Design and Simulation Study of an Online Monitoring Device for Radioactive Noble Gases in Thorium-Based Molten Salt Reactors

Authors: Liao Chuan, Zhou Xuemei, Liao Feng, Du Bingjie, Zhu Lixiao, Liu Wei

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Abstract

Thorium-based molten salt reactors emit radioactive noble gases such as 85Kr and 133Xe (characteristic nuclides) during operation. Accurately monitoring the activity concentration of each characteristic nuclide at the exhaust outlet is of great significance for understanding the operational status of the nuclear reactor and exhaust gas treatment system. The β-γ coincidence method can significantly reduce background and improve measurement precision, meeting monitoring requirements; therefore, a coincidence monitoring device composed of PIPS and HPGe was designed. Based on theoretical values of nuclide activity concentration in molten salt reactor exhaust gas, MCNP (Monte Carlo N Particle Transport Code) was used to design the optimal dimensions of the sampling chamber, study the influence of detectors on β and γ detection efficiency, and analyze the energy spectrum and Minimum Detectable Concentration (MDC) of the monitoring device. The results show that: the optimal dimensions for a cylindrical sampling chamber are a height of 5.3 cm and a radius of 4.25 cm; the monitoring device possesses nuclide identification capability and the MDC for characteristic nuclides is $10^{3-10}5$ mBq/m^3, meeting the design requirements. The results of this paper can provide a theoretical basis for the subsequent fabrication and testing of the monitoring device.

Full Text

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LIAO Chuan^{1,2}, **ZHOU Xuemei**^{1}, **LIAO Feng**^{1}, **DU Bingjie**^{1}, **ZHU Lixiao**^{1}, **LIU Wei**^{1}

^{1} Shanghai Institute of Applied Physics, Chinese Academy of Sciences, Shanghai 201800, China
^{2} University of Chinese Academy of Sciences, Beijing 100049, China

Abstract

Thorium-based molten salt reactors emit radioactive noble gases such as 85Kr and 133Xe during operation. Accurate monitoring of the activity concentrations of these characteristic nuclides at the exhaust outlet is crucial for understanding the operational status of the nuclear reactor and its off-gas treatment system. The β-γ coincidence method can significantly reduce background and improve measurement accuracy, meeting monitoring requirements. Therefore, a coincidence monitoring device composed of HPGe and PIPS was designed. Based on theoretical values of nuclide activity concentrations in molten salt reactor off-gas, MCNP (Monte Carlo N Particle Transport Code) was used to design the optimal sampling chamber dimensions, investigate the detector's influence on β and γ detection efficiencies, and analyze the device's energy spectrum and Minimum Detectable Concentration (MDC). The results show that the optimal cylindrical sampling chamber dimensions are 5.3 cm in height and 4.25 cm in radius. The monitoring device demonstrates nuclide identification capability with characteristic nuclide MDC values of 10^1–10^2 mBq/m^3, indicating excellent performance. These findings provide a theoretical basis for subsequent fabrication and testing of the monitoring device.

Keywords: Thorium molten salt reactor, Radioactive noble gases, β-γ coincidence, MCNP, Minimum detectable concentration

Introduction

As a candidate for fourth-generation advanced nuclear reactors, thorium-based molten salt reactors offer advantages and potential in efficient thorium utilization, high-temperature hydrogen production, waterless cooling, and compact reactor design [1]. During operation, molten salt reactors produce various radioactive gaseous effluents through chain fission reactions, primarily including airborne radioactive aerosols, iodine, and radioactive noble gases. Radioactive aerosols and iodine are almost completely adsorbed by the liquid molten salt, while radioactive noble gases—composed of activation products 37–41Ar and fission products 85–88Kr, 131–135Xe—are mixed with the cover gas. The purged cover gas enters the off-gas treatment system, which employs filters, delay coils, iodine adsorption beds, and activated carbon adsorption beds to treat airborne radioactive materials. After passing through the off-gas treatment system, the gas is discharged into the atmosphere, necessitating monitoring of radioactive nuclides at the exhaust outlet.

