Experimental study on the measurement of uranium casting enrichment by time-dependent coincidence method

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Abstract: Based on the time-dependent coincidence method, a preliminary experiment has been performed on uranium metal castings with similar quality (about 8–10 kg) and shape (hemispherical shell) in different enrichments using neutron from Cf fast fission chamber and timing DT accelerator. Groups of related parameters can be obtained by analyzing the features of time-dependent coincidence counts between source-detector and two detectors to characterize the fission signal. These parameters have high sensitivity to the enrichment, the sensitivity coefficient (defined as $(\Delta R/\Delta m)/\bar{R}$) can reach 19.3% per kg of ^{235}U. We can distinguish uranium castings with different enrichments to hold nuclear weapon verification.

Key words: uranium casting, enrichment, time-dependent coincidence method, verification

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1   Introduction

Uranium is the major material of nuclear components. The method for radiation detection of nuclear components is an important part of nuclear disarmament verification. Due to the low efficiency of the emission rate for neutron and gamma ray from the spontaneous fission of uranium (neutron emission rate is 0.312 s^{-1}·kg^{-1} for ^{235}U and 14 s^{-1}·kg^{-1} for ^{238}U), non-source detection is difficult and a more active method with a radiation source is considered.

As an active method, time-dependent coincidence measurement can be used in non-destructive detection of fissile material, and it plays an important role in the uranium component verification. Based on this, Oak Ridge National Laboratory has developed a nuclear material identification system (NMIS) for the verification of fissile material [1, 2]. There exist some simulation results for this method, but it is poor in experimental results.

A preliminary experimental research conducted on the basis of time-dependent coincidence measurement has been performed with HEU (highly enriched uranium) and DU (depleted uranium) castings with similar quality (about 8–10 kg) and shape (hemispherical shell). The neutron sources used in our study are Cf fast fission chamber and timing directionally tagged DT accelerator. By analyzing the features of time-dependent coinci-

dences of signals between source-detector and two detectors, many groups of related parameters can be obtained to characterize the fission signal. These parameters have high sensitivity to the change of enrichment, and the uranium castings with different enrichments can be distinguished.

2   The principle of measurement

When the uranium casting is irradiated by neutron source, ^{235}U easily undergoes fission and 2.5 prompt neutrons and prompt photons are emitted within 10^{-14} s in average. The experiment designed here is conducted based on the different cross section between ^{235}U and ^{238}U (See Fig. 1 [3]).

Taking NMIS system for example, the time-dependent coincidence method is shown in Fig. 2 [4]. The neutron source releases spontaneous fission neutrons and gamma rays and provides the accurate release time. Three possible processes will happen then:

(1) Neutrons and gamma rays from source can penetrate through the casting without any interaction with the uranium casting and then are recorded by the detector directly.

(2) Neutrons are scattered by the uranium casting and recorded by the detector.
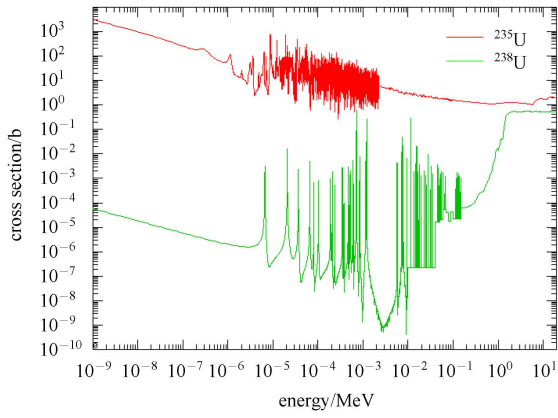
Fig. 1.   The fission cross section of ^{235}U and ^{238}U.

(3) The prompt neutrons and gamma rays from fission are recorded by the detector.

