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Uncertainty of Uranium Mass Measurement by Active Neutron Multiplicity

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Abstract: Recently, the international society pays more and more attention to nuclear material protection, control and accounting. The focus on the material unaccounted for (MUF) is enhanced. The uncertainty assessment of uranium mass measurement plays an essential role in estimating the production of uranium. Due to the relatively weak radioactivity of uranium, the active neutron multiplicity counting (ANMC) method is usually applied to assess the mass of uranium material. In this paper, the relations and parameters between the masses of the uranium objects and the features of the ANMCs are formulated by fitting the simulated results of different sets of uranium shells. The result indicates that, each mass of the objects could be obtained by analyzing the ANMCs of different orders. In order to study the effects of the detection system settings on the mass estimation, the propagation of the aleatoric and epistemic uncertainty of the method is quantitatively studied by the simulations with different detection conditions. The optimized settings of source intensity and detection coincidence gate width, which result in the minimal uncertainty of the mass estimation, are obtained for the simulated detection system.

Key words: nuclear material accounting; active neutron multiplicity counting; quantified uncertainty; mass measurement of uranium material

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1 Introduction

As the risk of nuclear proliferation and nuclear terrorism increasing, the international society attaches more importance to nuclear material protection, control and accounting (MPC&A). Mass assessment of the nuclear material is an essential issue of MPC&A. Many new technologies are applied in nuclear material accounting to reduce the quantity of MUF[1]. The weak radioactivity of uranium places restrictions on the precision of the estimation by passive measurement. Regarding this, active interrogations are introduced and studied to estimate the masses of uranium objects. In active interrogations, signals are induced by some radiation sources[2–3]. One possible solution to uranium mass estimation is the active neutron multiplicity counting (ANMC) method performed by well-detectors[4]. Since 1990s, the method is widely studied theoretically and experimentally. Ensslin *et al.*[5] derived the relation between the mass of plutonium and the corresponding neutron multiplicity counting in passive measurement. Timonthy *et al.*[6] conducted simulations on ANMCs with NCMP-DSP Monte Carlo N-Particle Transport Code. Belew *et al.*[7] experimentally proved that the ANMC distributions were related to the uranium mass by their ANMC experiment with ^{235}U. Krick *et al.*[8] studied the neutron multiplication and source-object coupling factor by ANMC detection with uranium. In China, the studies of design and optimization of ANMC detection system were carried out theoretically and experimentally[9–10]. Recently, Xiao Bo and Zhong Hua *et al.* studied the relation between the masses of high-enriched uranium objects and the detected ANMC distributions[11–12]. Li Gaochen *et al.*[13] improved the algorithm of the estimation by introducing some configuration related parameters.

Although many methods have been employed to improve the accuracy of the measurement, the MUFs

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physically exist in many procedures of nuclear material accounting. One of the most essential works in nuclear accounting is to quantitatively understand the MUFs. An explicit result of the uncertainty of the technologies could provide reference for MPC&A. In this paper, the method of quantification of margins and uncertainties (QMU) were applied to estimate the MUF of the nuclear material by quantitatively studying the uncertainty of the measurement[14–15]. Numerical simulations are performed by the numerical simulation platform to study the relation between the uncertainties and the settings of the detection system. As a result, the optimal settings for the detection system used in the simulation are obtained. For the optimal settings, the uncertainties of the mass estimations can be obtained.

This paper is structured as follows, in the second section of the paper, the mass estimation and uncertainty algorithm are introduced. In the third section, the simulation model, method and detection settings are discussed to show the relations between ANMCs and the parameters. In the fourth section, the uncertainties of mass estimations are studied by numerical simulations to optimize detection settings.

2 Uranium mass measurement by ANMC

In ANMC, the uranium objects are irradiated by external neutron sources, after which a series of neutrons emit. The number of the events that n neutrons are detected within one coincidence gate width is recorded as the value of n order ANMCs[4]. The number and temporal distribution of these emitted neutrons change according to the intensity of source neutron, the mass of the uranium object as well as some other factors. And therefore the analysis of ANMCs, could be used to estimate the mass of the uranium object.

2.1 Active neutron multiplicity counting

In the experiment, almost all the neutron emissions inheriting from one source neutron are detected within tens of microsecond. On the contrary, the time intervals between two successive source neutrons or background neutrons are much longer than those between the neutrons induced by one source neutron, when the intensity of the neutron source is not too strong. The total ANMC could be obtained by counting the neutron number within an elaborated gate widthes. The gate width is set as several tens of microseconds[5]. If the background neutrons are detected within one gate width, the detector would falsely consider the event as a multiplicity counting, which is called random ANMC. The value of random ANMCs could be acquired by counting the neutron number in a gate width after a delay[3]. So the real ANMCs could be obtained by subtracting the random ANMCs from the total ANMCs.

