5 cm³, 44.5 mg ²⁴⁴Cm)

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<tbody>
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<td>D₁/D₂</td>
<td>200</td>
<td>100</td>
<td>50</td>
<td>20</td>
<td>10</td>
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<td>1.2876</td>
<td>1.3293</td>
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<td>50018</td>
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<td>F₁ν₁+F₂ν₁</td>
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<td>634233</td>
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<td>661906</td>
<td>696417</td>
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<td>0.1004</td>
<td>0.1003</td>
<td>0.0999</td>
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<td>0.697</td>
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<td>4.678</td>
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</table>

The time constant 1/α₂ is several to ten times longer than 1/α₁, which is consistent to Rossi-alpha combined distribution though affected by the moderator of detector.

Table 7-4  UO₂ containing 10 kgU (1035.1 cm³, 89mg ²⁴⁴Cm)

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<td>1/(α₁τ₁)</td>
<td>0.1267</td>
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<td>1/(α₂τ₂)</td>
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<td>0.008</td>
<td>0.016</td>
<td>0.040</td>
<td>0.085</td>
<td>0.185</td>
<td>0.583</td>
</tr>
<tr>
<td>1/α₁</td>
<td>2.57E-07</td>
<td>2.57E-07</td>
<td>2.57E-07</td>
<td>2.56E-07</td>
<td>2.55E-07</td>
<td>2.53E-07</td>
<td>2.47E-07</td>
</tr>
<tr>
<td>1/α₂</td>
<td>1.00E-06</td>
<td>1.00E-06</td>
<td>1.00E-06</td>
<td>1.00E-06</td>
<td>1.00E-06</td>
<td>1.00E-06</td>
<td>1.00E-06</td>
</tr>
</tbody>
</table>
Regarding the time constant $1/\alpha_2$, the time constant and the ratio $\alpha_1/\alpha_2$ 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 | 13949 |
| $Y$ | 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 $F_1$ and $F_2$ 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 $M_L$, $F_1$ and $F_2$ 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 $F_2$ 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, $F_1$ 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 viewpoint of materials accountancy and safeguards, $^{235}$U, $^{239}$Pu and $^{241}$Pu are especially focused on, however $F_2$ 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 $(n,\gamma)$ reaction by $^{238}$U, $(n,\alpha)$ reaction by $^{10}$B and so on are also coupled linearly but negatively to the contributions, i.e.:

$$F_2 \propto \left[ \sigma_{\text{thm},(n,f)} W \right]^{235}\text{U} + \left[ \sigma_{\text{thm},(n,f)} W \right]^{239}\text{Pu} + \left[ \sigma_{\text{thm},(n,f)} W \right]^{241}\text{Pu} - \left[ \sigma_{(n,\gamma)} W \right]^{238}\text{U} - \left[ \sigma_{(n,\alpha)} W \right]^{10}\text{B}$$ \hspace{1cm} (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, $F_2$ 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 $103.5 \text{ cm}^2 \times 1 \text{ cm}$ for $\text{UO}_2$ containing 1 kg U. As previously mentioned, the time constant $1/\alpha_2$ and the ratio $\alpha_1/\alpha_2$ 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 \((\alpha, n)\) reactions correspond to \(^{244}\text{Cm}\) spontaneous fission is estimated\(^{26}\) to be 706 \(s^{-1}\) for 1 kgU (8.9 mg \(^{244}\text{Cm}\)), which is <1% of \(r = S_1 \nu s_1\).

