RESEARCH ON LEAK MONITORING OF THE PRIMARY CIRCUIT OF A PRESSURIZED WATER REACTOR BASED ON THE ^{13}N COINCIDENCE METHOD

by

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To assess the characteristics of ^{13}N decay, a monitoring method of the primary loop leakage rate based on the γ-γ coincidence method was proposed. In this work, sampling and measurement devices were designed, in which NaI (Tl) crystals were used as coincidence detectors. The Geant4 simulation method was used to study the relationship between the efficiency ε and capacity V change of different devices, and the εV value under the corresponding capacity was obtained. According to the value of εV, the optimal sampling and measurement device was determined, and then the detection efficiency of the device was calibrated experimentally. Taking the 600 000 kW reactor of Qinshan Phase II as the research object, the lower limit of detection was discussed. When the sampling point was located in the fume hood of the control rod drive mechanism, the theoretical lower limit of the coincidence method was 0.756 Lh^{-1}.

Key words: ^{13}N, coincidence method, sampling and measurement device, detection efficiency, lower limit of detection

INTRODUCTION

There are large quantities of equipment, pipelines, and valves in the primary circuit of a reactor. Under the forces of high temperature and high coolant water pressure, it is easy to produce weak links or induce the expansion of original defects and cracks in the boundary, leading to the leakage of coolant water [1, 2]. When the leakage is excessive, the technical specifications require the operation of the reactor to be restricted. Therefore, a complete and effective leakage monitoring system must be included in the reactor design. The primary loop coolant leakage monitoring system mainly monitors radioactivity, such as particulate matter and gaseous matter, in the containment. The judging leakage rate based on the concentration of ^{13}N in the containment is an effective method, and this method has been used in many nuclear power plants [3]. At present, gamma spectroscopy is used to obtain a concentration of ^{13}N, and the lower limit of the monitoring system is approximately 10 Lh^{-1}[4]. The ^{13}N energy spectrum method has a relatively high detection limit, and it is difficult to find a very small leak in the primary loop. Moreover, the energy spectrum method has high requirements for the stability of the entire system. To assess the characteristics of ^{13}N decay, low background, and relatively low system stability requirements of coincidence [5-10], a method for measuring the concentration of ^{13}N in the containment by the coincidence method was proposed.

THEORY

The elastic collisions between a neutron from nuclear fuel fission and a hydrogen nucleus in water produce recoil protons $^{1}H + n \quad n \quad p$

When the produced protons have energy higher than 5.5 MeV, they may interact with ^{16}O and produce ^{13}N via $^{16}O + p \quad ^{13}N + \alpha$

Here, ^{13}N is a positron emitter that has a half-life of 9.96 minutes, the emitted positrons lose their energy when they pass through matter, annihilate with abundant electrons surrounding them, and decay into two gamma-ray protons with equal energies of 0.511 MeV.

Theory of leakage measurement

The concentration of ^{13}N in the primary water is proportional to the power of the reactor. Once the high temperature and high pressure water in the primary circuit leaks into the containment, it will vaporize immediately. Therefore, the leakage rate of the primary circuit water can be obtained by detecting the intensity

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of gamma-rays with an energy of 0.511 MeV in the uniformly mixed gas in the containment.

The concentration of ^{13}N in the containment

If the reactor is operating at full power, the concentration of ^{13}N in the primary circuit coolant is N_1, and the primary circuit has a constant leakage rate V_L, then the number of ^{13}N nuclei per unit volume of air in the containment has a constant increase P_V. The change rate of ^{13}N nuclei N_v per unit volume is as follows

$$\frac{dN_v}{dt} \quad P_V \quad \lambda N_V \tag{1}$$

where λ is the decay constant of ^{13}N in units of h^{-1} and N_V can be derived from eq. (1)