Thus, signals from $N+1$ channels are obtained from the neutron sources and N detectors, and time-dependent coincidence can be calculated between any two signals. The correlation function $C_{xy}(\tau)$, which can statistically reflect the time-dependent characteristics between two random signals $X(t)$, $Y(t)$, is defined as follows,

$$C_{xy}(\tau) = \lim_{T \to \infty} \frac{1}{2T} \int_{-T}^{T} X(t)Y(t+\tau)\mathrm{d}t, \qquad (1)$$

where T is the observation time and τ is the delay time. From this formula the cross-correlation function between two detection channels is obtained as time-dependent signal.

2.1   Time-dependent signal [5]

Figure 3 shows the cross-correlation function between a detector and the Cf source ionization chamber measured by Oak Ridge National Laboratory. The ordinate represents the counts of C_{01} per Cf fission, and the subscript "01" means the cross-correlation function is between the signal 0 and the signal 1.

What we are concerned with is the tail of this function, because it reflects the induced fission strength in the measured uranium casting.

Figure 4 shows the cross-correlation function between two detection channels, and the subscript "12" means the cross-correlation function between signal 1 and signal 2.

The sharp peak at time lag of zero is primarily due to the simultaneous detection of two correlated prompt gamma rays from the same fission. The other correlations at longer time lags come from neutron-neutron pairs and gamma ray-neutron pairs.

2.2   The signature parameter

After analyzing the time-dependent signal obtained in experiment, we can get a corresponding signature parameter to study quantitatively uranium enrichment.

In this paper，we define the signature parameter R as the integration of time-dependent signal C in a specific time range $\tau_1 - \tau_2$:

$$R(\tau_1 - \tau_2) = \int_{\tau_1}^{\tau_2} C(\tau)\mathrm{d}\tau. \qquad (2)$$
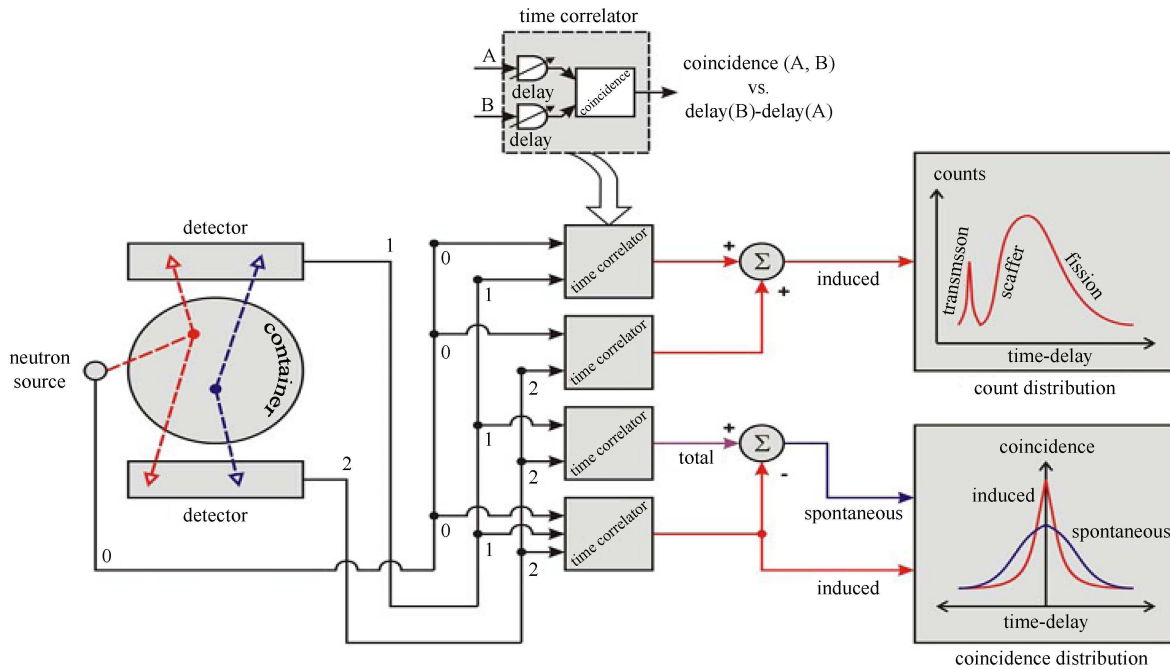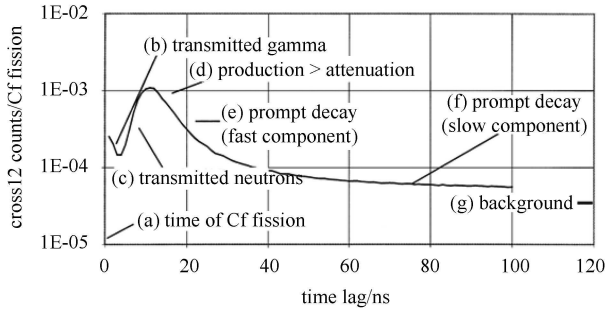