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% \(^{244}\text{Cm}\). It should be noted that the fission rate by thermal neutron \(F_2\) 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

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]

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:

\[
\begin{align*}
\{ \text{Expected number of correlated neutrons in interval } t \} &= \int_{t_2=0}^{t_2=t} \int_{t_1=0}^{t_1=t_2} \left\{ F\epsilon \times \left[ \frac{\nu(\nu - 1)}{2\alpha\tau^2} \exp^{-\alpha(t_2-t_1)} \right] \right\} dt_1 dt_2 \\
&= F^2\epsilon^2 \int_{t_2=0}^{t_2=t} \left[ t_1 \right]_{t_0}^{t_2} dt_2 + F\epsilon \frac{\nu(\nu - 1)}{2\alpha\tau^2} \int_{t_2=0}^{t_2=t} \exp^{-\alpha t_2} \left[ \frac{1}{\alpha} \exp^{\alpha t_1} \right]_{0}^{t_2} dt_2 \\
&= \frac{F^2\epsilon^2 t^2}{2} + F\epsilon \frac{\nu(\nu - 1)}{2\alpha^2\tau^2} \left[ t - \frac{1}{\alpha} (1 - \exp^{-\alpha t}) \right] \\
&= \frac{F^2\epsilon^2 t^2}{2} + F\epsilon \frac{\nu(\nu - 1)}{2\alpha^2\tau^2} t \left( 1 - \frac{1 - \exp^{-\alpha t}}{\alpha t} \right)
\end{align*}
\]
On the other hand, accidentals (≠ pair) is formulated as:

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

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

\[
= F^2 e^2 t - F^2 e^2 \frac{1}{\lambda} \left( 1 - \exp^{-\lambda t} \right)
\]

\[
= F^2 e^2 t \left( 1 - \frac{1 - \exp^{-\lambda t}}{\lambda t} \right)
\]

**Neutrons come from a Poisson Process**

\[
\text{PDF: } \frac{1}{\lambda} e^{-\lambda (t_2-t_1)} \times F_\epsilon [\text{counts/s}]
\]

where \( F \) is the number of fissions per unit time, \( \epsilon \) is efficiency of counter (counts per fission), \( \alpha \) is rate of decay regarding prompt neutrons, \( \tau \) is mean time between fissions and \( \lambda \) 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:

\[
N_c(t) = \frac{F^2 e^2 t^2}{2} + F^2 e^2 \frac{\nu (\nu - 1)}{2 \alpha^2 \tau^2} t \left( 1 - \frac{1 - \exp^{-\alpha t}}{\alpha t} \right) \quad (a1)
\]

\[
N_u(t) = F^2 e^2 t \left( 1 - \frac{1 - \exp^{-\lambda t}}{\lambda t} \right) \quad (a2)
\]

![Figure a2 Curve of the asymptotic term of \( N_c(t) \) and \( N_u(t) \) close to \( t=0 \).](image)

- 40 -
The first derivatives of coupled pair of correlated neutrons $N_{c}^{\text{coupled}}(t)$ and uncorrelated neutrons $N_{u}(t)$ are constants:

$$
\frac{dN_{c}^{\text{coupled}}(t)}{dt} = F \epsilon \frac{\nu(\nu-1)}{2 \alpha^2 \tau^2} = F \times \frac{\nu(\nu-1)}{2} \times \left(\frac{\epsilon}{\alpha \tau}\right)^2
$$

(a3)

$$
= F p m_{\text{eff}} \times \frac{\epsilon_n}{\alpha \tau} \left(\epsilon_n \frac{\nu}{p_l \nu}\right)^2 \quad \therefore \epsilon = \epsilon_n \frac{\nu}{p_l \nu}
$$

$$
= F p m_{\text{eff}} \times \frac{\epsilon_n}{\alpha \tau} \left(\epsilon_n \frac{\nu}{p_l \nu}\right)^2 \quad \therefore M_L = p_l M_T = p_l \left(\frac{\nu}{\alpha \tau}\right)
$$

$$
\frac{dN_{u}(t)}{dt} = F^2 \epsilon^2 = S^2 \text{ or } T^2
$$

(a4)

where $F_p$ is the number of spontaneous fissions per unit mass and time, $m_{\text{eff}}$ is effective mass of spontaneous fission nuclei, $\epsilon_n$ 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 $N_{c}^{\text{accidental}}(t)$.

