

ESTIMATION OF TRITIUM GENERATION AND DISCHARGE OF THE AP1000 REACTOR BASED ON HISTORICAL DISCHARGE DATA FROM THE U.S. PRESSURIZED WATER REACTORS

by

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During the normal operation of pressurized water reactors, tritium has contributed more than 95 % of the total radioactivity of all the radionuclides discharged into the environment and has been recognized as the key radionuclide in the design and operation of reactors. In the paper, the tritium production of the AP1000 reactor by the Westinghouse approach has been introduced and the diffusion fractions of tritium from fuel rods in the approach have been reviewed according to advances in research on the diffusion of tritium from zirconium alloys. The historical tritium discharge data from the predecessors with very similar core designs of the AP1000 reactor have been collected and sorted. According to the statistical results, the average tritium discharge approximates the tritium generated from neutron activation of boric acid and lithium hydroxide and it shows that the release fraction of tritium through fuel cladding with zirconium alloy may be neglected for the best-estimated condition. The release of tritium from secondary neutron sources has been validated by a comparison of historical tritium discharge from the predecessors with and without secondary sources. In addition, it indicates that local maximum tritium emissions from the U.S. Pressurized Water Reactors are remarkably affected by batch liquid release, especially before or during the overhauls. It will help recognize the tritium generation in the primary loops and optimize the management of radioactive emissions for the utilities of the AP1000 reactor in the future.

Key words: tritium, discharge, AP1000, pressurized water reactor

INTRODUCTION

Tritium is one of the most important radionuclides in the environment and the public during the normal operation of nuclear power plants owing to its long half-life (12.3 years) and predominant contribution to the total activity of all radionuclides discharged into the environment [1]. Tritiated water can enter readily into the human body through drinking, breathing and ingestion. Tritium discharges have been highly focused on safety analysis and environmental impact assessment of nuclear power plants and the public. Therefore, tritium has been recognized as one of the most important radionuclides in the industry [2]. The limits for the liquid and gaseous tritium discharges in pressurized water reactors (PWR) have been strictly authorized to control the impact on the environment and the public [3-5].

The AP1000 reactor is one of the Generation III reactors designed by Westinghouse and it has been approved after review by the U.S. NRC (Nuclear Regulatory Commission). Since AP1000 reactors are im-

ported into China, the attention was paid to the source of tritium because of its high production and discharge. The tritium production in the AP1000 reactor has been theoretically assessed by the standard Westinghouse approach and the origins of tritium include the diffusion of tritium from fuel rods and burnable absorbers, and the neutron activation of boric acid, lithium hydroxide and deuterium in the primary loops. The results of annual tritium production in the AP1000 reactor by the TRICAL computer code have been presented in tab. 1 [6]. The Westinghouse approach for tritium generation was developed in the 1970s and the assumptions used have been only partially validated by two years of operational feedback from the R. E. Ginna nuclear power plant [7]. It was not updated hereafter and does not match the latest operational feedback from a large number of PWR and the research progress on the diffusion of tritium from materials over the past decades.

In the paper, the mechanisms of tritium production by direct sources and indirect sources in pressurized water reactors have been introduced. One method has been developed to validate tritium production based

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Table 1. Results of tritium production in the AP1000 reactor

Source	Tritium production [TBqy ⁻¹]	
	Best-estimated	Conservative
<i>Direct source</i>		
Soluble boron	18.1	18.1
Soluble lithium	4.1	4.1
Deuterium	0.1	0.1
<i>Indirect source</i>		
Ternary fission	8.7	43.7
Burnable absorbers	1.3	6.9
<i>Total</i>	32.5	72.9

on the historical data on tritium discharges from PWR with very similar core designs of the AP1000 reactor. The statistical results will help to determine the source of tritium and help optimize the tritium discharge for the utilities of the AP1000 reactors in the future.

MECHANISMS OF TRITIUM PRODUCTION AND RELEASE

Recognition of sources

The origins of tritium in the primary loops in reactors are different in various types of reactors. All the origins of tritium in the PWR include two parts, the direct sources and the indirect sources. The direct sources designate neutron activation of soluble boric acid, lithium hydroxide and deuterium in the primary loops and all tritium produced by the direct sources will be discharged into the environment. The indirect sources of tritium have the potential contribution to the environment and the quantity of tritium diffused depending on the design of the material of fuel rods, the secondary neutron source rods and control rods. All the reactions for tritium generation in PWR have been summarized in tab. 2 [8, 9].