Fig. 2.   The functional design of NMIS.

Fig. 3. The right half ($\tau \geqslant 0$) of the cross-correlation function between a detector and the Cf source ionization chamber. The feature (a) shows the time when the spontaneous fission of Cf occurs. (b) shows the gamma ray which transmitted across the casting directly. (c) shows gamma ray scattered by the casting and transmitted neutrons. Then the scattered neutrons, and at last the induced fission neutrons and gamma rays (e and f). The background from accidental coincidences is described by feature (g).
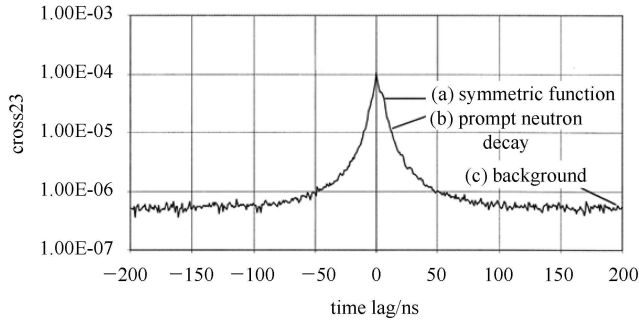

Fig. 4. The cross-correlation function between two detection channels. (a) shows the typically symmetric distribution at $\tau=0$, (b) shows the time decay and (c) is the background from accidental coincidences.

3   The experimental research on Cf fast fission chamber source

The experiment has two parts. In Part 1, a Cf fast fission chamber is used as neutron source. Because the neutron source strength of Cf is low (the fission rate is 10^4–10^5 s^{-1}), the time-dependent signal between two detectors cannot be measured efficiently (the count rate is about 10^{-2} s^{-1}). Thus, we use only one detector in Part 1 and measure the correlation between the signals from the source and that detector.

3.1   The experimental method

The Cf fast fission chamber [6] has ns-level response, which can give the exact time measurement of spontaneous fission. We take this signal as Channel 0 to be the start of time-dependent coincidence (CH0) and the

output signal from the detector as CH1. The high-speed data acquisition system synchronously collects and computes these two signals, and obtains the cross-correlation function $C_{01}(\tau)$. The measuring layout is shown as Fig. 5.
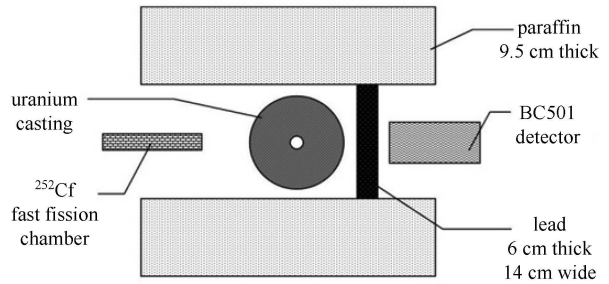

Fig. 5.   Sketch of top view of measurement configuration with uranium metal casting in Part 1.