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

(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.
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 $|z| \leq 1$:

$$G_X (z) \equiv \frac{1}{z^X} = \sum_{n=0}^{\infty} z^n P_X (X=n) \quad (b1)$$

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:

$$G_X (z) = \sum_{n=0}^{\infty} z^n P_X (X=n) = P_X (X=0) + \sum_{n=1}^{\infty} z^n P_X (X=n)$$

$$G'_X (z) = \sum_{n=1}^{\infty} n z^{n-1} P_X (X=n) = 1 P_X (X=1) + \sum_{n=2}^{\infty} n z^{n-1} P_X (X=n) \quad (b2)$$

$$G''_X (z) = \sum_{n=2}^{\infty} n(n-1) z^{n-2} P_X (X=n) = 2 P_X (X=2) + \sum_{n=3}^{\infty} n(n-1) z^{n-2} P_X (X=n)$$

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

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

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

$$G^{(k)}_X |_{z=1} = \sum_{n=k}^{\infty} n(n-1) \cdots (n-k+1) P_X (X=n) = X(X-1) \cdots (X-k+1) \quad (b4)$$

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:

$$G_X (z) = \sum_{k=0}^{\infty} \frac{M^{(k)}_P}{k!} (z-1)^k \quad \text{where} \quad M^{(k)}_P \equiv G^{(k)}_X |_{z=1} \quad (b5)$$

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:

$$G_{X_1+X_2+\cdots+X_N} (z) = G_{X_1} (z) \cdot G_{X_2} (z) \cdots G_{X_N} (z) \quad (b6)$$

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

$$G_S (z) = [G_X (z)]^N = [G_X (z)]^N = G_N (G_X (z)) \quad (b7)$$

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

\[
z[u_] := (1 - p) u + p y[h[u]]
\]

\[
\text{dh1} = \text{Flatten}\left[\text{Solve}\left[h^{(1)}[u] == z^{(1)}[u], h^{(1)}[u]\right]\right];
\]

\[
\text{dh2} = \text{Simplify}\left[\text{Flatten}\left[\text{Solve}\left[h^{(2)}[u] == z^{(2)}[u], h^{(2)}[u]\right] \text{/. dh1}\right]\right];
\]

\[
\text{dh3} = \text{Simplify}\left[\text{Flatten}\left[\text{Solve}\left[h^{(3)}[u] == z^{(3)}[u], h^{(3)}[u]\right] \text{/. dh1} \text{/. dh2}\right]\right];
\]

\[
\text{dh4} = \text{Simplify}\left[\text{Flatten}\left[\text{Solve}\left[h^{(4)}[u] == z^{(4)}[u], h^{(4)}[u]\right] \text{/. dh1} \text{/. dh2} \text{/. dh3}\right]\right];
\]

\[
\text{dh5} = \text{Simplify}\left[\text{Flatten}\left[\text{Solve}\left[h^{(5)}[u] == z^{(5)}[u], h^{(5)}[u]\right] \text{/. dh1} \text{/. dh2} \text{/. dh3} \text{/. dh4}\right]\right];
\]

\[
\text{dh6} = \text{Simplify}\left[\text{Flatten}\left[\text{Solve}\left[h^{(6)}[u] == z^{(6)}[u], h^{(6)}[u]\right] \text{/. dh1} \text{/. dh2} \text{/. dh3} \text{/. dh4} \text{/. dh5}\right]\right];
\]

\[
\text{dh7} = \text{Simplify}\left[\text{Flatten}\left[\text{Solve}\left[h^{(7)}[u] == z^{(7)}[u], h^{(7)}[u]\right] \text{/. dh1} \text{/. dh2} \text{/. dh3} \text{/. dh4} \text{/. dh5} \text{/. dh6}\right]\right];
\]

\[
\text{dhH} = \text{Simplify}\left[\text{H}^{(1)}[u] \text{/. dh1}\right];
\]

\[
\text{dh2} = \text{Simplify}\left[\text{H}^{(2)}[u] \text{/. dh1} \text{/. dh2}\right];
\]

\[
\text{dh3} = \text{Simplify}\left[\text{H}^{(3)}[u] \text{/. dh1} \text{/. dh2} \text{/. dh3}\right];
\]

\[
\text{dh4} = \text{Simplify}\left[\text{H}^{(4)}[u] \text{/. dh1} \text{/. dh2} \text{/. dh3} \text{/. dh4}\right];
\]