Current methods for monitoring radioactive noble gases include total β activity measurement [2], high-resolution HPGe γ spectrometry [3–4], and β-γ coincidence analysis [5–8]. When nuclear reactor operating conditions change, the

activity of released radioactive nuclides varies accordingly. The total β activity method uses detectors to measure the total β activity of various radioactive nuclides to monitor reactor operating status. Currently, TMSR-LF1 (Liquid Fuel Thorium-based Molten Salt Reactor) employs an ionization chamber to monitor total activity at the exhaust outlet, but this method cannot distinguish specific nuclide compositions, limiting understanding of reactor operating conditions. HPGe γ spectrometry leverages the high energy resolution of high-purity germanium detectors to acquire γ-ray energy spectra from various nuclides in the cover gas, identifying and calculating characteristic peaks to obtain activity information. However, radioactive noble gases in molten salt reactor cover gas have high activity [9], and some nuclides have extremely low γ-ray branching ratios (85Kr's γ branching ratio is approximately 0.438%). Certain nuclide spectral information is masked by others, resulting in a large MDC of 10^5 mBq/m^3 and poor detection performance. The β-γ coincidence method exploits the cascade effect during radioactive nuclide decay, using multiple detectors to simultaneously detect β and γ rays, screening for cascade decay coincidence signals to reduce background interference on characteristic nuclide spectra and achieve nuclide discrimination. This method effectively reduces background signal interference with target signals, making it suitable for monitoring radioactive noble gases in exhaust gas, addressing the inability of the current TMSR-LF1 off-gas monitoring system to identify nuclides and achieving detection requirements where characteristic nuclide MDC is lower than current theoretical activity concentration values.

Due to its significant background reduction capability, β-γ coincidence technology is widely applied by nuclear test ban organizations for low-activity xenon detection [10–12]. Based on coincidence technology, various detector configurations have been designed domestically and internationally: NaI+plastic scintillator [13], CsI+plastic scintillator [14], PIPS+CZT [15], plastic scintillator+HPGe [16], and other combinations of β and γ signal detectors. However, these designs focus solely on xenon and its isotopes, with limited research on other radioactive noble gases such as Ar and Kr. Li Yiqiu [17] and Zhou Dongdong [18] from Soochow University, and Feng Yajie [19] from Shanghai Jiao Tong University designed 4π phoswich detectors composed of plastic scintillator and CsI(Tl) scintillator for radioactive gas measurement, using a single photomultiplier tube to collect pulse signals from both scintillators and discriminating through pulse shape analysis. However, these studies were conducted in atmospheric environments with single nuclide objects, low activity concentrations, and low energy resolution requirements, making them unsuitable for the high energy resolution demands of this work.

In summary, to achieve online monitoring of mixed radioactive noble gases in molten salt reactor cover gas while meeting high energy resolution detection requirements, this paper designs a phoswich detector and conducts simulation studies on its relevant parameters using Monte Carlo programs, ultimately obtaining a geometric structure for the monitoring device with excellent performance to meet measurement demands.

1 Monitoring Principle and Structure

Theoretical activity concentration values of nuclides at the molten salt reactor exhaust outlet are shown in Table 1 . Based on activity concentrations and correlation with reactor operating status, 41Ar, 85Kr, 88Kr, and 133Xe were selected as characteristic nuclides for monitoring. The decay types, energies, and branching ratios of these four characteristic nuclides are listed in Table 2 [20].

All four monitored nuclides exhibit cascade effects of varying intensities. The β-γ coincidence method is used to detect coincidence events and calculate nuclide activity concentrations. The mathematical expression for this method is as follows [17]:

In Equation (1), A_i represents the activity concentration of the nuclide to be measured, Bq/m^3; $N_{\beta\gamma}$ is the β-γ coincidence event count; $\epsilon_{\beta\gamma}$ is the β-γ coincidence detection efficiency; and V_{gas} is the volume of the gas to be measured, m^3.