The whole detection system is located on an iron desktop which is 80 cm high from the ground, and the Cf fission chamber, the measured uranium castings and the detector are linearly placed. The Cf source has about 7×10^4 s^{-1} fission rate and is 24.5 cm away from the detector center. The detector used in this experiment is BC501 liquid scintillation detector which is sensitive to both neutrons and gamma ray. The size of scintillator is $\Phi50.8$ mm$\times50.8$ mm and the threshold for the detector is set such that neutrons with energy above 1.8 MeV will be detected. The uranium castings are hemispherical shell and pronely located in between the source and the detector. A 6 cm thick lead block is placed in between casting and detector so that most gamma rays can be shielded. There are many 9.5 cm thick paraffin blocks around the whole system.

The measured castings are 7.2 kg of HEU and 8.9 kg of DU, and their shapes are all hemispherical shell. Due to the limited conditions, the differences between the castings in mass and shape are ignored, and the only variable is enrichment. We measure three objects: HEU casting, DU casting and none, and the measuring time of each condition is 2048 s.

3.2   The experimental result

The correlation spectrum for HEU is shown as Fig. 6.

Divide the total $C_{01}(\tau)$ signal into two parts: the effective signals and the accidental coincidences. The effective signals caused by the source only appear in the right half of the function, and most of them are concentrated within 0–100 ns. The accidental coincidences between the source signals and the background signals are derived from the convolution of two random signals, and it shows a wide and low triangular peak symmetrical around 0 ns. So the total $C_{01}(\tau)$ accidental coincidences can be calculated from the left half, and this

background should be deducted during the data processing. The background is mainly caused by transmitted low-energy source neutrons, scattered neutrons by the casting and surrounding, and gamma rays from the fission chain.

The time-dependent signals of "HEU" and "none" are given in Fig. 7.
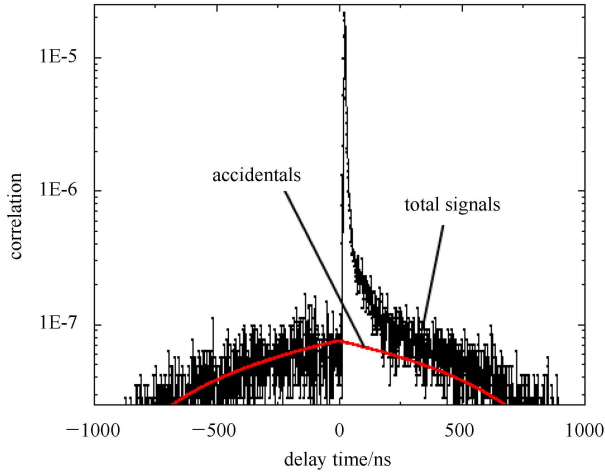

Fig. 6.   The coincidences in C_{01} signals.
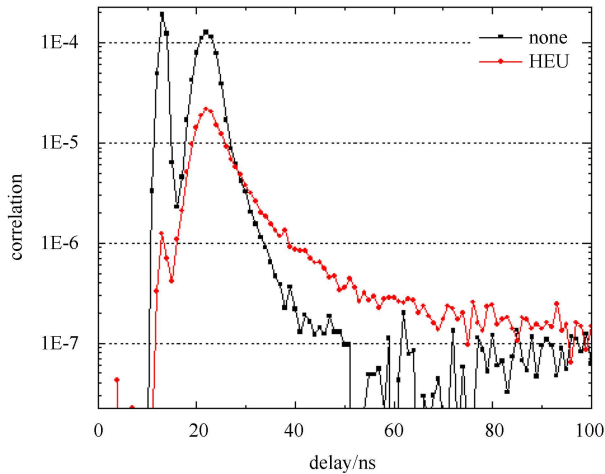

Fig. 7. $C_{01}(\tau)$ for HEU and none in Part 1.

As shown in Fig. 7 there is a crossover point at about 29 ns, and it means at this point the response for "none" is the same as that for "HEU". Before this point, the directly transmitted neutrons and scattered neutrons from the source take the main parts; while after this point, the signals "HEU" are much higher than "none" mainly because of the induced fission chain multiplication [2]. Thus the time-dependent signal after the crossover point can characterize the induced fission strength of measured casting.