\[
\text{dh5} = \text{Simplify}\left[\text{H}^{(5)}[u] \text{/. dh1} \text{/. dh2} \text{/. dh3} \text{/. dh4} \text{/. dh5}\right];
\]

\[
\text{dh6} = \text{Simplify}\left[\text{H}^{(6)}[u] \text{/. dh1} \text{/. dh2} \text{/. dh3} \text{/. dh4} \text{/. dh5} \text{/. dh6}\right];
\]

\[
\text{dh7} = \text{Simplify}\left[\text{H}^{(7)}[u] \text{/. dh1} \text{/. dh2} \text{/. dh3} \text{/. dh4} \text{/. dh5} \text{/. dh6} \text{/. dh7}\right];
\]

d) Another expression of probability mass distribution

Another expression of probability mass distribution of multiple leakage neutrons is possible. Instead of \(\eta_{ns-m}^{\nu}\) (factorial moment), \(\eta_{ns-m}^{\nu}\) that means directly the number of multiple neutron pairs is defined as:

\[
\eta_{ns-m}^{\nu} = \frac{\max_{\nu_{ns-m}^{\nu}} \left( \nu_{ns-m}^{\nu} \right) P_{\nu}}{m!}
\]

\[
\text{and } M_{L\eta} = M_{L\nu} \quad \text{(d1)}
\]

Probability mass distribution of multiple leakage neutrons using \(\eta_{ns-m}^{\nu}\) is given as:

\[
P_{m} (m = 1) = M_{L} (1 + \alpha_{r}) \eta_{s1}
\]

\[
P_{m} (m = 2) = M_{L}^{2} \left[ \eta_{s2} + \eta_{s2} M_{L\eta} (1 + \alpha_{r}) \eta_{s1} \right] \quad \text{(d2)}
\]

\[
P_{m} (m = 3) = M_{L}^{3} \left[ \eta_{s3} + 2 \eta_{s2} M_{L\eta} \eta_{s2} + \left( \eta_{s3} M_{L\eta} + 2 \eta_{s2}^{2} M_{L\eta} \right) (1 + \alpha_{r}) \eta_{s1} \right] \quad \text{(d3)}
\]

\[
P_{m} (m = 4) = M_{L}^{4} \left[ \eta_{s4} + 3 \eta_{s2} M_{L\eta} \eta_{s3} 
\right.
\]

\[
\left. + \left( 2 \eta_{s3} M_{L\eta} + 5 \eta_{s2}^{2} M_{L\eta}^{2} \right) \eta_{s2} 
\right.
\]

\[
\left. + \left( \eta_{s4} M_{L\eta} + 5 \eta_{s3} \eta_{s2} M_{L\eta}^{2} + 5 \eta_{s2}^{2} M_{L\eta}^{3} \right) (1 + \alpha_{r}) \eta_{s1} \right] \quad \text{(d4)}
\]
\[ P_m (m=5) = M_5^L \{ \eta_{s5} + 4 \eta_{i2} M_{L\eta} \eta_{s4} + (3 \eta_{i3} M_{L\eta} + 9 \eta_{i2}^2 M_{L\eta}^2) \eta_{s3} + (2 \eta_{i4} M_{L\eta} + 12 \eta_{i3} \eta_{i2} M_{L\eta}^2 + 14 \eta_{i2}^3 M_{L\eta}^3) \eta_{s2} + \left[ \eta_{i5} M_{L\eta} + (6 \eta_{i4} \eta_{i2} + 3 \eta_{i3}^2) M_{L\eta}^2 + 21 \eta_{i4} \eta_{i2}^2 M_{L\eta}^3 + 14 \eta_{i2}^4 M_{L\eta}^4 \right] (1 + \alpha_r) \eta_{s1} \} \] (d6)