$$N_V \quad \frac{1}{\lambda}(P_V \quad c\mathrm{e}^{\quad \lambda t}) \tag{2}$$

when $t = 0$, the leakage rate is zero, so N_V can be expressed as

$$N_V \quad \frac{P_V}{\lambda}(1 \quad \mathrm{e}^{\quad \lambda t}) \tag{3}$$

In engineering, five cycles are generally used to achieve uniform mixing, if the volume of the containment vessel is V (ignoring the volume of the ventilation pipe) and the ventilation rate is Q_s. then N_V can be expressed as

$$N_V \quad \frac{N_1 V_L}{\lambda V}\mathrm{e}^{\quad 5\lambda V/Q_s}(1 \quad \mathrm{e}^{\quad \lambda t}) \tag{4}$$

when t is the greater than five half-lives, the concentration of ^{13}N is stable. The N_V becomes

$$N_V \quad \frac{N_1 V_L}{\lambda V}\mathrm{e}^{\quad 5\lambda V/Q_s} \tag{5}$$

The number of ^{13}N nuclei in the measurement device

If the time of gas from the sampling point to the measurement device and gas through the device are t_1 and t_T, respectively, the number of ^{13}N nuclei in the device within t_T can be expressed as

$$N_V \quad \frac{N_1 V_L}{\lambda V}\mathrm{e}^{\quad 5\lambda V/Q_s} \tag{6}$$

where $t_1 = (L \quad S)/Q$, $t_T = V_s/Q$, Q is the sampling flow rate, L – the length of the sampling pipe, S – the section of the sampling pipe, and V_S – the capacity of the sampling device.

Transmission coefficient K_2

The transmission coefficient indicates that when the leakage rate is 1 Lh^{-1}, the gamma-ray count rate is measured by the measurement system. If the detection efficiency is ε, the count N_T of the detector system within t_T can be expressed as

$$N_T \quad \varepsilon N_0(1 \quad \mathrm{e}^{\quad \lambda t_T}) \tag{7}$$

by substituting eq. (5) and eq. (6) into eq. (7), the count N_T becomes

$$N_T \quad \frac{N_1 V_L}{\lambda V}\varepsilon Q t_T \mathrm{e}^{\quad \lambda Ls/Q}(1 \quad \mathrm{e}^{\quad \lambda V_S/Q})\mathrm{e}^{\quad 5\lambda V/Q_s} \tag{8}$$

Then, the count rate n_T can be expressed as

$$n_T \quad \frac{N_T}{t_T} \quad \frac{N_1 V_L}{\lambda V}\varepsilon Q \mathrm{e}^{\quad \lambda Ls/Q}(1 \quad \mathrm{e}^{\quad \lambda V_S/Q})\mathrm{e}^{\quad 5\lambda V/Q_s} \tag{9}$$

when the leakage rate of coolant V_L is 1 Lh^{-1}, the transmission coefficient K_2 becomes

$$K_2 \quad \frac{N_1}{3.6\lambda V}\varepsilon Q \mathrm{e}^{\quad \lambda Ls/Q}(1 \quad \mathrm{e}^{\quad \lambda V_S/Q})\mathrm{e}^{\quad 5\lambda V/Q_s} \tag{10}$$

Since the concentration of ^{13}N in the primary circuit water is proportional to the nuclear power, when the reactor is operated at any power P, the transmission coefficient K_2 can be expressed as

$$K_2 \quad \frac{N_1}{3.6\lambda V}\frac{P}{P_o}\varepsilon Q \mathrm{e}^{\quad \lambda Ls/Q}(1 \quad \mathrm{e}^{\quad \lambda V_S/Q})\mathrm{e}^{\quad 5\lambda V/Q_s} \tag{11}$$

