Assessment of tritium monitoring for radiation environment around the typical nuclear power plants in China

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Abstract

During the conventional operation, nuclear power plants discharge waste water containing amount of radioactivity, one of main ingredient is the artifcial radionuclide tritium. In this paper, the chemical characteristics and the level of tritium in the environment are studied. The monitoring medium and results of tritium in eight typical nuclear power plants are described. According to the monitoring data, the emission of tritium in heavy water reactor is relatively high, while that in pressurized water reactor is basically at the background level. Tips on topics related to strengthen the monitoring of radiation environment around the nuclear power plants have noted.

Keywords Tritium · Nuclear power plants · Monitoring · Environmental

Introduction

Waste water containing a certain amount of radioactivity were inevitably discharged into the environment accompanied with the operation of nuclear power plants, among which tritium is one of the main radionuclides $[1-3]$ $[1-3]$. Tritium is discharged mainly in the form of gaseous and liquid as radioactive wastes. Tritium is migrate and transform in diferent environments and organisms with the cycle of hydrogen when discharged into the environment. Tritium is absorbed by the body through eating, breathing, skin penetration and other ways to produce radioactive hazards. The activities of tritium released into surface water and other recipients from nuclear power plants are relatively high [\[4](#page-7-2)–[9\]](#page-7-3).

This paper describes the chemical properties of tritium, the sources, the typical nuclear power plants, and the supervision pattern for nuclear power plants in China. The monitoring data of tritium activity in various medium around the nuclear power plants and the supervisory monitoring data of

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Characteristics of tritium

Chemical property

Tritium is the only radioactive isotope of hydrogen, which has a half-life of 12.3 a, with the specific activity of 3.56×10^{14} Bq/g and beta-emitter. The average energy of releasing beta rays is 5.7 keV, and the maximum energy is 18. 6 keV. The low-energy beta rays emitted have a maximum range of about 5 mm in air, 5 μm in water, and 6 μm in skin. The chemical properties of tritium are displacement and permeability. Tritium can replace all chemical reactions involving hydrogen, and replace hydrogen atoms in compounds as C–T covalent bond is more stable than C–H covalent bond. It is easier to replace loosely bonded hydrogen atoms in organic molecules to form organic bonded tritium. Permeability means that tritium will be adsorbed and penetrated the materials like metal, plastic, and rubber structures. Radioactive pollution generate when Tritium were adsorbed on the surface of the structure [\[13](#page-7-6)[–16](#page-7-7)], which can be removed by high temperature heating or water washing,

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but which penetration into structural materials is difficult to be removed. Tritium penetration will cause structural damage, resulting in the performance of structural parts reduced. Chemical forms of tritium include tritiated water (HTO), hydrogen tritiated (HT), methane tritiated (CH3T), organic tritium (OBT) and tissue free tritium (TWFT). Among them, HTO accounts for more than 99%.

Tritium in ambient background

The main source of natural tritium is the interaction between high-energy ions (protons and neutrons) in cosmic rays and nitrogen and oxygen in the upper atmosphere. The yield of tritium in natural is about 7.2×10^{16} Bq/a, the tritium activity in natural is about 1.275×10^{18} Bq after tritium decay equilibrium in the atmosphere. Artifcial tritium is mainly released from nuclear tests, reactors, spent fuel reprocessing, accelerators, heavy water production and treatment facilities. The total activity of tritium released into the atmosphere is about 2.4×10^{20} Bq by nuclear weapon test, especially the test of airburst nuclear weapon, which is much higher than the annual amount of natural tritium. Since the ban on nuclear tests in 1996, tritium released from nuclear weapons tests were gradually decreased and tritium in the atmosphere drop off to the level of background, then the main sources of tritium were release from nuclear reactors and other nuclear facilities. The generation of tritium in nuclear reactor is mainly based on the triple fssion reaction of uranium and plutonium nuclear materials, and the activation reaction of neutrons with deuterium, lithium, boron, and other nuclides in coolant. Taking the pressurized water reactor as an example, the amount of tritium released to the environment through the gaseous phase is about 7.8×10^{12} Bq/ (GWe·a), and the amount released through the liquid phase is about 3.8×10^{13} Bq/(GWe·a) [[17–](#page-7-8)[20\]](#page-7-9). Background survey data show the content of tritium in water bodies of China before the frst commercial nuclear power plant put into operation in the early 1990. The results show that the average concentration of tritium in diferent water is as follows: water vapor (8.96 Bq/L)>Precipitation (5.42 Bq/L)>River/ river $(4.77 Bq/L)$ > Lake/reservoir water $(4.55 Bq/L)$ > Well water (4.04 Bq/L) > Spring water (2.38 Bq/L) > Sea water $(1.34 Bq/L)$ [\[1,](#page-7-0) [2\]](#page-7-10).