2.2 The uncertainty of mass measurement by real ANMC

Due to the impact of the active neutron sources, the relations between ANMC and the masses of uranium objects are more complicated than those of passive neutron multiplication counting measurements[5]. In this paper, the relations between the uranium masses and the ANMCs are derived from regression analysis with the normalized ANMCs. The real ANMCs are calculated by recording and counting all the detected neutrons inheriting from one source neutron. Three series of uranium metal shells are used in the simulations with the thickness of 0.5, 1 and 3 cm, respectively. The normalized ANMCs are calculated by dividing the n order ANMCs to the signal multiplicity counting which is displayed in Fig. 1 as circles. The fitted results are shown as lines.

The data listed in the upper panels from left to right are the second, third and fourth order normalized ANMCs, respectively, while the data shown in the bottom panels are the fifth, sixth and seventh order normalized ANMCs, respectively. Fig. 1 shows linear relations between ANMCs and the masses. The relation between the mass and the ANMCs could be established as,

$$m_{\mathrm{M}} = a_{\mathrm{M}}(t) \cdot \mathcal{N}_{\mathrm{M}} \tag{1}$$

with,

$$\begin{cases} \mathcal{N}_{\mathrm{M}} = \dfrac{N_{\mathrm{M}}}{N_1} \\ a_{\mathrm{M}}(t) = k_{\mathrm{M}} \cdot \exp(-t) + b_{\mathrm{M}} \end{cases}, \tag{2}$$

where, N_{M}, N_1, and $\mathcal{N}_{\mathrm{M}}$ are the M order ANMC, the first order ANMC and the normalized M order ANMC with t, k_{M} and b_{M} representing for the thickness of the uranium shell, and the coefficients, respectively. Finally, the relation between the mass, ANMC and thickness could be further identified by combining the formulas with different orders of ANMCs together,

$$\begin{cases} m = \dfrac{b_{\mathrm{M}} k_{\mathrm{M'}} N_{\mathrm{M'}} N_{\mathrm{M}} - b_{M'} k_{\mathrm{M}} N_{\mathrm{M}} N_{\mathrm{M'}}}{k_{\mathrm{M'}} N_{\mathrm{M'}} - k_{\mathrm{M}} N_{\mathrm{M}}} \\ t = -\ln\left(\dfrac{b_{\mathrm{M}} N_{\mathrm{M}} - b_{\mathrm{M'}} N_{\mathrm{M'}}}{k_{\mathrm{M'}} N_{\mathrm{M'}} - k_{\mathrm{M}} N_{\mathrm{M}}}\right) \end{cases}, \tag{3}$$

where variables with subscripts M' indicate the variables related to M' order ANMCs, respectively.

The unknown thickness in non-destructive detection process contributes to the epistemic uncertainty of mass measurement. Based on Function (1), the ranges of mass estimations could be derived from the empirical estimations of the thicknesses.

$$\begin{cases} m_{\max} = (k_{\mathrm{M}} \cdot \exp(-t_{\mathrm{e}_{\min}}) + b_{\mathrm{M}}) \cdot \mathcal{N}_{\mathrm{M}} \\ m_{\min} = (k_{\mathrm{M}} \cdot \exp(-t_{\mathrm{e}_{\max}}) + b_{\mathrm{M}}) \cdot \mathcal{N}_{\mathrm{M}} \end{cases}, \qquad (4)$$

where, $m_{\max}$ and $m_{\min}$ are the maximum and minimum estimations of the masses, with $t_{\mathrm{e}_{\min}}$ and $t_{\mathrm{e}_{\max}}$ representing for the minimum and maximum thicknesses, respectively. In this paper, this part of uncertainty is called the epistemic uncertainty from the parameters of the algorithm, σ_{param}.

The limited detection accuracy and restricted detection duration would bring in other two kinds of uncertainties, which are the uncertainty of acquiring the real ANMC from detected ANMC, and the aleatoric uncertainty. The detected ANMC by neutron well detector could not exactly reproduce the real ANMC[4]. In this paper, σ_{detc}. is used to indicate the uncertainty originating from the detection system.