Leakage rate of coolant

Once the high temperature and high pressure water in the primary circuit leaks into the containment, it will vaporize immediately and ^{13}N achieve uniform mixing in the air. The emitted positrons annihilate with abundant electrons surrounding them and decay into two gamma-ray protons with equal energies of 0.511 MeV. Therefore, the coincidence method can be used to measure the concentration of ^{13}N in air and deduce the leakage of the primary circuit. The coincidence count rate n, leakage rate V_L of the primary circuit, and transmission coefficient K_2 satisfy the following relationships

$$V_L \quad \frac{n}{K_2} \tag{12}$$

by substituting eq. (11) into eq. (12), the leakage rate becomes

$$V_L \quad \frac{3.6\lambda V n P_0}{N_1 P \varepsilon Q(1 \quad \mathrm{e}^{\quad \lambda V_S/Q})\mathrm{e}^{\quad 5\lambda V/Q_s}\mathrm{e}^{\quad \lambda LS/Q}} \tag{13}$$

when the reactor operates at full power, V_L can be expressed as

$$V_L \quad \frac{3.6\lambda V n}{N_1 \varepsilon Q(1 \quad \mathrm{e}^{\quad \lambda V_S/Q})\mathrm{e}^{\quad 5\lambda V/Q_s}\mathrm{e}^{\quad \lambda LS/Q}} \tag{14}$$

The lower limit of detection

In the presence of background and ^{13}N and with a 95 % confidence level, the minimum detectable count N_D of the measurement system can be expressed as

$$N_D \quad L_C \quad 1.645\sigma_{N_D} \tag{15}$$

when the net count is small and only concerns the statistical fluctuations of the count, N_D becomes [11]

$$N_D \quad 4.653\sqrt{N_b} \tag{16}$$

Converting the minimum detectable count to the minimum detectable activity A_1, some factors need to be considered, such as the decay branch ratio f, sample measurement time t, and absolute detection efficiency ε. Then, the A_1 can be expressed as

$$A_1 \quad \frac{N_D}{f\varepsilon t} \quad \frac{4.653\sqrt{N_b}}{f\varepsilon t} \tag{17}$$

For a primary loop monitoring system, the gas in the sampling container is a gaseous radioactive source. The activity A is related to the concentration of ^{13}N and the capacity V of the sampling device. The A can be expressed as

$$A \quad \lambda NV \quad \lambda KV_L V \tag{18}$$

where K is the constant under certain conditions and V_L – the leakage rate. When A is equal to A_1, the V_L – the minimum detectable leakage rate V_{LD} of the measurement system. Therefore, V_{LD} can be expressed as

$$V_{LD} \quad \frac{4.653\sqrt{\dfrac{n_b}{t}}}{K f \lambda \varepsilon V} \tag{19}$$

Equation (19) shows that reducing the background rate and increasing detection efficiency are effective methods to reduce the detection lower limit of the monitoring system to the primary circuit leakage rate.

MATERIALS AND METHODS

Design of sampling and measurement device

To improve the detection efficiency of the coincidence method, a sampling container was designed as a stainless-steel sealed cylinder with a wall thickness of 2 mm and an inner cavity height of 8 cm. The layout of the detector in the sampling container is shown in fig. 1 (devices are defined as A to J), where the cylinder represents the NaI(Tl) crystal and its size is 3 . Schematic diagrams A to D indicate that five detector crystals are placed vertically, one located in the center and the others located in different positions of the container. The E to G means that the center detector is placed vertically, and the surrounding four detectors are placed horizontally. The H to J means that four detectors are placed horizontally in different positions in the container.

Coincidence efficiency simulation

To study the coincidence efficiency of different sampling and measurement devices, the Geant4 program was applied to simulate the coincidence efficiency of devices A to J as shown in fig. 1. In the simulation, the ^{13}N nuclei were uniformly and randomly generated in the sampling container. The physical processes activated in the detector model of this work are Compton scattering, photoelectric effect for photons, ionization processes, multiple scattering and bremsstrahlung for electrons, ionization processes, multiple scattering, and annihilation for positrons [12]. A useful signal was taken when the energy deposition of gamma rays in the NaI(Tl) crystal exceeded the set threshold, and the coincidence only formed when any two detectors simultaneously generated useful signals. The simulation parameters are shown in tab. 1.