Mechanism for direct sources

According to the basic theory of neutron activation reactions, the production of tritium in the primary loops can be calculated as follows

$$\frac{d\bar{N}(t)}{dt} = V\bar{N}(t) (\sigma_{\epsilon} \bar{\phi}_{\epsilon} - \lambda) \bar{N}(t) \quad (1)$$

where \bar{N} is the average number of target atoms for tritium in the neutron region, t – the time of operation, V – the total volume of water in the neutron region, σ_{ϵ} – the microscopic cross-section of neutron with energy ϵ , $\bar{\phi}_{\epsilon}$ – the average neutron flux with energy ϵ in the neutron region, and λ – the decay constant of tritium.

The solution of the equation can be expressed as follows

$$\bar{N}(t) = V\bar{N}(t) \sigma_{\epsilon} \bar{\phi}_{\epsilon} (1 - e^{-\lambda t}) \quad (2)$$

Compared with the half-life of tritium, the decay of tritium in several fuel cycles can be negligible. The activity of tritium at time t can be presented as follows

$$\bar{A}(t) = \lambda V \sigma_{\epsilon} \bar{\phi}_{\epsilon} t \quad (3)$$

Therefore, the production rate R of tritium activity can be got as follows

$$R(t) = \frac{d\bar{A}(t)}{dt} = \lambda V \sigma_{\epsilon} \bar{\phi}_{\epsilon} \quad (4)$$

The relation between the neutron flux $\bar{\phi}_{\epsilon}$ and the reactor power P can be present as

$$P = \bar{\phi}_{\epsilon} N_f \sigma_f E_r V_a \quad (5)$$

where N_f is the atomic number density of fissile nuclides (main ²³⁵U in PWR) in the active region, σ_f – the microscopic cross-section of atoms of fissile

Table 2. Nuclear reactions of tritium production in PWR

Region	Nuclear reactions
Fuel*	U/Pu ¹ 0n FP1 FP2 ³ 1H
Antimony-Beryllium (in the secondary source)	⁹ 4Be ¹ 0n (n,α) ⁴ 2He ⁶ 2He ⁶ 2He β ⁶ 3Li ⁶ 3Li ¹ 0n (n,α) ⁴ 2He ³ 1H ⁹ 4Be ¹ 0n (n,T) ⁷ 3Li ³ 1H ⁷ 3Li ¹ 0n (n,nα) ⁴ 2He ¹ 0n ³ 1H
Boric acid (in the primary coolant and control rod)	¹⁰ 5B ¹ 0n (n,2α) ² 2He ³ 1H
	¹⁰ 5B ¹ 0n (n,nα) ⁶ 3Li ¹ 0n ⁴ 2He ⁶ 3Li ¹ 0n (n,α) ⁴ 2He ³ 1H
	¹⁰ 5B ¹ 0n (n,α) ⁷ 3Li ⁴ 2He ⁷ 3Li ¹ 0n (n,nα) ⁴ 2He ¹ 0n ³ 1H
Lithium hydroxide (in the primary coolant)	¹¹ 5B ¹ 0n (n,T) ⁹ 4Be ³ 1H ⁹ 4Be ¹ 0n (n,α) ⁴ 2He ⁶ 2He
	⁶ 2He β ⁶ 3Li ⁰ 1e ⁶ 3Li ¹ 0n (n,α) ⁴ 2He ³ 1H
	⁹ 4Be ¹ 0n (n,T) ⁷ 3Li ³ 1H ⁷ 3Li ¹ 0n (n,nα) ⁴ 2He ¹ 0n ³ 1H
Deuterium (in the primary coolant)	⁶ 3Li ¹ 0n (n,α) ⁴ 2He ³ 1H ⁷ 3Li ¹ 0n (n,nα) ⁴ 2He ¹ 0n ³ 1H
	² 1H ¹ 0n (n,γ) ³ 1H

*Note: FP1 and FP2 are different fission products of fuel, other than tritium

nuclides in the active region, E_r – the average recoverable energy per fission, and V_a – the volume of the active fuel region in the reactor core.

Then the production rate of tritium activity is

$$R = \lambda V \frac{\sigma_\varepsilon P}{\varepsilon N_f \sigma_f E_r V_a} \quad (6)$$

When the volume of the core and the fuel management of the reference reactors are similar to the AP1000 reactor, the distribution of neutron flux in the reactor, soluble boric acid concentration, and lithium hydroxide concentrations are comparable. So it is reasonable to regard the tritium discharges from the reference reactors as the tritium production by the direct sources in the AP1000 reactor.