The signals of "DU" and "HEU" are shown in Fig. 8.

From Fig. 8, it can be seen that there is a significant difference between the tails of two signals. The signal has a rise after 80 ns because of the neutron reflection from surroundings [7].
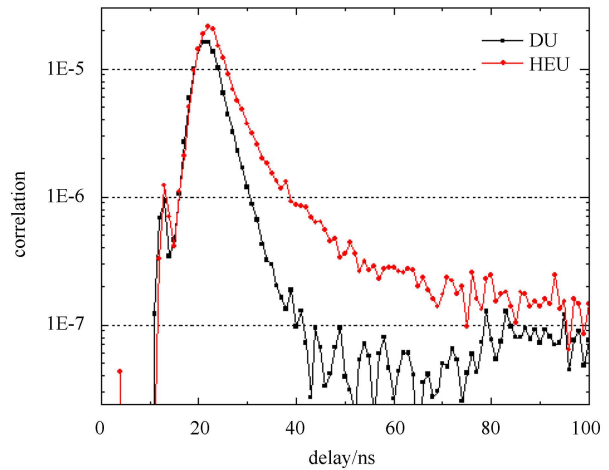

Fig. 8. $C_{01}(\tau)$ for HEU and DU in Part 1.

3.3   Distinguishing HEU and DU by the signature parameter

Considering the statistical fluctuation and the signal noise ratio (SNR), the signal integration within 29–70 ns is selected as signature parameter R_{12} in order to achieve the best discrimination. In this range, the SNR of HEU is 12.1 and the total correlation counts are 5667, while the SNR of DU is 1.6 and the total correlation counts are 4533.

Taking HEU enrichment for 93.15 wt%, and DU for 0.3 wt%, the measured uranium metal castings are characterized by the signature parameter $R_{01}(29$–70 ns), and the result is shown in Table 1.

The sensitivity coefficient was defined as [1]:

$$W = \frac{\dfrac{\Delta R}{\Delta m}}{\bar{R}}, \qquad (3)$$

where m is the ^{235}U mass in kilograms.

The signature parameter is a function of the ^{235}U mass in the uranium casting, and the sensitivity coefficient shows how sensitive it is. From Table 1 we can effectively distinguish HEU and DU castings by R_{01}.

Table 1.   Distinguishing the uranium castings in Part 1.

type of enrichment	mass/kg	$\varPhi$/mm	R_{01}
HEU	7.2	about 20/110	3.62E-5
DU	8.9	about 20/120	7.78E-6
sensitivity coefficient w_{01}=19.3%			

4   The experimental research on DT accelerator source

DT accelerator is used as neutron source in Part 2. Compared with the Cf fission chamber, the neutron source strength of DT accelerator can be as high as 10^6–10^7 s^{-1}. The count rate increases greatly and the time-dependent signal between two detectors can be measured efficiently. Thus there are two detectors in Part 2, and we measure the correlation between the signals from not only the source and one detector but between two detectors meanwhile.

4.1   The experimental method

The reaction T(d, n) ^{4}He radiates neutrons with 14.1 MeV and α particles with 3.5 MeV, and we fix the α-detector 5.6 cm away from the reaction site on accelerator to obtain the reaction time. We take this signal as CH0 and the output signals from two detectors as CH1&CH2. The cross-correlation function $C_{01}(\tau)$, $C_{02}(\tau)$, $C_{12}(\tau)$ can be obtained.

In order to decrease the influence of accidental coincidences, and reduce the computational load and the data storage, when measuring $C_{12}(\tau)$ we set CH0 as gated signal to select CH1&CH2. If the system gets a CH0 signal, it will record the CH1&CH2 signals in the following 100 ns, and the newer CH0 signal in this 100 ns will extend the gated time for another 100 ns.

