Analysis of measurement model of uranium multiplicity based on the liquid scintillators

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Abstract. In this paper, we point out the detection differences between the fluid scintillation detection system and the He3 detection system, and then build a special measurement system with four detectors. Based on the proposed measurement system, we analyze the fission neutrons distribution as well as the effects of self-multiplication, detector efficiency, detector record, neutron crosstalk on neutron number distribution and then establish the equations which describe the relationship between measuring parameters and samples parameters (equations). Finally using the model this paper proposed, we analyse the measuring parameters, especially the influence and the function of crosstalk rate.

1. Introduction
Comparing with the usual coincidence counting methods [1-3], the advantage of multiplicity measurement is which can measure and analyze the three count rate, besides the one and two count rate, so it can provide more information about the sample.

It is worth mentioning that there is a strong (a, n) reaction in the oxide samples of plutonium, that is, the a disintegration of plutonium produces the particles and the oxygen nucleus reaction in the oxide produces the neutrons, which are produced randomly, and usually as the background of a heavy count; different from the measurement of plutonium samples, in the measurement of uranium materials in this work, due to the excitation of fission through an external neutron inquiry source, the a disintegration of its own is negligible, so the (a, n) reaction is not considered.

The another differentia is spontaneous fission, after analyzing its own fission rate F from the measured data, the equivalent mass of 240Pu can be obtained by dividing the fission times per second of its unit mass by F; the induced fission of uranium sample, after analyzing the induced fission F from the measured data, it is necessary to know the function relation of F~m5, then the quality of 235U is obtained.

2. Measurement basis
As shown in figure 1, the measuring prototype consists of four BC501A liquid scintillation detectors [4]. An AmLi acquisition source (neutron emission intensity is about 10^7/s) is placed in the center of the upper and the lower surfaces. And U nucleus in the measured samples will be excited by AmLi to have induced fission (mainly for U5 fission, the contribution of U8 is relatively small). Then we can know the effective mass of U5 in the measured samples by fission neutron measurement and analysis.
Figure 1. Detector diagram.

When measuring, we use two corresponding AmLi in the upper and lower surfaces to make the multi-channel signals shift during their way from the detector to the discriminator switch, and use the timer counter for the measurement. The signals from the four-way liquid scintillation detector pass through the neutron gamma discrimination circuit respectively. The four-circuit discriminator has the same delay, and the logic pulse is output when the neutron signal is screened. We apply the instrument with one-circuit beginning and four-circuit timing function to measure the signal with neutron time information in the four-circuit discriminator. When there is a neutron signal output in any of the four circuits, the timer starts to count and record the incoming signals in 100ns. After a period of measurement, the recorded information is processed, and the total number of each detector is obtained. After that, singles, doubles, triples and quadruples are all obtained by solving relatively statistical computing.

Measurement instrumentation includes n/γ discriminator, MPD-4 (at most go into four input signals at the same time), the time of high resolution signal counter McS6a (at most go into the six input signals at the same time), and a connection McS6a used for computer data acquisition analysis. In addition, there are portable multichannel spectrometer, MCA2000, and an mpd-4 computer for debugging. The threshold value of the measurement is 1/4Cs, which corresponds to the optical pulse of 120keV Compton electron energy. The proton light response function of the BC501A detector shows that the corresponding neutron energy is 0.76MeV.

3. Mathematical model and data analysis algorithm
The data analysis method based on the He³ measurement system and the corresponding shift register electronics cannot be applied to the liquid scintillation detection system due to their significant differences. In this research, we attempt to describe the main physical process of neutron measurement by mathematical model, propose data analysis algorithm and then connect the data of the experiment with the undetermined parameters of the sample to construct the model [5-7]. Afterwards, the unknown parameters of the sample can be obtained by solving equations (induced fission rate F, self-multiplication coefficient M, etc.).