The value of $\sqrt{n_b}\,/\,\varepsilon V$ in the measurement system will change within a range with the change in the lower threshold of the data acquisition device. In this work, two typical thresholds E = 150 keV and E = 450 keV were set, and the corresponding efficiencies are the total coincidence efficiency ε_T and full energy coincidence efficiency ε_F, respectively.

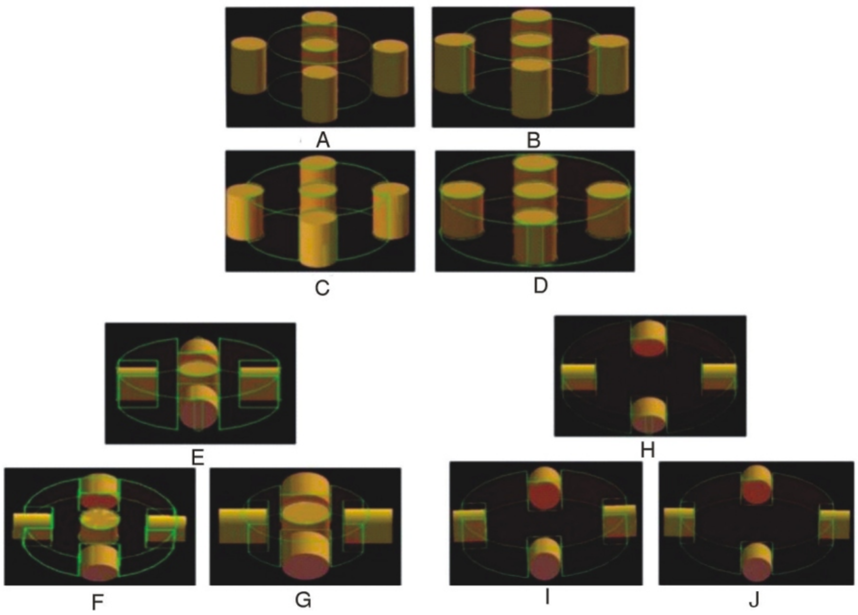

Figure 1. Diagram of detector in the container

Table 1. Simulation parameters of the material

Number	Material	Density [gcm^{-3}]	Element	Content [%]
1	NaI(Tl)	3.67	NaI	99.5
			TI	0.5
2	Al shell	2.702	Al	100
			C	0.12
			Si	0.75
			Mn	0.85
			Cr	19.1
3	Stainless steel	7.9	Ni	9.5
			S	0.2
			P	0.3
			Fe	68.8
			Ti	0.38

Total coincidence efficiency

To ensure that the simulation was most similar to the experimental measurement and to reduce the electronic noise of the measurement system during the experiment, in the simulation, the threshold energy E = 150 keV was used as the basic condition for obtaining coincidence events. If N sampling was carried out and produced n_T coincidence events, then the total coincidence efficiency ε_T can be expressed as

$$\varepsilon_T \quad \frac{n_T}{N} 100 \text{ [\%]} \tag{20}$$

Through simulation calculation, the relationship curve of the total coincidence efficiency with the capacity change is shown in fig. 2.

Full energy coincidence efficiency

In energy spectrum measurements, if the initial energy of the incident gamma-ray is E_0, then the starting point energy of the full energy peak is approximately 0.9 E_0. According to this rule, it is appropriate to select E = 450 keV as the lower threshold for rays with an incident energy of 511 keV.

If N sampling was carried out and produced n_T coincidence events, then the full energy coincidence efficiency ε_F can be expressed as