According to the momentum conservation, neutrons and α particles from DT reaction have opposite directions. Thus CH0 from α-detector only indicates a part of the neutrons in a specific direction. When the CH1&CH2 detectors are located outside the tapered regions of the specific direction, the interference from the source will decrease obviously. According to the characteristics of DT accelerator, the measuring layout is shown as Fig. 9.
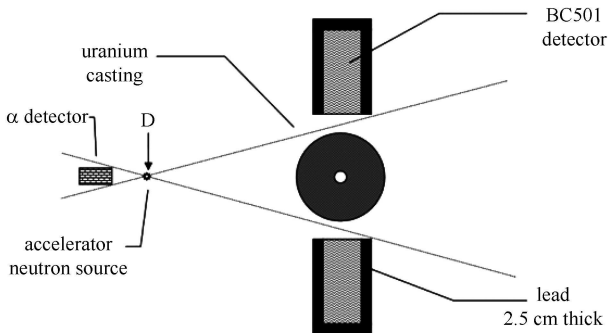

Fig. 9.   The sketch of the top view of the measurement configuration with uranium metal casting in Part 2.

The whole detection system is located in midair. The DT accelerator, the measured uranium castings and the detectors are placed at the same height. The fission rate is not stable and fluctuates in 10^6–10^7 s^{-1}. The two detectors are both BC501 liquid scintillation detectors whose sizes are $\Phi 2\times 2$ inches and $\Phi 5\times 3$ inches, and the low threshold is turned down to 0.5 MeV for suitable count rate. The detectors are located outside the tapered regions of the source neutrons, and wrapped up with 2.5 cm thick lead. The hemispherical shell castings are pronely located, and the casting center is 38 cm away from the source and 16.5 cm away from the detector center.

The measured castings are 10.7 kg of HEU and 8.9 kg of DU, and their shapes are all hemispherical shell. We measure three objects: HEU casting, DU casting and lead hemisphere (similar shape, radius 65 mm, mass 6.5 kg, as control experiment), and the measuring time of each condition is 1000 s. The lead casting is a control project for no-induced fission condition.

4.2   The experimental result

The analysis of the accidental coincidences of $C_{01}(\tau)$ and $C_{02}(\tau)$ in Part 2 is similar to that in Part 1, but the situation in $C_{12}(\tau)$ is more complex because the total accidental coincidences cannot be calculated directly. The $C_{12}(\tau)$ consists of the following four convolutions: $E1*E2$ (E=effective), $E1*B2$(B=background),
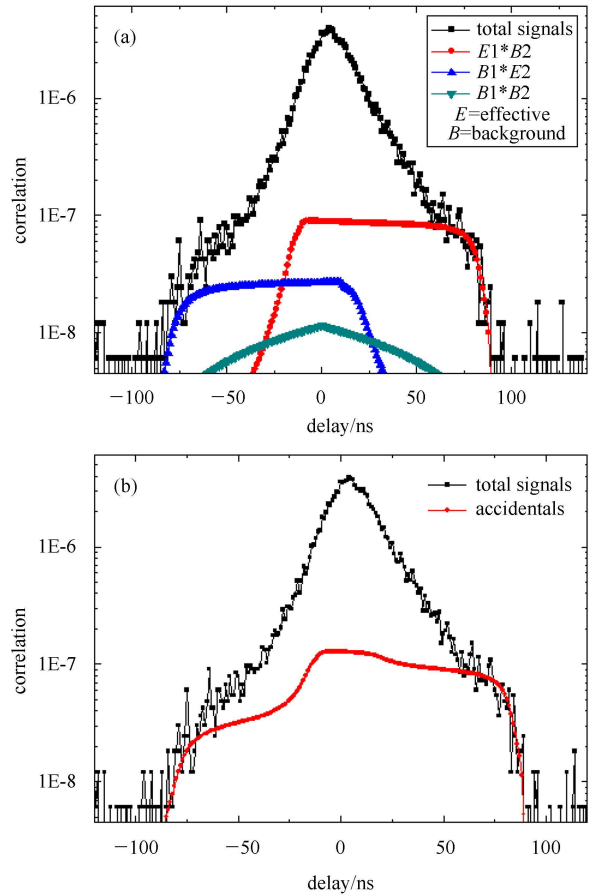

Fig. 10.   The coincidences in the C_{12} signals.

