Study on Production of Carbon-14 in Successive Fuel Cycles in Pressurized Water Reactor

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Abstract. Carbon 14 (C-14) is one of the most important radionuclides in radioactive gaseous effluents from nuclear power plants. The C-14 emissions of nuclear power plants at home and abroad are statistically analyzed based on the operation data traditionally. In the paper, the generation mechanism of C-14 from nuclear power plants are introduced. Based on a 1000 MWe pressurized water reactor, a three-dimensional core model was established by a Monte Carlo neutron transport program, and the multi-group neutron flux rate in each irradiation region of the successive fuel cycles of the pressurized water reactor was got. Then the production of C-14 of these fuel cycles are obtained. The results show that the C-14 production of the first and second cycles are higher than that of the subsequent cycles, especially for the first cycle, which is 18% higher than the equilibrium cycle.

Keywords: C-14, production, pressurized water reactor.

1 Introduction

As a new energy source, nuclear power is safe, clean and efficient. With the increase of human demand for energy and the prominence of environmental problems, the development of nuclear power has attracted more and more attention from countries around the world. While it has many advantages, the pollution caused to the environment by radioactive materials emitted during the operation of nuclear power plants cannot be ignored. Among the gaseous radioactive effluents from nuclear power plants, Carbon-14 (C-14) is the nuclide that contributes the most to the annual effective dose of the public, accounting for more than 50%[1]. C-14 has a half-life of 5,730 years and has the same physical and chemical properties as ordinary carbon, so it will be widely involved in the biological chain and food chain. C-14 can accumulate in the human body through inhalation and ingestion, and since C-14 undergoes β decay, it is very harmful to human body through irradiation. In the long term operation of nuclear power plants, the emitted C-14 can cause chronic toxic effects on the surrounding environment. Therefore, it is necessary to control the emission limit of C-14.

Internationally, the C-14 emissions from nuclear power plant have been controlled by United States, France and other countries[3]. In 2011, China formulated a new

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standard named Environmental radiation protection regulations for nuclear power plants (GB6249), to ensure that the radiation dose of radioactive substances emitted from nuclear power plants meets the limit requirements and remains at a reasonably low level[2]. The new standard specifies control values for airborne and liquid radioactive effluents, including noble gases, iodine, C-14, tritium and other nuclides, and for the first time sets limits for C-14 emissions. For reactors with 3,000MW thermal power, the annual emission limits for C-14 are 700GBq (gaseous) and 150GBq (liquid), respectively. In addition to the annual emission limit, the total amount of emissions in each quarter shall not exceed 1/2 of the annual emission limit, and the total amount of emissions in each month shall not exceed 1/5 of the annual emission limit. For one site with multiple reactors of the same type, the total annual emissions should be within four times the limit.

2 Statistical Analysis of C-14 Emissions from Nuclear Power Plants at Home and Abroad

According to the monitoring data of effluent emissions from nuclear power plants published by the United States Nuclear Regulatory Commission (NRC) on its website[4] and the European Commission Radioactive Discharges Database (RADD) of nuclear facilities provided by the European Union to its member states[5], C-14 emission data in gaseous and liquid effluents of relevant countries can be obtained, respectively. Based on the calculation of a 1GWh PWR nuclear power unit running at full power for one year (8000h), the annual emissions of C-14 in gaseous effluent are 12~5653.6GBq, with an average value of 246GBq, and the normalized emissions of C-14 in liquid effluent are in the range of 0.012~25.6GBq, with an average value of 9.6GBq. The average values are lower than the emission limits specified in GB6249-2011.

According to the requirements of GB6249-2011, for one site with multiple reactors of the same type, the total annual emissions should be within 4 times of the limit. That is, for one site with four 1GWh nuclear power units, the normalized emission limits of gaseous and liquid effluents are 700GBq and 150GBq per year (8000h), respectively. In fact, a typical nuclear power plant site has 6 units, which lowers the emission limits to 466GBq and 100GBq, and further lowers them if more units are planned. According to foreign emission data, the GB6249-2011 standard has sufficient margins for the current C-14 emission data of nuclear power plants in Europe and the United States.

At present, China's pressurized water reactors (PWR) have a variety of unit types, including M310 improved unit, AP1000, EPR, Hualong One, VVER, etc. According to Ref. [6], the C-14 production of AP1000 unit per year is expected to be 355GBq, of which the gaseous effluent production amount is 319GBq, the C-14 production amount of EPR units per year is expected to be 424.8GBq, and the gaseous effluent generation amount is 382.4GBq, which is significantly higher than the average emission of nuclear power units in Europe and the United States. The balance is small compared with the GB6249-2011 standard, and it is very likely to exceed the monthly and quarterly control values. Therefore, it is necessary to find technical methods to reduce the generation and emission of C-14 on the premise of accurately obtaining the C-14 emissions of each

stage of the unit. In this paper, the method of calculating the C-14 production by the reactor is introduced and the calculation results are analyzed based on a 1000MW pressurized water reactor.