3.1. Initial distribution of the fission neutron number - row vector E₀
The probability of a fission emission neutron is recorded in the sample, and the fission neutron quantity distribution can be denoted as row vector, \( E = (E₁, E₂, E₃, ..., E_{vmax}) \). The neutron number distribution of \( U₅ \) fission produced by different energy neutron induced \( U₅ \) is different, and the distribution table of \( U₅ \) induced fission neutron number is shown in table 1.

AmLi source neutrons reacting with the sample after the polyethylene is moderated by the active method to measure uranium samples. The biggest energy of AmLi source neutron does not exceed 1.5
MeV, and its average energy is 0.57 MeV. However, after polyethylene slowing down, the average energy of the sample space is 0.28 MeV, the desirable thermal neutron of the row vector $E$ of $U_5$ fission neutron number excited by AmLi source is distributed as $E=[0.17, 0.34, 0.31, 0.13, 0.027, 0.0026, 0.00014]$.

Table 1. The induced fission neutron number distribution in $U_5$ (The unit of En in the Table should be MeV).

| $E_n$ | $v=0$ | 1 | 2 | 3 | 4 | 5 | 6 | 7 | average |
|-------|-------|---|---|---|---|---|---|---|---------|
| 0     | 0.0097 | 0.00801 | 0.0073 | 0.00678 | 0.00578 | 0.00463 | 0.00364 | 0.0027 | 0.0018 |
| 1     | 0.0127 | 0.01349 | 0.01472 | 0.01603 | 0.01731 | 0.01840 | 0.01966 | 0.02102 | 0.0223 |
| 2     | 0.0152 | 0.01631 | 0.01742 | 0.01843 | 0.01935 | 0.02026 | 0.02108 | 0.02190 | 0.0227 |
| 3     | 0.0172 | 0.01823 | 0.01915 | 0.02000 | 0.02078 | 0.02150 | 0.02213 | 0.02270 | 0.0233 |
| 4     | 0.0191 | 0.02003 | 0.02086 | 0.02161 | 0.02226 | 0.02282 | 0.02329 | 0.02370 | 0.0242 |
| 5     | 0.0204 | 0.02123 | 0.02195 | 0.02261 | 0.02315 | 0.02360 | 0.02397 | 0.02433 | 0.0247 |
| 6     | 0.0211 | 0.02184 | 0.02247 | 0.02302 | 0.02347 | 0.02384 | 0.02413 | 0.02442 | 0.0248 |
| 7     | 0.0217 | 0.02239 | 0.02295 | 0.02342 | 0.02380 | 0.02410 | 0.02431 | 0.02454 | 0.0248 |
| 8     | 0.0221 | 0.02272 | 0.02318 | 0.02357 | 0.02390 | 0.02416 | 0.02435 | 0.02454 | 0.0248 |
| 9     | 0.0224 | 0.02297 | 0.02336 | 0.02370 | 0.02398 | 0.02421 | 0.02440 | 0.02459 | 0.0248 |
| 10    | 0.0226 | 0.02323 | 0.02354 | 0.02381 | 0.02404 | 0.02425 | 0.02444 | 0.02463 | 0.0248 |

3.2. The effect of self-multiplication on neutron distribution

The $^{235}U$ fission induced by the AmLi source neutron is called the primary induced fission, and the $^{239}U$ fission neutron induced by $^{235}U$ fission is called the secondary fission. Since it takes extremely short time (within 1 ns) to propagate, the fission neutron energy spectrum is a continuous spectrum, rather than single energy, and the induced fission cross section of each fission neutron varies due to their different energies. Self-multiplication coefficient $M$ can be used to describe the multiplication effect of the sample, which is defined as the ratio of the total fission neutron number from the primary fission and its subsequent fission chain to that of from its primary fission. The neutron number distribution of the primary fission is recorded as $E_0$ and the total number of neutrons emitted after the propagation is $E$. According to the definition of self-multiplication coefficient $M$, we get

$$M = \frac{\sum_{i=1}^{v_{\text{max}}} iE_i}{\sum_{j=1}^{v_{\text{max}}} iE_{0,j}}$$

Where $v_{\text{max}}$ represents the maximum number of the neutron distribution $E_0$ in the primary fission, $v_{\text{max}}=7$, and $v_{\text{max}}'$ refers to the maximum number of possible neutrons after multiplication. We can take a finite value, for example, $v_{\text{max}}'=14$, because the probability becomes very small when it is greater than 14. And the vector $E_0$ can be regarded as the known quantity.