As shown in Table 2, 85Kr has a cascade decay branching ratio of only 0.435%, making it nearly a "pure" β emitter, and its low-energy γ rays can be masked by Compton scattering from γ rays emitted by other nuclides such as 41Ar. Therefore, to accurately identify β and γ rays, the selected detectors must have good anti-Compton scattering performance. A double-layered anti-coincidence PIPS detector with 50 mm diameter and 500 m thickness was chosen for β-ray detection, while a P-type coaxial HPGe detector with annular NaI array for anti-Compton scattering was selected for γ-ray detection.

Based on the above physical analysis, to achieve high detection efficiency for β rays with weak penetration, the PIPS detector was placed inside the sampling chamber. Considering the low-temperature operating requirements of HPGe, the HPGe detector was positioned coaxially with the sampling chamber at a certain distance. To ensure radioactive noble gases remain in the sampling chamber for a sufficient duration, a long inlet and short outlet were designed at the top of the chamber. The longitudinal cross-sectional schematic and specific dimensions of the detector are shown in Figure 1 [Figure 1: see original paper] and Table 3 , respectively.

As shown in Figure 1, the space where the PIPS detector is located is the sampling chamber, whose dimensions are critical to the entire monitoring device as they affect the detection efficiency of β and γ rays emitted by exhaust nuclides. MCNP is a particle transport code developed by Los Alamos National Laboratory with advantages in nuclear reactor neutron transport and detector simulation. Below, based on MCNP6.1.1, we simulate and study the influence of sampling chamber dimensions and the distance between HPGe and the sampling chamber on ray detection to obtain reasonable detector geometric parameters, and analyze the energy spectrum and minimum detectable activity concentration for this configuration to determine whether it meets design requirements.

2.1 Sampling Chamber Optimization Design

As shown in the structural schematic Figure 1, the anti-coincidence PIPS detector is fixed at the top of the sampling chamber with a diameter of 50 mm, so the sampling chamber diameter must exceed 50 mm. Sampling chamber dimensions affect ray detection efficiency and detector count rate. Excessively high count rates may cause pulse pileup in detector signal acquisition, leading to pulse distortion. The mathematical expression is:

$$C = A \times V \times \epsilon \times BR$$

where C, A, V, ϵ, and BR represent count rate, activity concentration (Bq/m^3), sampling chamber volume (m^3), detection efficiency, and radiation branching ratio, respectively. Parameters A and BR are independent of sampling chamber dimensions, while dimension changes significantly affect V and ϵ. To identify optimal dimensions, simulation studies were conducted on sampling chamber radius varying from 3 cm to 5 cm and height from 3 cm to 11 cm, examining effects on ray detection efficiency and composite factor F (defined as the product of sampling chamber volume and ray detection efficiency). To simulate realistic conditions, the mixed field was set proportionally according to exhaust activity values, with air filling the sampling chamber and nuclides uniformly distributed, emitting rays isotropically in 4π geometry. The HPGe-to-chamber distance was temporarily set at 30 cm (referencing a demonstration fast reactor cover gas monitoring system). Detection efficiency and composite factor F versus sampling chamber dimensions are shown in Figure 2 [Figure 2: see original paper].

Figure 2 shows that when sampling chamber radius is constant, β-ray detection efficiency by the PIPS detector decreases with increasing chamber height. This occurs because β particles primarily lose energy through inelastic scattering; as height increases, the proportion of β particles reaching the PIPS detector decreases, reducing detection efficiency. Similarly, when height is constant, the efficiency variation mechanism follows the same principle. The composite factor is jointly determined by volume and detection efficiency, with volume having a greater influence than efficiency. Consequently, the composite factor shows the opposite trend to detection efficiency, increasing with both radius and height. For γ rays, when chamber height is constant, HPGe detection efficiency varies little across different radii because the 30 cm HPGe-to-chamber distance is large compared to chamber radius, resulting in small solid angle changes and minimal γ-ray detection efficiency variation. To achieve high detection efficiency while maintaining a moderate composite factor, sampling chamber dimensions should be radius 3–5 cm and height 4–6 cm under actual detection requirements.