$B1*E2$, $B1*B2$, and the sum of the last three parts is the total accidental coincidence. Through these analyses of $C_{01}(\tau)$ and $C_{02}(\tau)$ the three parts can be obtained separately, and then the total $C_{12}(\tau)$ accidental coincidence is calculated as shown in Fig. 10.

$C_{01}(\tau)$ of "HEU" , "DU" and "Lead" are shown in Fig. 11.

From Fig. 11, $C_{01}(\tau)$ in Part 2 has a low and narrow γ peak at about 12 ns, and a high and wide peak from the scattered and induced fission neutrons at about 22 ns. There are significant differences between these signals. Because the two detectors have the same type and symmetrical positions, $C_{02}(\tau)$ has similar characteristics to $C_{01}(\tau)$.
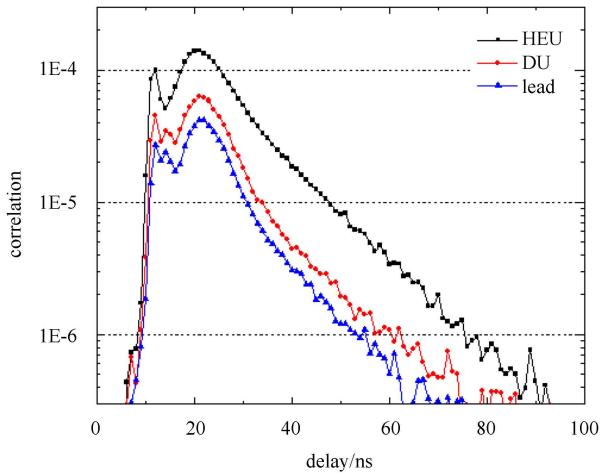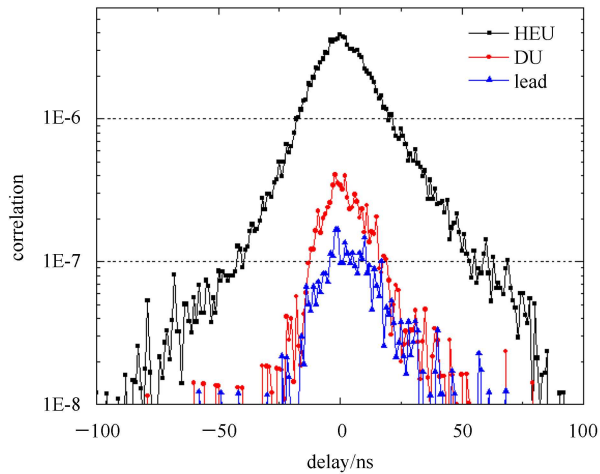

Fig. 11. $C_{01}(\tau)$ in Part 2.

Fig. 12. $C_{12}(\tau)$ in Part 2.

$C_{12}(\tau)$ of "HEU", "DU" and "Lead" are shown in Fig. 12.

Figure 12 shows that $C_{12}(\tau)$ in Part 2 have almost symmetric distribution about $\tau=0$, and from the middle to the two sides the signals are from gamma-gamma

correlations, neutron-gamma correlations and neutron-neutron correlations in turn. Obviously, there are significant differences between the three signals.

4.3 Distinguishing HEU and DU by the signature parameter

The measured uranium metal castings are characterized by the signature parameter R_{01} (36–80 ns) and R_{12} (−50–50 ns), and the result is shown in Table 2.