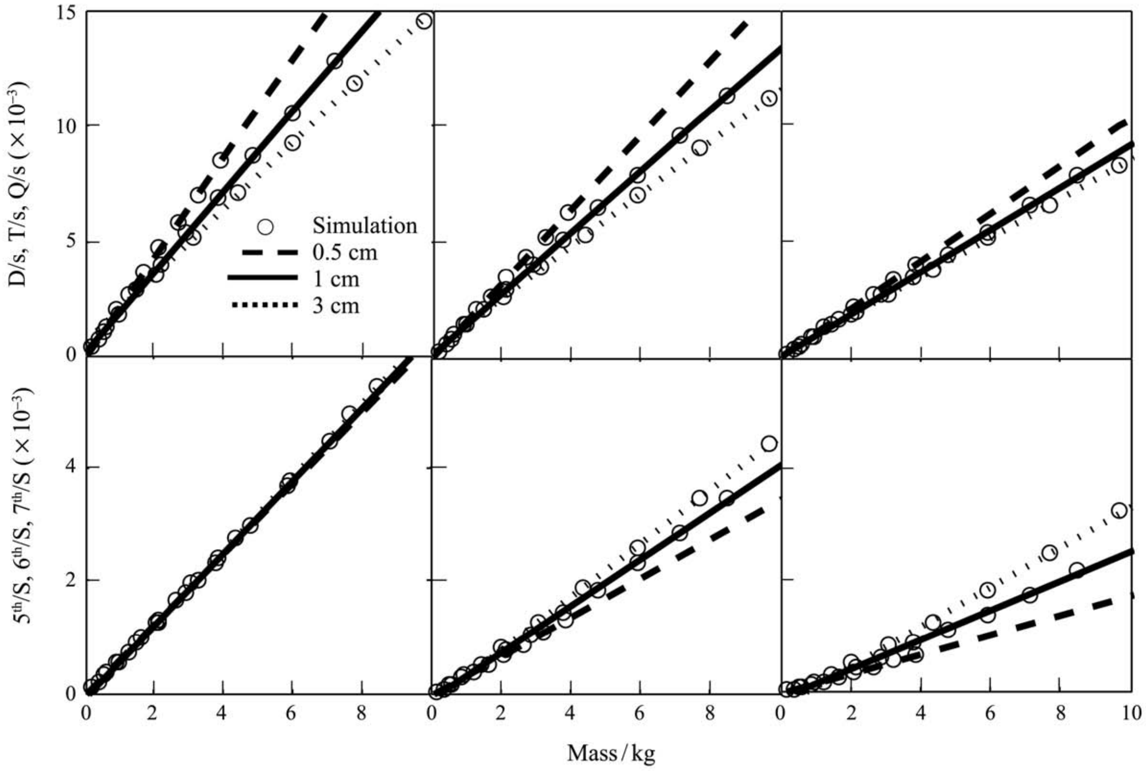

Fig. 1　The relations between uranium shell masses and the normalized ANMCs.

The circles, dashed lines, solid lines and dotted lines representing the simulated results, the fitted results for the series with the thicknesses of 0.5, 1 and 3 cm, respectively.

The detected neutron numbers, which are proportional to the detection duration, result in the aleatoric uncertainty. In this paper $\sigma_{\mathrm{alea.}}$ is used to represent the aleatoric uncertainty. According to the classic error expression, $\sigma_{\mathrm{alea.}}$ could be obtained by,

$$\sigma_{\mathrm{alea.}}(M) = \sqrt{\frac{1}{N_{\mathrm{M}}} + \frac{1}{N_1}}\ , \qquad (5)$$

where, M, N_{M} and N_1 refer to the multiplicity of neutron, the count for M order ANMC and the count of single event, respectively.

The total uncertainty of the mass estimation could be acquired by summarising the three kinds of the uncertainties.

$$\sigma_{\mathrm{total}}^2 = \sigma_{\mathrm{param.}}^2 + \sigma_{\mathrm{detc.}}^2 + \sigma_{\mathrm{alea.}}^2\ . \qquad (6)$$

The influences of the detection parameters on uncertainty and the propagation of the uncertainty in mass estimation will be studied in detail in the next section.

3　ANMC simulation and acquisition

In order to acquire the ANMCs, numerical simulations are carried out by the numerical simulation platform[16]. The real ANMC and the detected ANMC are two types of ANMC acquired in the calculation, the former of which could be acquired by recording and counting the number of neutrons induced by one neutron source, while the latter by counting the neutron number within each coincidence gate width[16]. The ANMCs with different intensities of the sources and the coincidence gate widthes are produced for various sets of uranium objects to study the influence of the detection settings on the uncertainties.