$M$ cannot be used directly to describe the microscopic mathematics of the multiplication, and therefore, it is necessary to define the secondary fission induced probability $q$ (the average probability $q$ of the secondary fission caused by the neutrons in the primary fission). In general, $q$ is less than 10%. For the Highly Enriched Uranium with 10 kg, its self-multiplication coefficient is less than 1.2, and $q$ is less than 0.1. Therefore, assume only one secondary fission is considered, no more than two out of the total neutrons in the primary fission will cause secondary fission.

The distribution of the neutrons in the secondary fission triggered by fission neutrons is $E_1=[E_{10}, E_{11}, \ldots, E_{1v_{\text{max}}}]$. Assume $n$ (the maximum $n=7$) neutrons in the primary fission cause $k$ secondary fission ($k$ neutrons lost), $\lambda$ neutrons is produced after its self-multiplication. Since $\lambda=k+m-n$, there is
tp(m,n) possibility to generate m neutrons (vmax’ refers to the maximum; let 2×vmax=14, vmax’ is large enough).

1. When n≤m and nxvmax≥m, there are more than one secondary fission occurred and no less than two neutrons produced (λ≥2).

\[ tp(n, m) = \sum_{k=\min(2,m)}^{m} C_k^m q^k (1-q)^{m-k} h(k, \lambda) \]

\[ h(k, \lambda) = C_k^1 E_{10}^{k-1} E_{n,1,\lambda} + C_k^2 E_{10}^{k-2} \sum_{j=1}^{\lambda-1} E_{1, j, \lambda-j} \] \( \lambda \geq 2 \)

Where, h(k, λ) represents the probability of λ neutrons generated if k neutrons cause k (k≤2) secondary fissions.

2. When n≥m and n-m≤2, at least k (k=n-m) fissions occur, while at most two fissions occur, which generates λ (λ≤k) neutrons.

\[ tp(n, m) = \sum_{k=n-m}^{\min(2,n)} C_k^n q^k (1-q)^{n-k} h(k, \lambda) \]

\[ h(k, \lambda) = \begin{cases} 
C_k^1 E_{10}^{k-1} E_{1,\lambda} & \text{if } \lambda = 1 \\
C_k^1 E_{10}^{k-1} E_{1,\lambda} + C_k^2 E_{10}^{k-2} \sum_{j=1}^{\lambda-1} E_{1, j, \lambda-j} & \text{if } \lambda > 1 
\end{cases} \]

3. In other cases, tp(m,n)=0.

From the above cases (1), (2) and (3), we find that the matrix tp can be expressed as the expression of q. tp is the matrix of vmax×(2×vmax), namely 7×14 matrix. After we obtain tp matrix, the total distribution of neutrons produced by AmLi and fission neutrons is distributed as

\[ E_n = \sum_{j=1}^{v_{\text{max'}}} E_{0,j}fp(j,n) \Lambda \Lambda 1 \leq n \leq 2 \times v_{\text{max'}} \]