Tritium in nuclear accident

In accident, the release of tritium includes instantaneous release and continuous release $[21-23]$ $[21-23]$ $[21-23]$ $[21-23]$. The continuous release usually occurs when there is a weak seal or crack in the tritium process loop, which release gaseous or liquid tritium at a certain leakage rate. The instantaneous release usually occurs when the high-pressure tritium storage container is broken or the tritium process circuit has a large size break. In terms of the form of tritium released in the accident, the reactor nuclear accident is dominated by HTO, with a small amount of HT and CH3T. Taking Qinshan CANDU heavy water reactor with a large amount of tritium production as an example, the gaseous tritium emissions account for about 94% of HTO, about 5% of HT and 1% of CH_3T respectively [[2\]](#page-7-10). The form of tritium in the process loop or pressure vessel depends on the purpose for which it is used, but it is usually tritium gas or HT. The amount of tritium released is usually preponderant due to the greater specifc activity of tritium, which also makes it known as the characteristic radionuclide of some nuclear accidents. For example, the amount of tritium released in the Fukushima nuclear accident is about 3.4×10^{15} Bq.

Monitoring of tritium

Background

By March 2022, China has 54 nuclear power plants in operation (excluding nuclear power plants of Taiwan), and 18 are under construction, with a total installed capacity of 54.63695 million kilowatts, ranking third in the world. Nuclear power plant reactor has a very complex system, with the development of many diferent structures, diferent application of reactors has been developed. The most common classifcation for the reactors is according to the coolant and moderator, such as pressurized water reactor, heavy water reactor, high temperature gas cooled reactor, sodium cooled fast reactor and so on. Figure [1](#page-2-0) and Table [1](#page-2-1) show the typical nuclear power plants in Mainland China. The National Nuclear Safety Administration (NNSA) as an independent national regulator formulated the national radiation environment monitoring program, which clarifes the content of supervised monitoring of the radiation environment around the nuclear power plants. China's nuclear power plant are in safe and stable operation. The fuel element cladding integrity of each unit meets the requirements of the nuclear power plant technical specifcations. The reactor coolant system and the leakage rate of the containment vessel are far below the limits of the technical specifcations, and that safety barrier of the unit is efective.

According to the Law on the Prevention and Control of Radioactive Pollution and the Provisions on Environmental Radiation Protection of Nuclear Power Plants (GB 6249-86), the peripheral radiation environment monitoring of nuclear power plants adopts a dual-track system. The fact is, the nuclear power plant is responsible for routine monitoring and the state environmental authorities are responsible for supervisory monitoring. Ninety percent of China's nuclear power plants are coastal plants, when a nuclear power plant is in operation, the radionuclides

Fig. 1 Schematic diagram of Distribution of the typical nuclear power plants in China

Table 1 Typical technical of nuclear power plants

emitted to the environment. The radionuclides come from fission products of nuclear fuel chain reaction (such as 85 Kr, 133 Xe, 137 Cs, 134 Cs, 131 I, 90 Sr, etc.) and activation products (such as ³H, ⁵⁴Mn, ⁵⁸Co, ⁶⁰Co, ¹¹⁰mAg, ¹²⁴Sb, etc.) [[24,](#page-7-13) [25\]](#page-7-14). In order to protect the environment and the public around nuclear power plants, national environmental authorities set emission limits for radioactive gases and liquids for each nuclear power plant.