Mechanism for indirect sources

The fuel rod and secondary neutron source (SNS) are the most important indirect sources for tritium discharge in a pressurized water reactor since a large quantity of tritium has been accumulated in them. The normalized tritium production by ternary fission in fuel rods is estimated as high as the order of magnitude of 500 TBq per GW of electricity per year according to the fission yield of tritium, but the tritium production in the secondary sources cannot be simply evaluated because it depends on the cladding design of the secondary sources and the neutron flux. The amount of tritium released from the indirect sources into the primary coolant can be expressed in the form as follows

$$A_i^{RCP} = \eta_i A_i^{ID} \quad (7)$$

where A_i^{RCP} is the activity of tritium in the primary loops from fuel rods and secondary neutron sources, η_i – the release fraction of tritium through the cladding of fuel rods and secondary neutron sources, A_i^{ID} – the amount of tritium produced in the fuel rods and secondary neutron sources.

The release of burnable absorbers and neutron activation of deuterium in the primary loops in AP1000 can be neglected owing to the tiny contribution to the total tritium in comparison with other sources.

Review of tritium production by the Westinghouse approach

There are two potential problems with the tritium generation in the AP1000 reactor by the Westinghouse approach. One is that the fraction of tritium released from the fuel may be too high, *i.e.* 2 % for the best-estimated condition and 10 % for the conservative condition. The hydrogen diffusion mechanism and experiments have been extensively researched in

the past decades and the release fraction of diffusion through fuel rod with zirconium alloy cladding in PWR has been proved very low due to the outside oxidation layers of cladding, *e. g.*, 0.81 % in the Belgian-3 reactor [10], as low as $3 \cdot 10^{-5}$ by effusion and permeation experiments [11]. The other is that the diffusion of tritium from the secondary neutron source has not been recognized in the AP1000 reactor. Tritium generated by the activation of beryllium in the secondary neutron source can penetrate the stainless-steel cladding readily and then release into the primary coolant in the PWR, which has been verified by the experiment [12], the measurements [13] and the comparison of the tritium discharge before and after the abandoning of the secondary neutron source in many PWR across the world [9]. Although the production of tritium in fuel rods and secondary sources is well understood, there is no consensus on the release fraction of tritium from these indirect sources in the industry. Differences in the assumptions of these release fractions will inevitably bring great uncertainty to the analysis of tritium production. Therefore, the historical data on tritium emissions from PWR with very similar core designs of the AP1000 reactor has been collected and analyzed, and the statistical results have been proposed as the tritium production and emissions in the AP1000 reactor.

STATISTICAL ANALYSIS AND DISCUSSION

Data analysis

In terms of tritium sources, eight reactors from the operating PWR reactors in the U.S. have been recognized as referenceable reactors or predecessors of AP1000 reactors owing to the similarity of the thermal capacity of the reactor, average linear power ($\sim 18.01 \text{ kWm}^{-1}$), fuel cladding (zirconium alloy) and the total active volume of the core [14]. The predecessors have been sorted into two groups, *Group 1* with secondary sources and *Group 2* without secondary sources. The main information on predecessors and the AP1000 reactor has been listed in tab. 3. The materials of the fuel rod cladding in AP1000 and these predecessors are zirconium alloy. Although the geometric configuration of the reactor core is different, the active volumes of the reactor core are comparable and the relative difference is merely around 4.9 %. In addition, the material of the secondary source cladding is double encapsulated stainless steel for AP1000 and these predecessors if used.

In the study, the historical data of annual tritium discharge of the predecessors were collected [15] and have been normalized to full power operation by the records of the annual loading factor of reactors [14]. The annual loading factors of each reactor have been shown in fig. 1.

Table 3. Information on AP1000 and predecessors

Reactor	Thermal capacity [MW]	No. of assemblies	Active height [cm]	Fuel configuration	SNS*	Discharge data (unit*year)
AP1000	3415	157	14	17 17	w/	2019~2021
<i>Group 1</i>						
A	3411	193	12	17 17	w/o	2000~2019
B	3411	193	12	17 17	w/o	2001~2019
C	3438	193	12	17 17	w/o	2001~2011
D	3438	193	12	17 17	w/o	2001~2011
<i>Group 2</i>						
E	3459	193	12	17 17	w/	2001~2019
F	3468	193	12	17 17	w/	2000~2019
G	3455	193	12	17 17	w/	2001~2019
H	3455	193	12	17 17	w/	2001~2019