3 Principles and Methods of Calculation

3.1 The Principle of the Generation of C-14

There are three main sources of C-14 in the atmosphere[7]: (1) Continuously produced by the N-14 in the upper atmosphere through nuclear reactions; (2) Atmospheric nuclear tests conducted in the mid-to-late 20th century. Since atmospheric nuclear testing was banned, atmospheric C-14 levels have approached pre-test levels; (3) Generated by various nuclear reactors.

There are 5 elements known to be able to produce carbon-14, and their mechanisms and cross-sections are listed in Table 1, where the first three cross-sections are for thermal neutrons.

Table 1. C-14 production mechanisms and cross-sections

Parent nucleus	Mechanism	Cross-section (barns)
${}^{14}_{7}N$	(n,p)	1.82
${}^{17}_{8}O$	(n,α)	0.24
${}^{13}_{8}O$	(n,γ)	0.9×10^{-3}
${}^{15}_{7}N$	(n,d)	2.5×10^{-7}
${}^{16}_{8}O$	$(n,3He)$	5.0×10^{-8}

In PWR nuclear power plants, C-14 is produced through the first three reactions, the first two being the most dominant pathways. The C-14 produced in PWRs is mainly distributed in two areas: (a) fuel (fuel pellets, fuel cladding, fuel assembly lattice, etc.) and structural materials; (b) Primary circuit coolant.

Research literature [8,9,10] show that the majority of C-14 in fuels is confined to fuels and structural materials and does not have a direct impact on the current environment. As the primary circuit coolant flows through the core area, the N-14 and O-17 atoms contained in the coolant are neutron-activated and have a chance to produce C-14. The content of O-17 in the coolant is the same as the abundance of O-17 in the oxygen element in nature, and the N-14 in the coolant is mainly introduced by nitrogen dissolution and the addition of hydrazine to the downstream of the RCV system.

3.2 Calculation Formula

According to the reaction principle[7], the formula for calculating the rate of C-14 generation in the reactor is as follows:

$$A = \frac{dN}{dt} = N\sigma\phi\left(1 - e^{-\lambda t}\right). \tag{1}$$

Due to the long half-life of C-14, the above equation can be simplified to the following equation during the operational life of the reactor:

$$A = \lambda NV \sum_g \sigma_g \phi_g t; \tag{2}$$

where A is the total activity value of C-14 corresponding to time t, λ is the decay time constant of C-14, N is the number density of the target nucleus per unit volume, V is the irradiated effective volume, σ_g is the nuclear reaction cross-section of different energy groups, ϕ_g is the neutron flux rate of different energy groups within the reactor, and t is the reactor operation time. The microscopic cross-section of ${}^{17}_{8}O(n,\alpha){}^{14}_{6}C$ and ${}^{14}_{7}N(n,p){}^{14}_{6}C$ is shown in Fig.1.

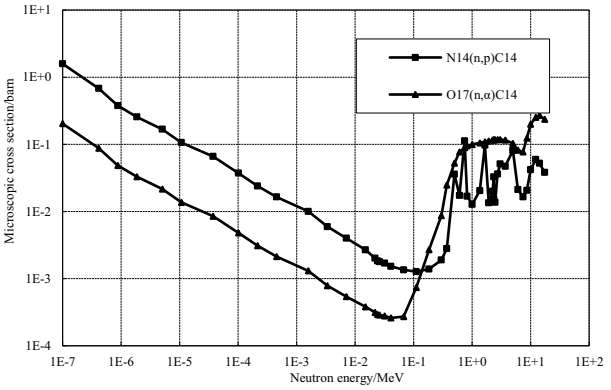

Fig. 1. Microscopic cross section

In addition, the following data are required for the calculation, including the natural abundance of N-14 and O-17, the rated thermal power of the core, the rated electrical power, and the atomic content of N-14 in the coolant.

3.3 Calculating Process

Reactor Core Description.

The reactor core consists of 177 full M5 AFA3G fuel assemblies with an enrichment of 4.45%. Each fuel assembly is arranged in a 17×17 square array. Consisting of 264 fuel rods, 24 guide tubes and 1 instrument tube. In the cold state, the center distance between adjacent fuel assemblies is 21.504cm, the height of the active segment is 365.76cm, and the equivalent diameter of the core is 323cm. The rated thermal power is 3150MWt.