3.1 The configuration of the detection system and properties of the object

Simulations are carried out to show the process of uncertainty estimation and the influence of detector settings to the uncertainty. The well-type detector with high pressure ^{3}He gas tubes and polyethylene are used in the simulation. The structure of the detector is shown in Fig. 2. Usually well-type detector consists several dozens of ^{3}He tube. In our simulation, the ^{3}He tubes are simplified as an cylindrical shell located in the polyethylene shell. The inner and outer diameters of the detection area are chosen by fitting the typical dei-away-time and efficiency of the well detectors[4].

High-enriched uranium (HEU) shells with ^{235}U concentration of 90% are chosen as an example. In the simulation, 14.1 MeV neutrons are used as the external neutron source.

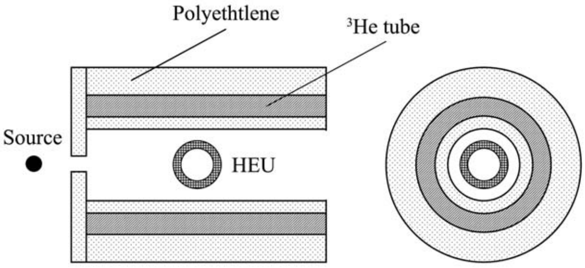

Fig. 2 The configuration of simulated detection system.

3.2 The influence of source intensity on active multiplicity detecting result

The ANMCs are simulated with different source intensities. HEU shell with 3 cm inner radius and 4 cm outer radius is used as an example to show the influence of source intensities on detected ANMCs. Fig. 3 demonstrates the real and detected ANMC distributions. In the simulation, the source intensities are $I = \{10^3,\ 10^5,\ 10^6\}$ (neutron/s). To keep the total counting of the neutron constant, the detection duration is selected as $t = 10^8/I$, respectively.

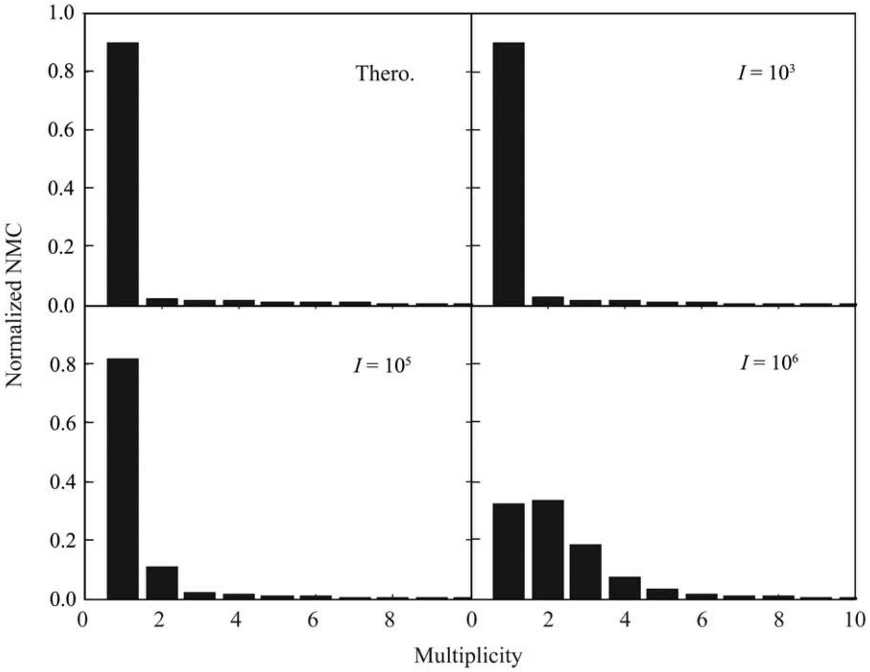

Fig. 3 The neutron multiplicity counting for sources with various neutron intensities.

As shown in Fig. 3, when the intensity of the neutron source is high, due to the epistemic uncertainty from detection, the detected ANMCs diverge from the real ANMC. In the simulation, 64 μs coincidence gate width is used. As the incident neutron intensity decreasing to 10^3 neutron/s, the detected ANMCs and the real ANMC tend to be agreed. When the intensity is higher, the discrepancies become greater. Although, the real ANMC couldn't be acquired experimentally, the relations between ANMCs and the uranium mass maintain valid when the intensity of source is higher, if the parameters could be calibrated by experimental or theoretical data with a stronger source. In Section 4.1, we will discuss the uncertainty of the redeliberation methods in detail.