For high β-ray detection efficiency, a PIPS detector with 50 mm diameter was adopted. Considering detector bottom fixation and sampling chamber inlet/outlet holes, the sampling chamber volume was set at 300 ml. Based on the 50 mm PIPS diameter, ten cylindrical sampling chamber diameters from 55

mm to 100 mm were constructed, with corresponding heights calculated using the cylinder volume formula. With the sampling chamber 30 cm from HPGe, the mixed field was set proportionally according to exhaust outlet activity theoretical values, with each source emitting rays according to its decay scheme energy-emission probability. Using the same method as Section 2.1, detection efficiencies for β and γ rays were calculated, with simulation results shown in Figure 3 [Figure 3: see original paper].

Figure 3 shows that at 300 ml volume, as sampling chamber radius increases, both β-ray and γ-ray detection efficiencies increase when radius is less than 4.25 cm. However, when radius exceeds 4.25 cm, β-ray detection efficiency continuously decreases while γ-ray detection efficiency plateaus. This occurs because increasing radius enlarges the effective detection area for both PIPS and HPGe detectors, increasing particle-matter interaction probability and detection efficiency. Conversely, further size increase reduces effective β-detectable particle numbers, but since the sampling chamber radius already exceeds the HPGe crystal radius, no significant change in γ-ray detection efficiency occurs. Therefore, from the perspective of both ray detection efficiencies, the optimal sampling chamber dimensions are radius 4.25 cm and height 5.3 cm.

2.2 γ Detection Efficiency Simulation

The distance between the HPGe detector and sampling chamber affects the solid angle of incident γ rays, thereby influencing detection efficiency. The optimal distance for the thorium-based molten salt reactor radioactive noble gas monitoring system remains unclear. Therefore, using the sampling chamber dimensions from Section 2.2 (r = 4.25 cm, h = 5.3 cm) as a standard, simulations were conducted for HPGe-to-chamber distances from 1 cm to 35 cm to study γ-ray effects, using mixed source terms. Results are shown in Figure 4 [Figure 4: see original paper].

Figure 4 shows that γ-ray detection efficiency continuously decreases as the distance between HPGe and the sampling chamber increases. This is because increased distance leads to greater photon attenuation, scattering effects, and scattering angles, reducing the proportion of photons reaching the HPGe detector. From a pure detection efficiency standpoint, closer distance yields better monitoring device performance. However, high detection efficiency means more photons reaching the HPGe detector, and detectors have dead time that may cause signal distortion. Based on Table 1 and Equation (2), detector count rates at various positions were calculated as shown in Table 4 .

The HPGe detector has a pulse width of 24 s [21–22], with a maximum acceptable count rate of 4.17×10^4 per second. However, the calculated count rates do not account for background, electronic noise, and other factors, so actual HPGe count rates should be higher than values in Table 4. After estimating background and noise impacts, an HPGe-to-chamber distance of 5 cm is optimal, providing relatively high γ-ray detection efficiency with lower count rates,

avoiding pulse pileup from excessive particle numbers.

2.3 Energy Spectrum Simulation

Energy spectrum analysis determines radioactive nuclides and their quantities by measuring characteristic γ-ray energies and intensities in the sampling chamber. Whether the detection system can accurately identify relevant nuclides is an important performance indicator [23]. Therefore, this study conducts energy spectrum simulation analysis for the previously determined optimal dimensions and distance using MCNP6.1's F8 and E8 cards.