Calculation of Neutron Flux Rate.

In this paper, the multi-group neutron flux rates of the first four cycles (first, second, third and equilibrium cycles) of the pressurized water reactor are calculated. The calculation of neutron flux rate in the irradiated region is based on the Monte Carlo program (JMCT), which can accurately consider the three-dimensional core power

distribution. The neutron flux rates of the core region, upper reflector layer, lower reflector layer, radial reflector layer and descent area are calculated separately. Since parameters such as the power of the core assembly will change with the burn-up, in order to accurately describe the parameters such as core power and energy spectrum, the calculation is expressed by the average weights of several burn-up points. In the calculation, the core neutrons are divided into 53 energy groups.

Calculation of Source Power

The source power sampling probability is calculated by the power distribution and ν/κ value. The three-dimensional core power distribution is divided into N segments in the axial direction, and the radial module adopts the homogeneous power value of the assembly. The ν/κ value is the number of fission neutrons produced per unit of fission energy. The source power can be calculated using the following formula:

$$S = \frac{\nu}{\kappa} \frac{P_t}{177N} P_f \frac{1}{CV}; \tag{3}$$

where C is the unit conversion constant, V is the volume of the assembly of this segment, N is the number of axial segments in the assembly calculation, P_t is the total power of the core, and P_f is the relative power share of this segment.

Determination of Fission Energy Spectrum.

Due to the different burn-up depths of reactor assemblies, the accumulation of nuclides such as Pu-239, Pu-240, Pu-241 and Pu-242 at different locations in the core is also different. Since the isotope of Pu produces more neutrons per unit of energy than the isotope of U, and the energy spectrum of the fission neutrons produced by the isotope of Pu are harder and more penetrating than those produced by the isotope fission of U, the influx of burnup on the neutron source strength and fission neutron spectrum is considered in the calculation of the neutron flux rate of multiple groups in the reactor.

In this calculation, a mixed fission energy spectrum is determined based on the burnup of the assemblies, the fission neutron share of the corresponding major fission nuclide, and the fission energy spectrum of each nuclide.

4 Calculation Results and Analysis

The results of the multi-group neutron flux rate of four cycles of the reactor are shown in Fig. 2 to Fig. 7. Figures 2 to 5 are the neutron flux rates in each irradiation region of the four cycles, respectively. It can be seen that for the same cycle, the neutron flux rate is different in different regions, where the reactor core has the largest neutron flux rate. Fig. 6 and Fig. 7 compare the neutron flux rates of the reactor core region of the four cycles for the fast and thermal energy group, respectively. It can be seen that the thermal neutron flux rates of the first and second cycles are greater than those of the third and fourth cycles, but there is no significant difference in the neutron flux rates of the higher energy group.