\[ P_m (m=6) = M_6^L \{ \eta_{s6} + 5 \eta_{i2} M_{L\eta} \eta_{s5} + (4 \eta_{i3} M_{L\eta} + 14 \eta_{i2}^2 M_{L\eta}^2) \eta_{s4} + (3 \eta_{i4} M_{L\eta} + 21 \eta_{i3} \eta_{i2} M_{L\eta}^2 + 28 \eta_{i2}^3 M_{L\eta}^3) \eta_{s3} + \left[ 2 \eta_{i5} M_{L\eta} + (14 \eta_{i4} \eta_{i2} + 7 \eta_{i3}^2) M_{L\eta}^2 + 56 \eta_{i3} \eta_{i2}^2 M_{L\eta}^3 + 42 \eta_{i2}^4 M_{L\eta}^4 \right] \eta_{s2} + \left[ \eta_{i6} M_{L\eta} + (7 \eta_{i5} \eta_{i2} + 7 \eta_{i4} \eta_{i3}) M_{L\eta}^2 + (28 \eta_{i4} \eta_{i2}^2 + 28 \eta_{i3}^2 \eta_{i2}) M_{L\eta}^3 + 84 \eta_{i3} \eta_{i2}^3 M_{L\eta}^4 + 42 \eta_{i2}^4 M_{L\eta}^5 \right] (1 + \alpha_r) \eta_{s1} \} \] (d7)

\[ P_m (m=7) = M_7^L \{ \eta_{s7} + 6 \eta_{i2} M_{L\eta} \eta_{s6} + (5 \eta_{i3} M_{L\eta} + 20 \eta_{i2}^2 M_{L\eta}^2) \eta_{s5} + (4 \eta_{i4} M_{L\eta} + 32 \eta_{i3} \eta_{i2} M_{L\eta}^2 + 48 \eta_{i2}^3 M_{L\eta}^3) \eta_{s4} + \left[ 3 \eta_{i5} M_{L\eta} + (24 \eta_{i4} \eta_{i2} + 12 \eta_{i3}^2) M_{L\eta}^2 + 108 \eta_{i3} \eta_{i2}^2 M_{L\eta}^3 + 90 \eta_{i2}^4 M_{L\eta}^4 \right] \eta_{s3} + \left[ 2 \eta_{i6} M_{L\eta} + (16 \eta_{i5} \eta_{i2} + 16 \eta_{i4} \eta_{i3}) M_{L\eta}^2 + (72 \eta_{i4} \eta_{i2}^2 + 72 \eta_{i3}^2 \eta_{i2}) M_{L\eta}^3 + 240 \eta_{i3} \eta_{i2}^3 M_{L\eta}^4 + 132 \eta_{i2}^5 M_{L\eta}^5 \right] \eta_{s2} + \left[ \eta_{i7} M_{L\eta} + (8 \eta_{i6} \eta_{i2} + 8 \eta_{i5} \eta_{i3} + 4 \eta_{i4}^2) M_{L\eta}^2 + (36 \eta_{i5} \eta_{i2}^2 + 72 \eta_{i4} \eta_{i3} \eta_{i2} + 12 \eta_{i3}^3) M_{L\eta}^3 + (120 \eta_{i4} \eta_{i2}^3 + 180 \eta_{i3}^2 \eta_{i2}^2) M_{L\eta}^4 + 330 \eta_{i3} \eta_{i2}^4 M_{L\eta}^5 + 132 \eta_{i2}^6 M_{L\eta}^6 \right] (1 + \alpha_r) \eta_{s1} \} \] (d8)

Compared to the expression using \( \overline{\nu_{s5}}^m \), 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 $|z_2| \leq 1$:

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

From equation (a4):

$$X = \partial G_{XY}(z_1, z_2) \bigg|_{z_1=1} = \partial \frac{\partial G_{XY}(z_1, z_2)}{\partial z_1} \bigg|_{z_1=1} \quad (e2)$$

$$Y = \partial G_{XY}(z_1, z_2) \bigg|_{z_2=1} = \partial \frac{\partial G_{XY}(z_1, z_2)}{\partial z_2} \bigg|_{z_2=1} \quad (e3)$$

and:

$$X(X-1) = \partial^2 G_{XY}(z_1, z_2) \bigg|_{z_1=1} = \partial^2 \frac{\partial G_{XY}(z_1, z_2)}{\partial z_1^2} \bigg|_{z_1=1} \quad (e4)$$

$$XY = \partial^2 G_{XY}(z_1, z_2) \bigg|_{z_1=z_2=1} = \partial^2 \frac{\partial G_{XY}(z_1, z_2)}{\partial z_1 \partial z_2} \bigg|_{z_1=z_2=1} \quad (e5)$$