* w/ and w/o denote whether SNS has been loaded in the core or not

Figure 1. Records of the annual average load factor of predecessors

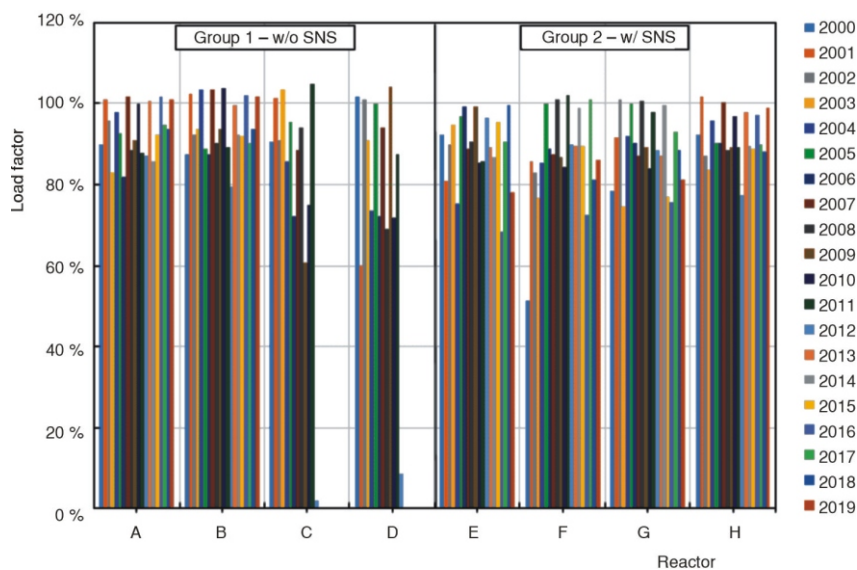
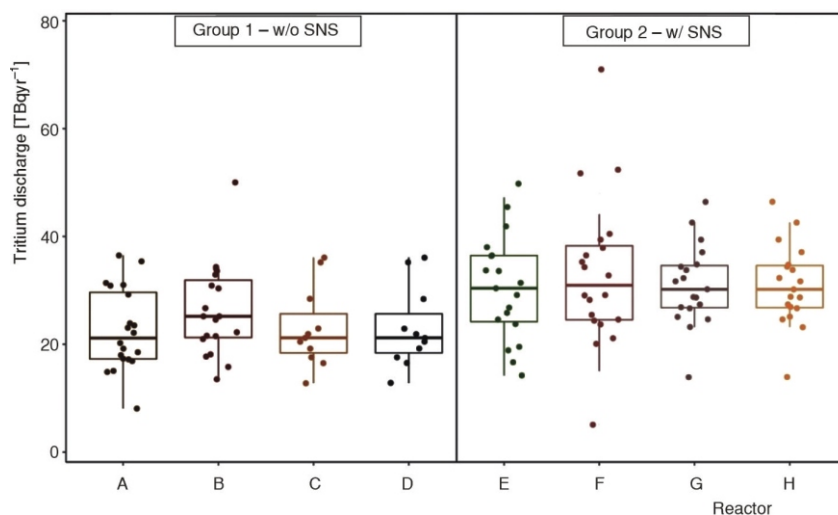


Figure 2. Distribution of annual tritium discharge in predecessors



The statistical results of the historical tritium discharge in these operating reactors have been shown in fig. 2. For the reactors of group 1, the discharged tritium originates from the activation of boric acid and hydroxide lithium, and the potential release from fuel rods. The average tritium discharge in the reactors of *Group 1* is 23.9 TBqyr⁻¹ ($\sigma = 1.2$ TBqyr⁻¹) and just ap-

proximates the tritium production from the direct source (22.3 TBqyr⁻¹) by the Westinghouse TRICAL code [6]. Combining theoretical analysis and the statistics, it shows that the tritium diffused from the fuel rods with zirconium alloy cladding indeed is so low qualitatively and the intact fuel rods of the AP1000 reactor can't be one of the most important origins for the