Table 2.   Distinguishing the uranium castings in Part 2.

type of enrichment	mass/kg	Φ/mm	R_{01}	R_{12}
HEU	10.7	about 20/130	3.27E-4	1.13E-4
DU	8.9	about 20/110	8.52E-5	8.56E-6
Sensitivity coefficient			w_{01}=11.8%	w_{12}=17.3%

In 36–80 ns of $C_{01}(\tau)$, the SNR of HEU is 1.2 and the total correlation counts are 98933, while the SNR of DU is 0.7 and the total correlation counts are 32521. In −50–50 ns of $C_{12}(\tau)$, the SNR of HEU is 79 and the total correlation counts are 18848, while the SNR of DU is 6.0 and the total correlation counts are 1608.

Table 2 shows that we can effectively distinguish HEU and DU castings with R_{01} and R_{12}.

5   Discussion

5.1   Application of the measurement system

Two fundamental approaches could provide confidence that declarations concerning items in a nuclear-weapon arms control regime are true. The attribute approach is based on the intrinsic characteristics of nuclear weapons and their components. The template approach compares the radiation signature from an inspected item with a known standard for a weapon or component of the same type [8].

From the experimental result we know that using the time-dependent coincidence method, the measured HEU and DU castings can be effectively distinguished by different neutron sources (Cf fast fission chamber and DT accelerator), different time-dependent signals (C_{01} and C_{12}) and different expressions (the cross-correlation function and signature parameter). However, the method has some limitations in that the castings should have only one variable of enrichment. It could be a problem in measuring enrichment whether the measured castings have different masses or shapes. According to this characteristic, the system is suitable for template measurement, such as nuclear material transfer. For attribute measurement, a known standard or other measuring method is needed.

Meanwhile, the research still has a long way to go, such as optimizing the signature parameter, preferable

analysis for SNR, frequency-domain and auto-correlation functions.

5.2 The comparison between two time-dependent signals

From the experimental results of Part 2, compared with C_{01}, the advantages of C_{12} are:

(1) C_{12} has higher sensitivity coefficient. It means C_{12} has a better distinguishable effect for uranium castings of different enrichments.

(2) C_{12} has a better SNR. It means the influence from accidental coincidences observably decreases in C_{12}.

The disadvantage of C_{12} is lower count rate, longer measuring time or more powerful neutron source is needed for efficient measurement of C_{12}.

5.3 The comparison between two neutron sources

The characteristics of two neutron sources can be obtained from the experimental results. The advantage of Cf fast fission chamber is that its neutron energy is lower than the DT accelerator, thus the strength of gamma rays in nearby environment is also much lower. It makes the SNR better.

While the advantages of DT accelerator are:

(1) The DT accelerator has much stronger neutron intensity, and makes the count rate much higher and measuring time shorter. So C_{12} can be measured efficiently.

(2) The DT accelerator neutrons have the single energy, and the interference of energy spread is decreased.

(3) When the detectors are located outside the specific tapered region, the interference of source will decrease observably.

(4) The DT accelerator has a switch, and has no radioactivity in manufacture, transportation and assembling. It means the accelerator is much safer for operating personnel.

In further experiment, the DT neutron tube will take the place of DT accelerator as neutron source. The neutron tube has not only all advantages of the accelerator mentioned above, but also more characteristics such as convenient to carry and easy to operate.

6　Conclusion

In this paper, an experimental research conducted on the basis of time-dependent coincidence measurement has been performed with HEU and DU castings with similar mass (about 8–10 kg) and shape (hemispherical shell). The neutron sources are Cf fast fission chamber (7×10^4 s^{-1}) and timing DT accelerator (10^6–10^7 s^{-1}), and the measuring time is 2048 and 1000 s. The experimental results show that the uranium castings with different enrichments can be effectively distinguished by different time-dependent signals (C_{01} and C_{12}) and different expressions (the cross-correlation function and signature parameter), the sensitivity coefficient $(\Delta R/\Delta m)/\bar{R}$ can be as high as 19.3%. This system is suitable for template measurement such as nuclear material transfer, and useful for uranium attribute analysis.

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