Monitoring program

Radiation environment monitoring by NPP

The sampling points for gamma doses rate of the environmental radiation locate within 50 km of nuclear power plants generally, other projects are within a radius of 20–30 km, and the major monitoring range about 10 km around the nuclear power plant. The monitoring of the sea areas focuses on region near the discharge outlet of nuclear power plants. Environmental monitoring focuses on environmental mediators and radionuclides that have the greatest impact on residents who live near nuclear power plants. Environmental radioactivity monitoring consists of atmospheric radioactivity monitoring, terrestrial radioactivity monitoring and Marine radioactivity monitoring.

Monitoring objects include environmental gamma radiation dose rate, aerosol (gamma spectrum, gross activity), air $({}^{3}H, {}^{14}C, {}^{131}I)$, fallout and rain (gamma spectrum, 3 H), surface water (gamma spectrum, 3 H), drinking water (gamma spectrum, 3 H, gross alpha, gross beta), groundwater (gamma spectrum, ${}^{3}H, {}^{90}Sr$), the factory groundwater (γ spectrum, ${}^{3}H$), seawater (γ spectrum, ${}^{3}H$, ${}^{90}Sr$), soil, sediment, terrestrial and aquatic organisms. At the same site carry out the samples monitoring at the intake and discharge port of the nuclear power plants.

Radioactive effluent monitoring by NPP

Composition and number of radionuclides, entering terrestrial and freshwater ecosystems from NPP due to its emissions, are strictly regulated. In the environmental impact assessment stage of nuclear power plant, the management targets of diferent discharge concentrations are set according to the sources of radioactive waste in each nuclear power plants. The volume and concentration were controlled by nuclear power plants when discharge radioactive effluents. Radioactive effluents include gaseous effluents and liquid effluents. During environmental impact assessment phase of the nuclear power plant, diferent waste discharge concentration management targets will be set, depending on the type of the reactors and the power units. Effluent monitoring program generally include outlet sampling monitoring and effluent on-line monitoring. Inert gas (41Ar, 85Kr, 88Kr, 131mXe, 133Xe, 133mXe, 135Xe and so on), iodine, particles, ${}^{14}C$, ${}^{3}H$ ware monitored for gaseous effluents. Liquid effluent monitoring ${}^{14}C$, ${}^{3}H$, other characteristic nuclides $\binom{51}{r}$ Cr, $\binom{54}{r}$ Mn, $\binom{55}{r}$ Fe, $\binom{58}{r}$ Co, $\binom{60}{r}$ Co, $\binom{65}{r}$ Zn, ⁹⁵Nb, ^{110m}Ag, ¹³¹I, ¹³⁴Cs, ¹³⁷Cs, ¹⁰⁶Ru and so on). Critical residents' dose estimation were performed for the impact on the environment of nuclear power plants.

Supervisory monitoring

Radiation ambient air quality monitoring

Radiation can be produced from other sources in addition to nuclear power plants, such as from nuclear fuel cycle facility, uranium mining and metallurgy plant industrial, nondestructive testing machines and medical of X-ray radiation therapy for cancer patients' settings. There are 500 automatic monitoring stations located near the nuclear facilities, cities, important border ports and other sensitive areas. Gas samples were used to analyze gamma doses rate, ³H, 131 I, radon. Aerosols samples were used to analyze gamma nuclides, 90 Sr, 137 Cs, 210 Po and 210 Pb. Seawater samples were collected to analyze 3 H. Fallout samples were used to analyze gamma nuclides, ^{90}Sr , ^{137}Cs .

The developed network of radiation monitoring of ecosystems environment make it possible to register a change in the situation and to identify impact of nuclear facilities operation on the environment and resident around.

Radiation environment monitoring

Several principles will be considered when the provincial environmental protection agencies are formulating supervisory monitoring programs, as follows: (1) Focus on monitoring the changes of radionuclide levels and environmental radiation levels,which around the residents near nuclear facilities and key routes (the waters near the liquid discharge of nuclear power plants); (2) Some monitoring points are the same as those in the background survey and environmental laboratory of nuclear power plants, so as to facilitate the comparison and analysis of monitoring results; (3) Compatible with emergency monitoring, the scheme should be able to provide current situation and real-time data for environmental assessment under accident conditions.