3.3 The influence of coincidence gate width on counting rate

In ANMC measurement, in order to distinguish the influence of the neutrons produced by different sources, the coincidence gate width should not be too long[4]. Generally, thousands of nanoseconds are used as the gate width, so that the real ANMC could be reproduced by the experimental data in NMC detection[5]. The effects of the gate widthes on ANMCs are illustrated in Fig. 4. Different symbols represents the gate widthes from 6.4 to 819.2 μs.

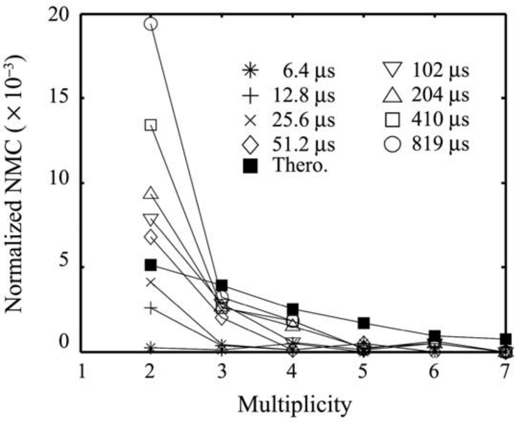

Fig. 4 The ANMC with various correlation gate widthes.

4 The uncertainty of mass estimation

In this section, we focus on the the detection settings with significant influence on the uncertainty of mass estimation. As shown in Function (8), three factors affect the uncertainty of the mass estimation, the uncertainty from the unknown thickness of the uranium shell, the uncertainty from the discrepancy between the real ANMCs and the detected ANMCs, and the uncertainty from limited neutron counts and the detection duration. If the relation between the masses and the ANMCs could be calibrated for the detection system, the uncertainty originating from the discrepancy between the real ANMCs and the detected ANMCs could be reduced. In the cases, the uncertainty related to the unknown thickness and the aleatoric uncertainty with different orders ANMCs could be calculated by,

$$\sigma^2_{\mathrm{cal.}} = \frac{1}{n-1}\sum_{i=1}^{n}\left(a_{\mathrm{M}}(t_{\mathrm{e}})\cdot\frac{N_{\mathrm{M}}}{N_1}-\bar{m}\right)^2 , \qquad (7)$$

where, t_{e} is the empirical thickness. In this paper, based on the hypothetic model suggested by Steven Fatter in 1990, 1 cm is used as the empirical thickness[18].

4.1 The influence of source intensity on the mass uncertainty

In order to discuss the influence of source intensity on the uncertainty of uranium mass, the uncertainties for ANMCs with different source intensities are simulated and compared with each others. In the Fig. 5, the solid squares represent the real distribution of ANMC, and the hollow symbols represent the detected ANMCs simulated with source intensities, $I = \{10^3,\ 10^4,\ 10^5,\ 10^6,\ 10^7\}$ (neutron/s), respectively. The value of the intensities represented by different symbols are displayed in the figure. In the simulation, the detector efficiency is 80%, the coincidence gate width is 6.4 μs, the resolution time of the detector is 50 ns, and the detection duration is 1 000 seconds. As a result, the total number of source neutron being simulated is $1\,000\times I$.

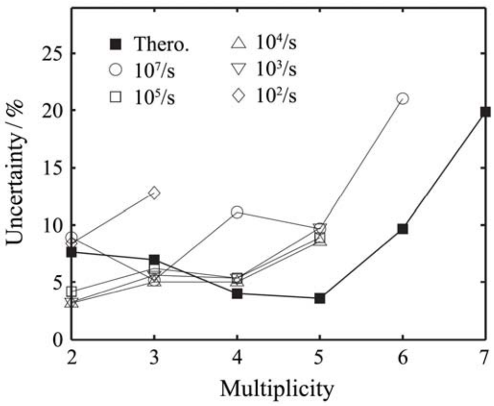

Fig. 5 The average uncertainty of uranium shell obtained by ANMC distribution with various neutron source intensities.