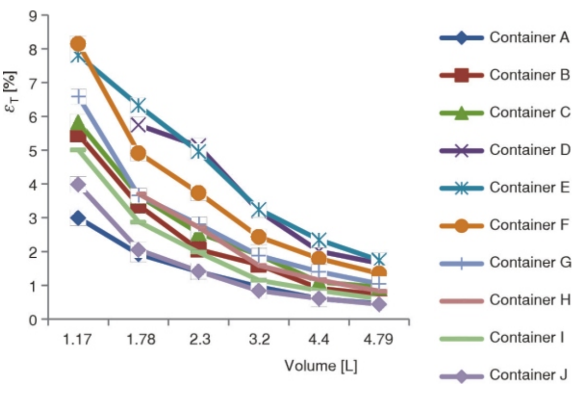

Figure 2. Curve of total coincidence efficiency with volume

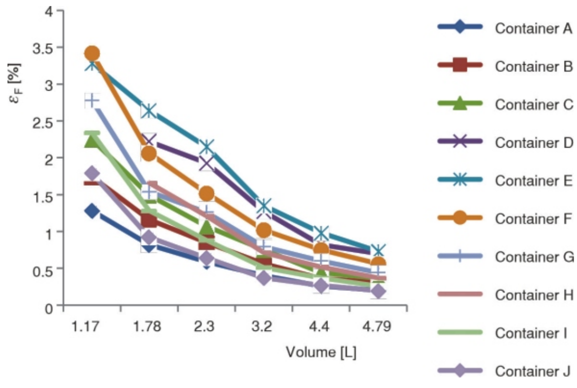

Figure 3. Curve of full energy coincidence efficiency with volume

$$\varepsilon_F \quad \frac{n_F}{N} 100 \text{ [\%]} \tag{21}$$

The relationship curve of the full energy coincidence efficiency with the capacity change is shown in fig. 3.

Figures 2 and 3 show that ε_T and ε_F of D and E are relatively high, and ε_T is much higher than ε_F. When the position of the crystal in the container does not change, the coincidence efficiency declines with the capacity increase of the device. When the capacity is constant, the efficiency gradually increases as the volume of the detector crystal increases in the container.

Comprehensive factor εV

The primary loop leakage monitoring system requires a high detection efficiency of the sampling and measurement device and a larger value of the comprehensive factor εV. When the ^{13}N concentration in the container is certain, the value of εV determines the lower limit of the leakage detection system curve of εV with capacity change, as shown in figs. 4 and 5.

When the capacity V = 2.3 L, the ε_T V of devices D and E are 11.776 and 11.408, respectively, and, the ε_F V are 4.493 and 4.945, respectively. If only the εV value is considered, the geometric structure of device E is better than that of device D, however, in actual use, the shielding and installation space of the device should also be considered. When capacity V = 2.3 L, the radii of D and E are 13.2 cm, and if the thickness of the shielding lead chamber is 15 cm, then the shielding room radii of D and E are approximately 30 cm and 60 cm, respectively. Considering the εV and the installation space comprehensively, the sampling and measurement device should be designed similarly to a D device (all five detectors are placed vertically inside the container) with a capacity of V = 2.3 L.

Coincidence efficiency experimental test

To verify the accuracy of the simulation results and calibrate the coincidence efficiency of the device,