Although the supervision and monitoring programs difer between NPP, but the monitoring items is consistent includes: air absorbed dose rate monitoring as well as radioactive nuclide activity concentration monitoring of air, water, soil, and biological environment medium. Among them, the biological monitoring must also include instructions biological, such as algae, mosses, oyster, etc. These indicator organisms are not necessarily part of the direct food chain, but they have the function of concentrating radionuclides and are a sensitive indicator of environmental pollution. The radiation environment monitoring around the nuclear power plant focuses on the artifcial radionuclides released from the nuclear power plant. The real-time impact and long-term cumulative trend impact of the operation of the nuclear power plant on the surrounding environment are monitored by comparing with the background radiation level. The efective dose of radioactive emitted from the nuclear power plant to the surrounding public is estimated.

Monitoring quality assurance

All monitoring institutions should have metrological certifcation qualifcation and establish corresponding quality system. Tritium were measured by liquid scintillation counter. The personnel, experimental environment, instruments and equipment, quality management and other aspects shall meet the requirements of radiation environment monitoring to ensure quality system efective operation.

The latest effective national or industry standards should be adopted as monitoring methods generally. A value traceability plan should formulate and implement regularly for the instruments and equipment that may have infuence on the accuracy or validity of monitoring results. The sampling equipment and auxiliary measurement equipment should be ensure used within the validity period. Conduct verifcation for instruments and equipment according to the usage condition. Sampling must be carried out according to the sampling points, monitoring items, frequency and time determined by the monitoring scheme. The corresponding technical specifcations shall be met during sampling. The quality control during sample transportation, handover and storage should be ensured.

Data of tritium

Monitoring points

By sorting out the monitoring point data of the monitoring scheme, the map of the number of sampling points of tritium activity from diferent objects in China's typical nuclear power plants is obtained as shown in Fig. [2](#page-4-0).

Figure [2](#page-4-0) shows the monitoring medium of tritium activity cover air, rain, drinking water, surface water, ground water, sea water, and sea organism. According to the different types of reactors and the kilowatts of the nuclear power plants, the numbers of the monitoring points for the tritium activity of diferent nuclear installations are slightly diferent in China.

The number of sampling points from sea water is more than other medium. The total number of monitored tritium activity sites in Qinshan nuclear power plant is 54, which is the largest among all nuclear power plants with 25 of Hongyanhe, 31 of Haiyang, 36 of Tianwan, 32 of Ningde, 29 of Daya Bay, 26 of Fangchenggang and Changjiang Respectively. In order to supervise the early warning of radiation caused to the surrounding environment during the operation of the nuclear power plants more efectively, the number and frequency of monitoring points have been strengthened. The monitoring points and numbers were

optimized to make the monitoring work more representative, scientifc, and feasible.

Distribution of monitoring targets

The emission characteristics of tritium in nuclear power plant is related to the reactor type and operation condition, and HTO is the main component. The formation of HT in nuclear reactors were related to the radiative decomposition of coolant. The construction of $CH₃T$ is related to the oxidation or reduction state of the coolant. The purifcation resin is also one of the sources of tritium-containing organic matter in the liquid effluent of pressurized water reactor. As is shown in Fig. [3](#page-5-0), among all the samples for tritium activity monitoring, the proportion of seawater is the highest of 26.64%. The proportion of groundwater, rainwater and air in the sampling medium was 16.22, 14.67 and 13.51%, respectively. The distinguishing distribution accords with the number of reactor unit and the complexity of reactor type in each nuclear power plant.

Result

This study has showed the assessment monitoring data for tritium activity by comparing supervisory monitoring with nuclear power plant monitoring data (Table [2\)](#page-6-0). The self-monitoring results of Daya Bay NPP and Fangchenggang NPP were exhibited with mean range, other monitoring results were expressed with interval range values.