3.3. The neutron number distribution detected by the detector- transfer matrix Q

Let the neutron number distribution detected by the whole detection system be a row vector D, the transfer matrix from neutron number distribution E launched by the sample to the detected neutron number distribution D is Q, which satisfies

\[ D_{v_{\text{max'}}} = E_{v_{\text{max'}}} Q_{v_{\text{max'}} \times v_{\text{max'}}} \]

\[ Q = \begin{bmatrix} 
C_1^1 e & 0 & L & 0 \\
C_2^1 e & C_2^2 e^2 & L & 0 \\
C_3^1 e (1-e)^2 & C_3^2 e^2 (1-e) & L & 0 \\
M & M & M & M \\
C_{v_{\text{max'}}}^1 e (1-e)^{(v_{\text{max'}}-i)} & C_{v_{\text{max'}}}^2 e^2 (1-e)^{(v_{\text{max'}}-2)} & L & C_{v_{\text{max'}}}^v_{v_{\text{max'}}} e^{v_{\text{max'}}-i} \end{bmatrix} \]

Q(i, j) = C_j^i e^i (1-e)^{(i-j)}, \ldots, i \geq j

Q (i, j) represents the probability that fission produces i neutrons and the detector can detect j, and it is only related to the total detection efficiency.
3.4. The neutron number distribution threshold - transfer matrix U
When measuring, let the threshold value be about 1/4Cs, and the corresponding neutron energy 0.76MeV. If the number over the threshold to the total number of neutrons in the fission neutron is u, the probability a single neutron over threshold is u.

Neutrons emitted from a sample may not be detected by the threshold even if all of them are in the detector. However the neutrons under threshold will be detected due to the superposition of the energy deposition associated r rays, which makes the actual threshold rate is higher than that calculated by a quarter of the 1/4Cs theory. In the algorithm, it means the actual value of a single neutron threshold is higher than its theoretical value, that is, its actual threshold is less than 1/4Cs. If the subthreshold neutron does not contribute the neutron counting independently, the transfer matrix \( U \) of the threshold can be described as follows

\[
U = \begin{bmatrix}
C_i^1 u & 0 & L & 0 \\
C_i^1 u (1-u) & C_i^2 u^2 & L & 0 \\
C_i^1 u (1-u)^2 & C_i^2 u^3 (1-u) & L & 0 \\
M & M & M & M \\
C_i^{\text{max}} u (1-u)^{(v_{\text{max}}-1)} & C_i^{\text{max}} u^2 (1-u)^{(v_{\text{max}}-2)} & L & C_i^{v_{\text{max}}'} u^{v_{\text{max}}'} \\
\end{bmatrix}
\]

\( U(i, j) = C_i^j u^j (1-u)^{i-j} \) \( (i \geq j) \),\( U(i,j)=0 \) \( (i<j) \), where \( U(i,j) \) means there is a probability of \( j \) over the threshold in \( i \) neutrons.

3.5. Neutron number distribution recorded by the detector - transfer matrix P
If the detector detects \( j \) neutrons, it does not necessarily mean there are \( j \) coincidence counting because \( j \) neutrons may have entered into the same detector, or in some cases, two or more detectors. The transfer matrix from the detected neutrons number distribution \( D \) to the recorded neutrons number distribution \( R \) is \( P \), that is, \( P \) satisfies the equation: \( R_{1\times 4} = D_{1\times v_{\text{max}}} P_{v_{\text{max}}'\times 4} \).

\[
P_{i,1} = \left( \frac{e_a}{e} \right)^i + \left( \frac{e_b}{e} \right)^i + \left( \frac{e_c}{e} \right)^i + \left( \frac{e_d}{e} \right)^i \quad (i \geq 1)
\]

\[
P_{i,2} = \left( \frac{e_a + e_b}{e} \right)^i + \left( \frac{e_a + e_c}{e} \right)^i + \left( \frac{e_a + e_d}{e} \right)^i + \left( \frac{e_b + e_c}{e} \right)^i + \left( \frac{e_b + e_d}{e} \right)^i + \left( \frac{e_c + e_d}{e} \right)^i - 3P_{i,1} \quad (i \geq 2)
\]

\[
P_{i,3} = \left( \frac{e_a + e_b + e_c}{e} \right)^i + \left( \frac{e_a + e_b + e_d}{e} \right)^i + \left( \frac{e_a + e_c + e_d}{e} \right)^i + \left( \frac{e_b + e_c + e_d}{e} \right)^i + \left( \frac{e_b + e_c + e_d}{e} \right)^i + \left( \frac{e_b + e_c + e_d}{e} \right)^i - 2P_{i,2} - 3P_{i,1} \quad (i \geq 3)
\]

\[
P_{i,4} = 1 - P_{i,1} - P_{i,2} - P_{i,3} \quad (i \geq 4)
\]