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

$$\text{Cov}(X, X) = \overline{X(X-1)} - (\overline{X})^2 = \overline{X^2} - \overline{X} - (\overline{X})^2 = (\overline{X} - \overline{X})^2 - \overline{X} \quad (e6)$$

Thus:

$$\text{Var}(X) = \text{Cov}(X, X) + \mu(X) \quad (e7)$$

$$\text{Var}(X) = M_{P_X}'' + M_{P_X}' - \left( M_{P_X}' \right)^2 \quad (e8)$$

Normal covariance of $X$ and $Y$ is defined as:

$$\text{Cov}(X, Y) = \overline{XY} - \overline{X} \overline{Y} = \frac{(X - \overline{X})(Y - \overline{Y})}{\text{covariance}} \quad (e9)$$

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

$$\frac{\partial^2 G}{\partial X^2} \bigg|_{X=1} = N_1(N_1-1) \text{ is not self-covariance of } N_1 \text{ and}$$

$$\frac{\partial^2 G}{\partial X \partial Y} \bigg|_{X=Y=1} = N_1N_2 \text{ is not covariance of } N_1 \text{ and } N_2.$$
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:

$$M_L = l M_T = l \left[ 1 + p \nu \nu_1 + (p \nu \nu_1)^2 \ldots \right] = \frac{l}{1-p \nu \nu_1} = \frac{1-p-p_c}{1-p \nu \nu_1} \approx \frac{1-p}{1-p \nu \nu_1} \quad (f1)$$

Another expression derived in the body is:

$$M_L = \frac{1}{S_1 \nu s_1} \left( S_1 \nu s_1 + F_1 \nu^1_1 + F_2 \nu^2_1 \right) \quad (f2)$$

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

$$\frac{1}{n} \frac{dn}{dt} = k_{\text{eff}} \left( 1 - \beta_{\text{eff}} \right) - \frac{1}{l_s} \approx \frac{k_{\text{eff}} - 1}{l_s} \quad \text{if delayed neutron is sufficiently small} \quad (f3)$$

$$\rho = \frac{k_{\text{eff}} - 1}{k_{\text{eff}}} \quad (f4)$$

$$\Lambda = \frac{l_s}{k_{\text{eff}}} \quad (f5)$$

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

where $n$ is the number of neutrons in arbitrary generation, $\beta_{\text{eff}}$ is the ratio of the number of delayed neutrons to the one of total neutrons, $k_{\text{eff}}$ is effective multiplication factor, $\rho$ is reactivity and $\Lambda$ is prompt neutron generation time. These equations satisfy the relation below that is the definition of $\alpha$:

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

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

$$\frac{1}{\alpha \tau} = \frac{1}{1-p \nu} = \frac{1}{1-k_{\text{eff}}} = M_T \quad (f8)$$

Therefore:

$$M_L = l M_T = \frac{l \nu}{\alpha \tau} = \frac{l}{\alpha l_s} \therefore \frac{\tau}{l_s} = \nu \quad (f9)$$

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 $10^6$. 

- 46 -
Regarding equation (f9), physical meaning of $\frac{\nu}{(\alpha \tau)} = M_F$ is clear because $\frac{1}{(\alpha \tau)}$ is the number of induced fissions by one source neutron and $\frac{\nu}{(\alpha \tau)}$ is the number of neutrons induced by one source neutron. Physical meaning of $\tau = \overline{\nu}l_s$ is also clear because $\tau$ is mean time between fissions and $l_s$ is neutron lifespan.