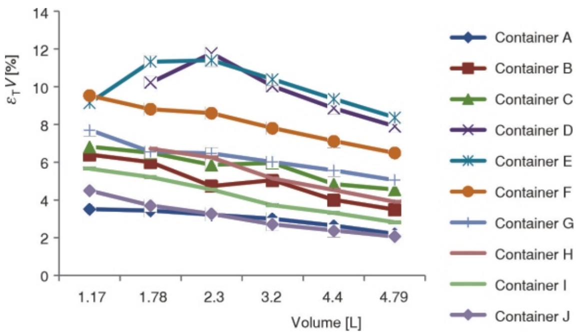

Figure 4. Curve of ε_TV with volume change

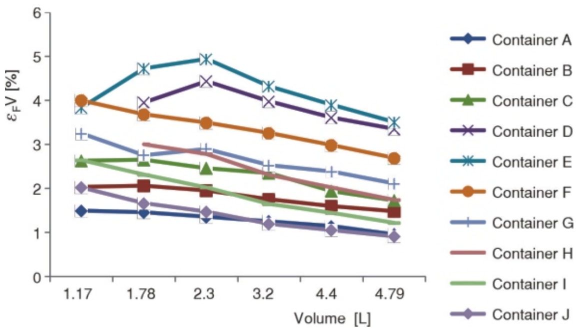

Figure 5. Curve of ε_TV with volume change

a sampling container with a capacity of 2.3 L was produced. All parameters are identical to those in the preliminary design, as discussed. The measurement system consisted of five 3"×3" NaI(Tl) detectors, which were purchased from BeiJing Zhong Guang Detector Co. Ltd CHINA. The ^{13}N-NH_3 gas used in the experiment was produced by a medical cyclotron, and its activity was measured by an activity meter. Under the effect of CCl_4 as a booster, ^{13}N-NH_3 was pushed into a sampling container for the following measurement. The detector and sampling container used in the experiment are shown in fig. 6.

The coincidence experiment system consists of a sampling container, NaI(Tl) detector, amplifier, single-channel pulse amplitude analyzer, coincidence (anti-coincidence) circuit, and intelligent scaler.

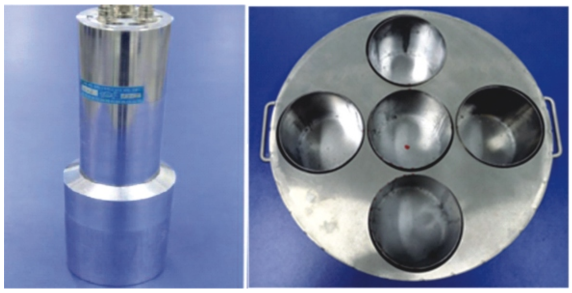

Figure 6. Detector and sampling container used in the experiment

Table 4. Comparison of coincidence efficiency

Parameter	Simulation efficiency [%]	Experimental efficiency [%]	Relative error [%]
Total coincidence efficiency	5.12	5.05	1.37
Full energy coincidence efficiency	1.93	1.89	2.07

To assess the different activities of ^{13}N-NH_3, an efficiency study was carried out, and the results are shown in tab. 2 and tab. 3.

The results of three different activities and their average values are shown in tabs. 2 and 3. For different activities, coincidence efficiency is basically unchanged. A comparison of the simulation efficiency and experimental efficiency of device *D* is shown in tab. 4.

The simulation and experimental results show a slight discrepancy of 3 %, which further validates the Geant4 predictions. The results show that device *D* is the best sampling and measurement device, and it can adapt to the change in the reactor primary loop leakage rate.

Comparison of measurement method

In the monitoring work, due to the influence of environmental background and the detector's background, there is a background count in the coincidence measurement system. The value of $\sqrt{n_b} / \varepsilon V$ varies with the change in the lower threshold of the count system, and the threshold corresponding to the minimum $\sqrt{n_b} / \varepsilon V$ is the optimal operating threshold of the sys-

Table 2. Total coincidence efficiency

Container number	Capacity [L]	Number of decay	Count of true coincidence	Coincidence efficiency [ε/%]	εV	$\bar{\varepsilon}$[%]
D	2.30	816700	41080	5.03	11.569	5.05 ± 0.03
		576982	29137	5.05	11.615	
		408347	20703	5.07	11.661	

Table 3. Full energy coincidence efficiency

Container number	Capacity [L]	Number of decay	Count of true coincidence	Coincidence efficiency [ε/%]	εV	$\bar{\varepsilon}$[%]
D	2.30	847070	16518	1.95	4.485	1.89 ± 0.06
		600064	11341	1.87	4.301	
		423810	7882	1.84	4.232	

Table 5. Results of different threshold

Container number	Capacity [L]	Threshold energy [keV]	Background [cps]*	Coincidence efficiency [%]	$\sqrt{n_b}/\varepsilon V$
D	2.30	150	2.08	5.05	0.124
		450	0.17	1.89	0.095

* counts per second

tem. In this work, the abovementioned experimental system was used to discuss the coincidence background and efficiency when the threshold energy was 150 keV and 450 keV. The results are shown in tab. 5.

When the lower threshold energy of the system is 450 keV, the $\sqrt{n_b}/\varepsilon V$ value is smaller. Therefore, the full energy coincidence mode helps to reduce the lower limit of the measurement system.