Where, \( e_j \) represents the average detection efficiency of \( j \) \( (a, b, c, d) \) detector; \( P_{i,j} \) represents \( i \) detected neutrons and recorded as the probability of \( j \) neutrons. In this case, when \( i < j \), \( P_{i,j}=0 \). And \( P \) depends on the relative efficiency of detectors rather than the absolute value of the total efficiency.
3.6. The correction of P matrix by Crosstalk- transfer matrix T

High-energy neutrons may scatter to other detectors after they enter a detector, which is called crosstalk. If the initial energy of the neutron is higher than twice the value of threshold, it will be possible that the energy deposited in both detectors is enough to pass the threshold, which, in turn, produces two neutron signals.

We make the following two assumptions to simplify the problem because the crosstalk rate is much less than 1. Because the number of the initial energy of neutron is two times higher than the threshold value is very small.

(1) If a neutron has a crosstalk, it enters another detector rather than the third or the fourth one.
(2) Only one of the $n$ neutrons detected by the detector has a crosstalk.
(3) When a crosstalk occurs, the probability for an ejected crosstalk neutron to enter into other detectors is the same. (if no crosstalk neutron can be detected by the detector located over against, the $T$-matrix below should be changed accordingly.)

Then we make the matrix $T$ of $P$ affected by a crosstalk as

$$T_{i,j} = P_i(1-\varphi)^j (i \geq 1)$$
$$T_{i,2} = P_i C_i \varphi(1-\varphi)^{j-1} + P_j \sum_{j=0}^{1} C_i (\frac{1}{3})^j \varphi^j (1-\varphi)^{j-1} (i \geq 1)$$
$$T_{i,3} = P_i C_i (\frac{2}{3}) \varphi(1-\varphi)^{j-1} + P_j \sum_{j=0}^{2} C_i (\frac{2}{3})^j \varphi^j (1-\varphi)^{j-1} (i \geq 2)$$
$$T_{i,4} = 1 - T_{i,1} - T_{i,2} - T_{i,3} (i \geq 3)$$

$T(1,3)=T(1,4)=T(2,4)=0$

3.7. The establishment of equations and solutions.

According to the previous discussion, it can be known that the counting rate $R_j$ satisfies the following equation (matrix).

$$R_{1,4} = F \times (E_{\text{source}} Q_{\text{source}} \times U_{\text{source}} T_{\text{source}})$$

where, $R$ is 1x4, and $R(j)$ is the measured $j$-coincidence counting.

$F$ is the induced fission rate of AmLi source, and its unit is times/s;

$E$ is the number of neutrons emitted by the sample, and which is related to $q$;

$Q$ is the neutron number transfer matrix detected by the detector, which is related to $e$;

$U$ is the transfer matrix of the neutron number passing threshold, and $U$ is only related to the neutron passing threshold $u$.

$T$ is the detector recorded transfer matrix of the $j$-coincidence count. And $T$ is determined by the relative efficiency of the detectors (we calculate the same value by using the total number of detected neutrons minus background count) as well as the average rate of crosstalk $\varphi$.

The measured data includes the total number of neutrons, namely, $c_2, c_3, c_4, c_5$, recorded by each detector, as well as the total count rate (single, double, triple, and quadruple), namely, $R_1, R_2, R_3, R_4$.