Secondary additional different expression is possible using $\epsilon_n$, $\epsilon_1$ and $\epsilon_2$:

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

This equation satisfies 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):

$$M_L^1 = \frac{1}{S_1 \overline{\nu_{s_1}}} \left( S_1 \overline{\nu_{s_1}} + F_1 \overline{\nu_{i_1}} \right) = 1 + \frac{\overline{\nu_{i_1}}}{\alpha_1 \tau_1} \quad (f11)$$

$$M_L^2 = \frac{1}{S_1 \overline{\nu_{s_1}}} F_2 \overline{\nu_{i_1}}^2 = \frac{\overline{\nu_{i_1}}^2}{\alpha_2 \tau_2} \quad (f12)$$

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 $\alpha$, $\gamma$, proton, fission, total, elastic, nonelastic ($\equiv$ TOT–EL), inelastic ($\equiv$ NON–absorptions), respectively;
- $^{16}$O is eliminated due to small (n, $\gamma$) cross section (0.00019 barns for thermal neutron);
- $^{237}$Np(n, $\gamma$) $\rightarrow$ $^{238}$Np, $^{241}$Am(n, $\gamma$) = $^{242m}$Am or $^{243}$Am (via $^{242}$Am) are added for safeguards.
$^{241}\text{Pu}$

Cross section [barns] vs. Energy [MeV]

- 1012 barns for thermal by (N,F) reaction
- PU-241(N,NON) is not included in the library.

$^{242}\text{Pu}$

Cross section [barns] vs. Energy [MeV]

- 21.27 barns for thermal by (N,G) reaction
- PU-242(N,NON) is not included in the library.

$^{241}\text{Am}$

Cross section [barns] vs. Energy [MeV]

- 684.3 barns for thermal by (N,G) reaction
- AM-241(N,NON) is not included in the library.
This is a blank page.
### 国際単位系（SI）

#### 表1. SI基本単位

<table>
<thead>
<tr>
<th>基本量</th>
<th>名称</th>
<th>記号</th>
<th>定義</th>
</tr>
</thead>
<tbody>
<tr>
<td>長さ</td>
<td>メートル</td>
<td>m</td>
<td>1m = 1/25,196,850 広域の釣金の長さ</td>
</tr>
<tr>
<td>質量</td>
<td>キログラム</td>
<td>kg</td>
<td>0℃,101.3kPaの水の密度が1g/mLとなる量</td>
</tr>
<tr>
<td>時間</td>
<td>秒</td>
<td>s</td>
<td>1s = 9,192,631,770周期の光の餌</td>
</tr>
<tr>
<td>電気</td>
<td>アンペア</td>
<td>A</td>
<td>1A = 1C/s</td>
</tr>
<tr>
<td>熱力学温湿度</td>
<td>ケルビン</td>
<td>K</td>
<td>0K = -273.15℃</td>
</tr>
<tr>
<td>物質的</td>
<td>モル</td>
<td>mol</td>
<td>1mol = 1/6.0221429×10^23個の物質</td>
</tr>
<tr>
<td>光度</td>
<td>ユニット</td>
<td>cd</td>
<td>1cd = 1lm/sr</td>
</tr>
</tbody>
</table>

#### 表2. 基本単位を用いて表されるSI組立単位の例

<table>
<thead>
<tr>
<th>組立単位</th>
<th>記号</th>
<th>定義</th>
</tr>
</thead>
<tbody>
<tr>
<td>電流密度</td>
<td>A/m^2</td>
<td>1A/m^2 = 1C/(s·m^2)</td>
</tr>
<tr>
<td>熱流密度</td>
<td>W/m^2</td>
<td>1W/m^2 = 1J/(s·m^2)</td>
</tr>
<tr>
<td>面電圧</td>
<td>V/m</td>
<td>1V/m = 1J/(C·m)</td>
</tr>
<tr>
<td>加速度</td>
<td>m/s^2</td>
<td>1m/s^2 = 1m/(s^2)</td>
</tr>
</tbody>
</table>