RESULTS

The lower limit of detection in the ^{13}N leakage monitoring system is related to the system background, detection efficiency, and reactor power. In this work, the Qinshan Phase II 600,000 kW nuclear power plant is used to calculate the theoretical lower limit of the coincidence method. When the sampling point is located in the hood of the control rod drive mechanism, the calculation parameters are shown in tab. 6.

Substituting the relevant parameters into eq. (10), eq. (17), and eq. (19), the performance parameters of the coincidence monitoring method are obtained and shown in tab. 7.

The error of the theoretical detection lower limit is affected by errors in the background count rate, detection efficiency, and sampling container volume. The errors in the background count rate, detection efficiency, and sampling container volume are approximately 2.4 %, 3.2 %, and 3 %, respectively. Based on the error transmission rule, we know that the error of the theoretical detection limit is approximately 5 %.

Table 6. Calculation parameters

Parameter	Value
Half-life of ^{13}N [min]	9.96
Concentration of ^{13}N in coolant water [13] [cm^{-3}]	4.781 10^6
Volume of the hood [m^3]	2.5 10^2
Cooling ventilation speed [m^{3}h^{-1}]	5.0 10^3
Length of sampling pipe [m]	80
Diameter of sampling pipe [cm]	2
Capacity of sampling container [L]	2.30
Sampling rate [Lmin^{-1}]	35
Coincidence efficiency [%]	1.89
Measurement time [s]	900
Count rate of background [cps]	0.169

Table 7. Performance parameters of the coincidence monitoring method

Transmission coefficient [cps/(Lh^{-1})]	Detectable activity [Bq]	Detection lower limit [Lh^{-1}]
0.084	3.36	0.756

CONCLUSION

Monitoring coolant water leakage is an important strategy to prevent reactor safety accidents, and monitoring the content of ^{13}N in containment is an important method for leakage monitoring. Based on the characteristics of the positron, a method to measure the content of ^{13}N by γ-γ coincidence is proposed. In this work, simulations and experiments were used to optimize the coincidence sampling and measurement device and calibrate the detection efficiency of the coincidence measurement device. Taking the 600 000 kW reactor of Qin Shan Phase II as the research object, when the sampling point is located in the fume hood of the control rod drive mechanism, the theoretical lower detection limit of the coincidence method is 0.756 Lh^{-1}. This result can meet the nuclear power plant's requirement of 1 Lh^{-1}.

AUTHORS' CONTRIBUTIONS

Y. Zhao completed the simulation, and experiment research and wrote the manuscript. G. P. Qu and J. L. Zhou completed experiment research and revised the article.

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ПРАЋЕЊЕ ЦУРЕЊА ПРИМАРНОГ КОЛА РЕАКТОРА СА ВОДОМ ПОД ПРИТИСКОМ ЗАСНОВАНО НА КОИНЦИДЕНТНОЈ ^{13}N МЕТОДИ

Да би се процениле карактеристике распада ^{13}N предложен је метод праћења брзине цурења примарне петље заснован на методи - коинциденције. У овом раду пројектовани су уређаји за узорковање и мерење у којима су кристали NaI(Tl) коришћени као коинцидентни детектори. Методом Geant4 симулације проучаван је однос између ефикасности ε и промене капацитивности V различитих уређаја, а добијена је вредност εV испод нивоа одговарајуће капацитивности. Према вредности εV одређен је оптимални уређај за узорковање и мерење, а затим је експериментално калибрисана ефикасност детекције уређаја. Узимајући као објекат истраживања реактор Qinshan Phase II снаге 600 000 kW, анализирана је доња граница детекције. Када је место узорковања било лоцирано у димној комори погонског механизма контролне шипке, теоријска доња граница методе коинциденције била је 0,756 Lh^{-1}.

Кључне речи: ^{13}Н, коинцидентна метода, уређај за узорковање и мерење, ефикасност детекције, доња граница детекције