According to $R=FEQT$, there are five parameters in the established equations, namely, $F, q, u, e, \varphi$. In fact, it can be proved that $u$ and $e$ in $R$ vector expressions with individual elements always appear together in the form of $u \times e$, that is, $R$ vector contains 4 unknown parameters, namely $F, q, u \times e, \varphi$. Among them, $u \times e$ is the total detection efficiency (for the convenience, the total efficiency we is used to represent $u \times e$). $\varphi$ value is about a few parts per thousand, which is only related to the fission neutron energy spectrum of the sample and the detection system. Therefore, it can be regarded as system parameter (approximately use Cf$^{252}$ to scale), and in this case, there are three undetermined parameters for the sample.
4. Model test and analysis

4.1. Model experiment and its solution

In the experiment, the calibrated intensity of Cf-252 is 17200/s (the number of neutrons emitted per second), and the calibration date is in August 17, 1998, the synthetic uncertainty is 3.5%, the half-life is 2.646 years. The experiment date is December 7, 2017, the experiment source strength is 292.9/s, and the fission times per second is 77.7, so the neutron emission intensity in the experiment is 77.7 ± 2.7. And the measurement data of Cf-252 is shown in Table 2.

| the measurement data          | count     | counting rate |
|-------------------------------|-----------|---------------|
| one weight coincidence        | 3501751   | 20.195        |
| two weight coincidence        | 373600    | 2.155         |
| three weight coincidence      | 14688     | 0.08471       |
| four weight coincidence       | 169       | 9.746e-4      |

When the crosstalk is not considered, the unknown parameter are $F$ and $u\varepsilon$. Choose two from one two three four weight counts can be solved. The total efficiency calculated is about 8.4%, and the spontaneous fission rate is 81.

When the crosstalk is considered, the unknown parameter $F$, $u\varepsilon$ and $\psi$ can be solved by one two three weight counts. The calculation result, that is $F=81.86$, $u\varepsilon=0.08283$ and $\psi=0.001042$.

4.2. Analysis of model test results

In Part 2 we describe the main physical process and its self-multiplication process of the fission neutron detection in matrix form to establish the system of equations describing measurement parameters (single, double, triple, quadruple, which conforms to the net count rate) and the relationship among sample parameters (source induced fission rate, the secondary induced fission probability and detection efficiency $u\varepsilon$, crosstalk rate). And then the equations can be solved to get the undetermined parameters of the sample.

Compared to thermal neutron measurement, there is a few coincidences by fast neutron measurement. Theoretically speaking, there should be no string disturbance in the base neutron counting of AmLi source, and the double and triple counting should come from accidental coincidence [8-11]. Based on the calculation method proposed in this research, we can give the result of nonzero crosstalk rate. As the accidental coincidence is not considered in this algorithm, the nonzero crosstalk rate value is used to describe the accidental coincidence (the number of measured coincidence counting increases if crosstalk and accidental coincidence occur.)

If crosstalk rate can, to some extent, describe the crosstalk and accidental coincidence, the crosstalk rate is not only related to the fission neutron energy spectrum of the sample but also to its fission rate. Therefore, it is not accurate to use the crosstalk rate of Cf ²⁵² as the scale value.

Through the experiment, we find that the crosstalk rate is far lower than 1% based on the crosstalk rate 0.05% from the background of AmLi source analysis and the crosstalk rate of 1% from the Cf²⁵² source analysis. It shows that taking the result from Cf²⁵² as the scale value can meet the requirements. And there are other factors such as the coincidence counting statistic, mathematics calculation model and the ion deviation of actual measurement background that will affect the accuracy of analysis.

5. Conclusions

In this paper, we point out the differences between the fluid scintillation detection and He³ detection, and then build a measurement system. Based on the proposed system, we establish the equations which describe the relationship between measuring parameters and samples (equations). Finally using
the model this paper proposed, we analyze the measuring parameters, especially the influence and the function of crosstalk rate.

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