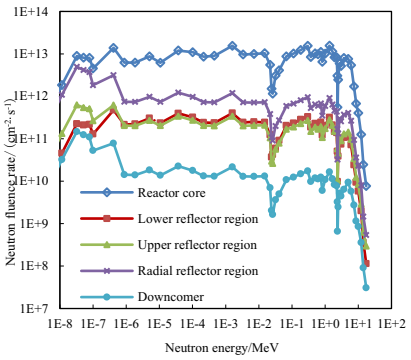

Fig. 2. Neutron flux rate of cycle 1

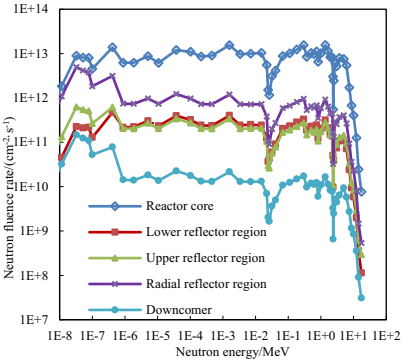

Fig. 3. Neutron flux rate of cycle 2

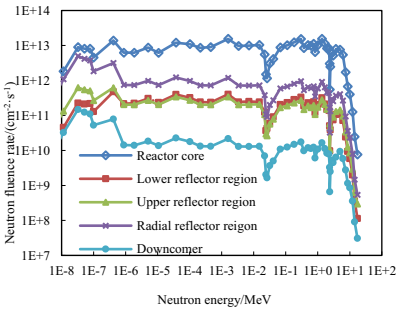

Fig. 4. Neutron flux rate of cycle 3

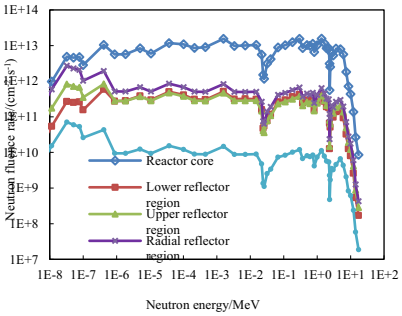

Fig. 5. Neutron flux rate of cycle 4

Table 2 lists the annual C-14 production amount in four cycles of different irradiation areas. According to the data in Table 2, with the increase of the number of cycles, the production gradually decreases and tends to be stable. The first cycle produced the highest amount of C-14, reaching 1.18 times that of the equilibrium cycle, and the second cycle producing 1.07 times the production of C-14 of the equilibrium cycle. The difference in the amount of C-14 produced in different cycles mainly comes from the difference in the amount of reactor zone, which is due to the gradual increase in the average enrichment of the fuel loaded in the core. In the beginning cycles, the enrichment of the fuel is low. To maintain the same power level, the thermal neutron flux rate is higher than that of the equilibrium cycle. The neutron flux rate of the fast group in the core area is almost unchanged but the neutron flux rate of the thermal group gradually decreases, so the activation reaction rate of the coolant region of the core in the first two cycles will be greater than that of the equilibrium cycle, and the amount of C-14 generated in the coolant will be correspondingly greater than that of the equilibrium cycle.

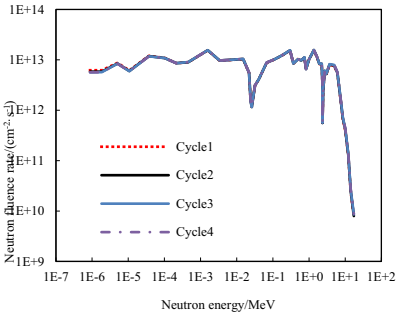

Fig. 6. Neutron flux rate in reactor core area in fast neutron energy region of 4 cycles

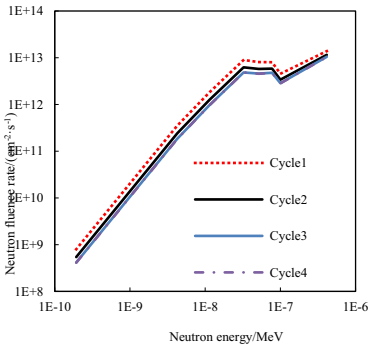

Fig. 7. Neutron flux rate in reactor core area in thermal neutron energy region of 4 cycles

Table 2. Annual production amount of C-14 in each irradiation region of four cycles

Cycle	Reaction	Production amount of C-14(GBq/a)						
		Reactor core	Lower reflector	Upper reflector	Radial reflector	Downcomer	sum	total
1	$^{17}_{8}O(n,\alpha)^{14}_{6}C$	357.3	4.3	14.0	5.9	0.4	382	477
	$^{14}_{7}N(n,p)^{14}_{6}C$	85.5	1.3	5.5	2.8	0.2	95	
2	$^{17}_{8}O(n,\alpha)^{14}_{6}C$	324.7	5.3	18.5	5.3	0.3	354	432
	$^{14}_{7}N(n,p)^{14}_{6}C$	66.0	1.6	7.2	2.5	0.2	78	
3	$^{17}_{8}O(n,\alpha)^{14}_{6}C$	309.1	5.4	18.8	3.4	0.2	337	404
	$^{14}_{7}N(n,p)^{14}_{6}C$	56.4	1.6	7.3	1.6	0.1	67	
4	$^{17}_{8}O(n,\alpha)^{14}_{6}C$	309.0	5.5	18.9	3.5	0.2	337	404
	$^{14}_{7}N(n,p)^{14}_{6}C$	56.1	1.6	7.4	1.6	0.1	67	

5 Conclusions

In this paper, the neutrons in the core are divided into 53 energy groups, the amount of C-14 produced by a 1000MW pressurized water reactor from the first to the fourth (equilibrium) cycle is calculated, and the calculation method is introduced in detail. The results show that the C-14 production of this reactor type decreases with the increase of the number of cycles.

Currently, the application value for C-14 emissions from typical pressurized water reactors is determined based on the value from equilibrium cycle. According to the analysis in this paper, the application value of the reactors should be based on the cycle with the largest emissions, i.e., the first cycle, or around 1.2 times of the emission value from the equilibrium cycle to meet the requirements of the Environmental radiation protection regulations for nuclear power plants (GB6249). Therefore, the reactors in operation should control the C-14 emissions in the first three cycles. In addition, the

design of pressurized water reactors should take the neutron activation effect on the production of C-14 in fuel management of different cycles into consideration.

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