#### 表3. 固有の名称と記号で表されるSI組立単位

<table>
<thead>
<tr>
<th>組立単位</th>
<th>名称</th>
<th>記号</th>
<th>定義</th>
</tr>
</thead>
<tbody>
<tr>
<td>エンタルピー</td>
<td>ジュールモル当量</td>
<td>J/mol</td>
<td>1J/mol = 1Kmol当量のエンタルピー</td>
</tr>
<tr>
<td>エネルギー</td>
<td>ジュール</td>
<td>J</td>
<td>1J = 1Nm</td>
</tr>
<tr>
<td>動力学</td>
<td>ジュールモル当量</td>
<td>J/mol</td>
<td>1J/mol = 1Kmol当量のエンタルピー</td>
</tr>
</tbody>
</table>

#### 表4. 国際単位系の定義

<table>
<thead>
<tr>
<th>名称</th>
<th>定義</th>
</tr>
</thead>
<tbody>
<tr>
<td>ペバラスカール</td>
<td>1/60s</td>
</tr>
<tr>
<td>モメント</td>
<td>Nm</td>
</tr>
</tbody>
</table>

#### 表5. SIの接頭語

<table>
<thead>
<tr>
<th>接頭語</th>
<th>記号</th>
<th>定義</th>
</tr>
</thead>
<tbody>
<tr>
<td>本位</td>
<td>·</td>
<td>1</td>
</tr>
<tr>
<td>十</td>
<td>d</td>
<td>10</td>
</tr>
<tr>
<td>オーグ</td>
<td>d</td>
<td>10^3</td>
</tr>
<tr>
<td>万</td>
<td>d</td>
<td>10^6</td>
</tr>
</tbody>
</table>

#### 表6. 国際単位系の定義

<table>
<thead>
<tr>
<th>名称</th>
<th>記号</th>
</tr>
</thead>
<tbody>
<tr>
<td>安培</td>
<td>A</td>
</tr>
<tr>
<td>ジュール</td>
<td>J</td>
</tr>
<tr>
<td>ジャルペルト</td>
<td>J/m^2</td>
</tr>
</tbody>
</table>

#### 表7. シリウスのSI単位で表される数値

<table>
<thead>
<tr>
<th>名称</th>
<th>数値</th>
</tr>
</thead>
<tbody>
<tr>
<td>真空電気透過程</td>
<td>1.66027016×10^-19</td>
</tr>
<tr>
<td>真空波長</td>
<td>1.66027016×10^-19</td>
</tr>
</tbody>
</table>

#### 表8. シリウスのSI単位で表される数値

<table>
<thead>
<tr>
<th>名称</th>
<th>数値</th>
</tr>
</thead>
<tbody>
<tr>
<td>キュリ</td>
<td>1.66027016×10^-19</td>
</tr>
<tr>
<td>デーサル</td>
<td>1.66027016×10^-19</td>
</tr>
</tbody>
</table>

#### 表9. シリウスのSI単位で表される数値

<table>
<thead>
<tr>
<th>名称</th>
<th>数値</th>
</tr>
</thead>
<tbody>
<tr>
<td>カリウム</td>
<td>1.66027016×10^-19</td>
</tr>
<tr>
<td>チタニウム</td>
<td>1.66027016×10^-19</td>
</tr>
</tbody>
</table>

#### 表10. SI単位で表される数値

<table>
<thead>
<tr>
<th>名称</th>
<th>記号</th>
<th>定義</th>
</tr>
</thead>
<tbody>
<tr>
<td>キュリ</td>
<td>Ci</td>
<td>1.66027016×10^-19</td>
</tr>
<tr>
<td>デーサル</td>
<td>da</td>
<td>1.66027016×10^-19</td>
</tr>
</tbody>
</table>

#### 表11. SI単位で表される数値

<table>
<thead>
<tr>
<th>名称</th>
<th>記号</th>
<th>定義</th>
</tr>
</thead>
<tbody>
<tr>
<td>カリウム</td>
<td>K</td>
<td>1.66027016×10^-19</td>
</tr>
<tr>
<td>チタニウム</td>
<td>Ti</td>
<td>1.66027016×10^-19</td>
</tr>
</tbody>
</table>