The epistemic uncertainty and aleatoric uncertainty play different roles as the intensity of the source varying. When source intensity increases, the epistemic uncertainty will dominate the total uncertainty. While for weak neutron source, the effective neutron counting deceases, the aleatoric uncertainty dominates. Fig. 5 shows that the mass uncertainty will achieve its minimum with 10^4 neutron/s neutron source. When the source intensity is low, such as 10^2 neutron/s, due to the large aleatoric uncertainty the relations between the mass and the ANMC distributions become unstable, higher order ANMCs distributions can't be used for mass estimation. So optimal source intensity to give the minimal uncertainty of mass estimation could be suggested as 10^4 neutron/s for this detection system within 1 000 s detection duration.

4.2 The influence of coincidence gate width on uranium mass

Similar with the influence of source intensity, the coincidence gate width is another factor which could affect the uncertainty of mass estimation. The uncertainties of mass estimation obtained with different gate widthes are displayed in Fig. 6, with the solid square and hollow symbols representing the uncertainty obtained from real ANMCs and from detected ANMCs. In the simulation, 10^4 neutron/s neutron sources are used.

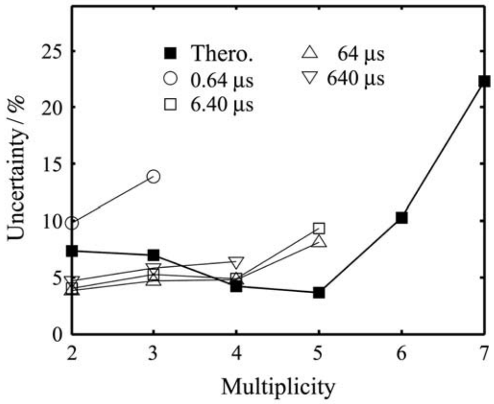

Fig. 6 The average uncertainty of uranium shell obtained by ANMC distribution with various correlation gate widthes.

The detected ANMCs displayed in Fig. 6 are calculated with different gate widthes. The solid squares and the hollow symbols represent the results obtained from real distribution of ANMC, the detected ANMCs simulated with different gate widthes, respectively. As shown in Fig. 6, both extending or narrowing the gate width would lead to larger mass uncertainty. Only when the time width is approaching to hundred microsecond, the estimating uncertainty achieves its minimum. As a result, besides extending the detecting duration, it is possible to reduce the uncertainty of mass estimation by selecting the optimal setting to the detection system.

5 Conclusions

In this paper, we analyze the origins and influencing factors of the mass uncertainty of uranium mass measurement by ANMC method. By realizing the ANMC calculation in the numerical experiment platform, we obtain detected ANMC and the real ANMC of uranium metal shell with different settings of the detection systems. Our simulations indicate that for each series of uranium shell with the same thickness, the ANMC distributions are linearly dependent on the mass of the shell. The slope of line is related to the thickness of the shell. Three kinds of uncertainties exist in the mass estimation, the epistemic uncertainty from unknown parameter, the epistemic uncertainty from real ANMC detection and the aleatoric uncertainty. In the processes of mass estimations, the thicknesses of the shell couldn't be effectively measured. The indetermination of the thickness becomes one of the most significant origin of the epistemic uncertainty. The other kind of epistemic uncertainty originates from the difficulties in resuming the real ANMC from detected ANMC, which is related to the detection settings. Aleatoric uncertainty is related to the total counting of the detection, which could be improved by increasing the detection duration.

In this paper, the optimal settings to give the minimum epistemic uncertainty are discussed in detail by numerical simulation and analysis. For the detection system used in this paper with 1000 second detection duration, the optimal source intensity is about 1×10^4 neutron/s, and the optimal coincidence gate width is about several hundreds of microsecond.

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主动中子多重性法估算铀材料质量的不确定性

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摘要： 近年来，国际社会对核材料保护、控制和衡算日益加强。对不明材料损失量 (MUF) 的关注逐渐提升。铀材料质量不确定性测量在估算铀材料生产量中扮演重要角色。由于铀材料自发裂变相对较弱，主动中子多重性法被应用于估算铀材料质量。通过拟合对不同系列铀金属壳的数值模拟结果，获得了描述铀材料质量与主动中子多重性特征之间的算法和参数。得到的关系表明，可以通过分析不同重数中子多重性探测结果获得铀部件的质量。对不同探测条件下的模拟结果的定量分析，确定了探测系统设置对铀质量估算的影响，以及认知不确定性和随机不确定在估算过程中传播对质量估算的影响。对不确定度的分析获得了本文模拟采用的探测系统的最佳源强和探测时间窗设置，在此设置下，质量估算的不确定性最小。

关键词： 核材料衡算；主动中子多重性法；量化不确定性；铀材料质量测量

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