In actual energy spectrum measurements, peaks often broaden due to the intrinsic statistical distribution of charge carriers, charge collection efficiency variations, and electronic noise contributions. In MCNP, Gaussian Energy Broadening (GEB) is a special treatment method for simulating peak broadening in detectors, typically expressed as Full Width at Half Maximum (FWHM). The mathematical expression is:

$$\mathrm{FWHM} = a + b\sqrt{E} + cE$$

where E is the incident γ-ray energy, and a, b, and c are energy-related parameters with units of MeV, MeV^(1/2), and MeV^(-1), respectively. The energy broadening coefficients for ORTEC's P-type coaxial HPGe are [24]:

Source information was set using data from Tables 1 and 2, simulating 1×10^9 photons. The normalized energy spectrum is shown in Figure 5 [Figure 5: see original paper].

As shown in Figure 5, due to Compton scattering, 88Kr, 41Ar, and 85Kr form obvious Compton platforms. 88Kr forms single and double escape peaks between 1.5–2.0 MeV due to electron-positron pair production. The simulated energy spectrum of the monitoring device shows characteristic peaks for each nuclide, enabling identification of all four characteristic nuclides listed in Table 2. Full-energy peak areas can also be used for radioactive activity calculations. Figure 5(b) shows detailed information for the 88Kr peak at 0.83 MeV, demonstrating Gaussian broadening due to the GEB card setting.

3 Minimum Detectable Activity Concentration

The minimum detectable activity concentration (MDC) is defined as the lowest activity concentration reliably detectable in a measured sample at a given confidence level. MDC assesses detection system sensitivity in a given measurement environment—the minimum activity concentration a nuclide must achieve for reliable detection. Numerous MDC calculation methods exist, with Currie's method [25] and ISO 11929 standard [26] being most common. Currie's method employs classical statistical theory but ignores system uncertainties. The ISO

11929 standard improves upon Currie's method using Bayesian statistical theory, considering detection system uncertainties and calculating MDC via Monte Carlo methods, but does not account for acquisition/processing time or nuclide decay during processing. This paper adopts the MDC calculation formula from the ARSA β-γ coincidence monitoring device, which considers gas acquisition, processing, and nuclide decay based on uncertainty. The mathematical expression is [27]:

$$\mathrm{MDC} = \frac{\mathrm{DL}}{\epsilon_\beta \epsilon_\gamma BR_\beta BR_\gamma V_{\mathrm{air}}}$$

where DL is the detection limit. Using Currie's method, where DL = $2\sqrt{2}\sigma_0$, Equation (4) becomes:

$$\mathrm{MDC} = \frac{2\sqrt{2}\sigma_0}{\epsilon_\beta \epsilon_\gamma BR_\beta BR_\gamma V_{\mathrm{air}}}$$

where ϵ_β and ϵ_γ are β and γ detection efficiencies; BR_β and BR_γ are respective branching ratios; λ is the specific nuclide decay constant; V_{air} is the gas collection system volume, m^3; t_{collect}, t_{process}, and t_{signal} represent gas collection time, processing time (e.g., decay), and signal processing time, s; and σ_0 is the deviation from background and other influencing counts, expressed as [28]:

$$\sigma_0 = \sqrt{\mathrm{BckCnt}_{\mathrm{total}} + \mathrm{InterferenceCnt}_{\mathrm{total}} + \mathrm{MemoryCnt}_{\mathrm{total}}}$$

where BckCnt, InterferenceCnt, and MemoryCnt represent counts from environmental background, nuclide interactions, and detector memory effects, respectively. Using ARSA detection system timing parameters, and since this device performs real-time online monitoring without decay processing, t_{process} is set to 0. V_{air} is calculated through the ratio of this monitoring device's sampling chamber effective volume to ARSA's effective volume:

$$V_{\mathrm{air}} = V_{\mathrm{eff}} \times \frac{V_{\mathrm{air,ARSA}}}{V_{\mathrm{eff,ARSA}}}$$