Fig. 3 Percentage of monitoring targets for tritium activity of China's typical nuclear power plants

The supervisory monitoring results of tritium activity in the air for the Fangchenggang NPP were higher than that of NPP self-monitoring. Other monitoring results and supervisory monitoring data were smaller than the power plant monitoring value. The results of tritium activity monitoring at Qinshan nuclear Power Station were highest, because it has a heavy water reactor, which produces a high amount of tritium. Taking the maximum tritium value of 10,900 Bq/L in the air of Qinshan NPP as an example, the efective value from total tritium to be accumulated will be $E = 104(L)$ (maximum estimated) $\times 10,900$ $(Bq/L) \times 365 = 4.14 \times 10^8$ Bq/a, much lower than the annual discharge limit of 4.5×10^{14} /a in GB6249-2011. The monitoring data of some projects were missing, the reason is that the power plant is disabled to carry out the project or the sample is not representative according to the actual objective situation. All the monitoring data did not exceed the emission standards for safe operation stipulated by the state. Analytical results show negligible radiation hazards to the surrounding populace.

Uncertainty

The main factors contributing to measurement uncertainty include low level radioactive measurement, radioactive decay and background fuctuation, the count statistical deviation" is the fundamental factor of measurement uncertainties. The evaluation of uncertainty mainly considers several aspects as follow: sample count rate uncertainty measured by Liquid scintillation spectrometer $u(N)$, instrument efficiency

scale uncertainty $u(E)$, sample weighing uncertainty $u(m)$ and measurement standard deviation uncertainty *u*(*C*).

$$
u(N) = \frac{\sqrt{\frac{N_x}{t_x} + \frac{N_b}{t_b}}}{N_x - N_b}
$$
\n(1)

in which t_r =time for sample measurement (min); t_b =time for background measurement (min); N_r = Total count rate for sample (min⁻¹); N_b =count rate for background (min⁻¹);

Synthetic uncertainty *u*:

$$
u = \sqrt{u(N)^2 + u(E)^2 + u(m)^2 + u(C)^2}
$$
 (2)

expanded uncertainty *U*:

 $U = ku$ (3)

in which $k=2$; confidence is 95%.

According to the compiling statistical, the uncertainty of the data in the Table [2](#page-6-0) is 8–15%.

Conclusions

The tritium emissions from nuclear power plants are required monitoring by nuclear power plant and supervision monitoring by provincial monitoring agencies. Tritium emissions mainly in air and liquid form from nuclear power plants.

According to the migration characteristics of tritium, the main monitoring media of tritium activity in nuclear power plants are gas, sea water, surface water and groundwater. According to the control value of environmental radiation protection of NPP (GB6249), the control values in water of tritium, carbon—14 and other nuclides in pressurized water reactors is 7.5×10^{13} Bq/a, 1.5×10^{11} Bq/a, 5.0×10^{10} Bq/a respectively; tritium, carbon—14 and the remaining nuclides from heavy water reactors is 3.5×10^{14} Bq/a, 2×10^{11} Bq/a, 2×10^{11} Bq/a respectively. For reactors with a thermal power greater than or less than 3000 MW, the mission limits can appropriate adjust to own situation. Each nuclear facility has designated emission limits of years authorized by NNSA. The radioactive release reported by the research reactors with supervisory monitoring system showed on the annual report of the radiation environment monitoring around the radiation facilities [\[26](#page-7-15)]. The emission of tritium from heavy water reactor is relatively high, while that in pressurized water reactor is basically at the background level.

Due to its chemical uniqueness, the effective dose coeffcients of diferent forms of tritium difer greatly. The values of the background tritium concentration, the concentration of tritium in process facilities and the concentration of tritium in accident scenarios contradict greatly. When the tritium activity monitoring scheme is formulated, specifc research should be carried out according to the specifc requirements of the monitoring purpose, the type of the source, measuring range, measuring precision, and measuring time.

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Declarations

Conflict of interest The authors declare that they have no known competing fnancial interests or personal relationships that could have appeared to infuence the work reported in this paper.

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