where V_{eff}, $V_{\mathrm{eff,ARSA}}$, and $V_{\mathrm{air,ARSA}}$ represent this monitoring device's sampling chamber effective volume, ARSA detection system sampling chamber effective volume, and ARSA system gas collection volume, respectively. Other parameters in Equation (5) such as detection efficiency, branching ratio, and decay constant can be obtained from previous sections. In the σ_0 influencing factors, background and nuclide interactions in complex environments are two key factors, with the latter severely affecting full-energy peak counts in low-energy

regions. Memory effects can be reduced by more than three orders of magnitude by coating the detector inner surface with a thin Al_2O_3 layer [29], so σ_0 can be simplified to:

$$\sigma_0 = \sqrt{\mathrm{BckCnt_{total}} + \mathrm{InterferenceCnt_{total}}}$$

Through simulation analysis of single-nuclide and multi-nuclide mixed radiation fields, changes in Region of Interest (ROI) counts for characteristic nuclides were obtained to determine counting variations caused by nuclide interactions. Specific interaction effects for characteristic nuclides are shown in Table 5 .

In β-ray interaction analysis, all four characteristic nuclides have β-ray detection efficiency of 8.60×10^{-2}, with maximum deviation for 133Xe single-source detection efficiency at 8.53×10^{-2} (0.7% deviation), indicating minimal impact. However, as shown in Table 5, using only γ-ray interaction analysis yields large count values, resulting in high background in minimum detectable activity concentration calculations and poor MDC performance of approximately 10^5 mBq/m^3, demonstrating that HPGe-only analysis of molten salt reactor exhaust nuclides is inadequate under these complex atmospheric conditions. Xiao Simin et al. [30] used PIPS+HPGe with 15 cm low-background lead shielding, analyzing 133Xe β-γ coincidence ROI region [β: 10–350 keV, γ: 77–85 keV] with total background counts of about 50 in 12 hours. After time conversion and using this background value, minimum detectable activity concentrations for the four characteristic nuclides were estimated, with standard deviations calculated using error propagation formulas. Table 6 shows the minimum detection activity concentrations for characteristic nuclides.

The minimum detectable activity concentrations for the four characteristic nuclides range from 10^1 to 10^2 mBq/m^3. 85Kr has a larger MDC value due to its small γ decay fraction and long half-life resulting in a small decay constant. System performance can be improved by increasing detection efficiency or adjusting measurement time to achieve smaller detectable activity concentrations for thorium-based molten salt reactor radioactive noble gas online monitoring systems.

Conclusion

This paper optimized the design of a monitoring device for nuclide identification and activity measurement of major radioactive noble gases at thorium-based molten salt reactor exhaust treatment outlets. The device uses HPGe and PIPS detectors to discriminate and measure β and γ rays online. Based on MCNP simulation results: (1) Without determined sampling chamber dimensions, the monitoring device's ray detection efficiency decreases with increasing chamber height and radius; (2) At 300 ml sampling chamber volume, optimal performance occurs at radius 4.25 cm and height 5.3 cm; (3) Optimal HPGe-to-chamber distance is 5 cm, providing 3.8% γ-ray detection efficiency with count

rate 6.77 × 10^3, below HPGe's maximum acceptable rate; (4) The monitoring device can identify 4 characteristic nuclides with MDC at 10^1–10^2 mBq/m^3, superior to HPGe-only MDC of 10^5 mBq/m^3, with xenon MDC at 3.21 × 10^2 mBq/m^3, demonstrating excellent performance. These simulation results provide theoretical reference for subsequent device fabrication, performance studies, and efficiency correction. Future work will analyze interactions among the four characteristic nuclides in detail and compare with experiments to validate conclusions.

Author Contributions

LIAO Chuan: investigation, simulation, data processing, manuscript writing; ZHOU Xuemei: conceptual design, manuscript revision and finalization; LIAO Feng and DU Bingjie: algorithm design; ZHU Lixiao: figure design; LIU Wei: overall plan review and funding acquisition.

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Note: Figure translations are in progress. See original paper for figures.