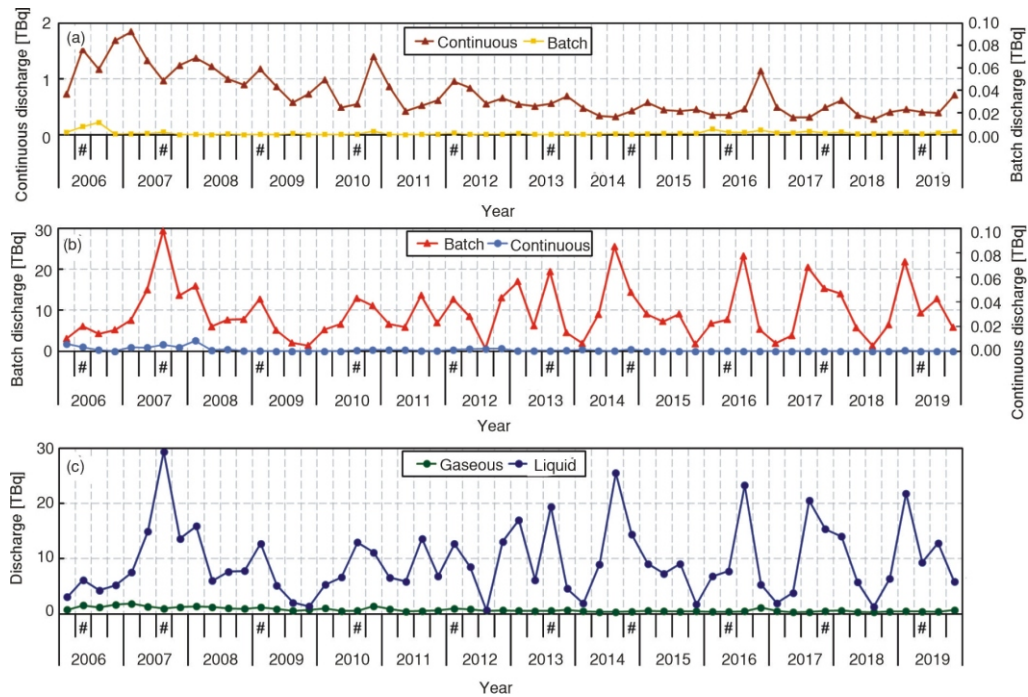


Figure 3. Distribution of quarterly tritium discharges in reactor F: (a) Gaseous discharge, (b) Liquid discharge, and (c) Total discharge. # represents the quarter with the shutdown

tritium discharged into the environment. From the perspective of tritium production, the most difference between the predecessors in two groups is whether the secondary neutron source assemblies have been used in the reactor core. The comparison of average tritium discharges between reactors of group 1 and reactors of group 2 has indicated the release of tritium from secondary sources because the average tritium discharges (31.4 TBqy^{-1}) from the reactors of group 2 are remarkably higher than that from the reactors of group 1. The quantity of tritium released from secondary sources depends on the irradiation conditions, *e. g.*, the local neutron flux and irradiation time of secondary source rods in these reactors. It proves that the secondary neutron source should be the most important indirect origin of the tritium production in the AP1000 reactors and should be considered in the determination of the tritium discharge.

The diffusion behaviors of hydrogen (tritium) through zirconium and stainless steel are much different, and some investigators have shown that the permeation of tritium through zirconium is orders of magnitude lower than in stainless steel [16-18]. Some parts of tritium in the pellet matrix can migrate to the gap and the fuel cladding subsequently by a diffusion mechanism. In comparison to stainless steel, most of these hydrogen atoms can be picked up by the zirconium and then exist in the form of zirconium hydride. It is noted that the solubility of hydrogen in zirconium (more than 150 ppm (parts per million) typically) is much higher than that in stainless steel (less than 5 ppm) [19]. In addition, the oxide layers of zirconium can also prevent the further diffusion of hydrogen atoms to the primary loops [11].

Therefore, a large proportion of tritium in the secondary neutron source can penetrate through the stainless-steel cladding, but most tritium generated in fuel rods can't always penetrate through the zirconium cladding to the primary loops.

Fluctuation of tritium discharges

In operating a PWR, tritium generated in the primary loops in the reactor can be discharged into the environment in two ways, *i. e.* batch release and continuous release, and the respective contribution for tritium release is different. Taking the reactor *F* as an example, the quarterly tritium discharge of units and the quarters in which the overhaul took place has also been presented in fig. 2. It shows that the gaseous tritium discharges do not change remarkably (from 0.24 TBq to 1.85 TBq), but the liquid tritium discharges fluctuated significantly and the local maximum liquid discharge always occurs before or during the period of an overhaul period. The quarterly discharge of liquid tritium with overhaul (10-29.4 TBq) is much higher than that during power operation (around 5 TBq). The average liquid tritium discharge from 2006 to 2019 is 29.3 TBqy^{-1} , but the maximum liquid tritium discharge reaches 71.0 TBqy^{-1} in 2007, which is outside the range of the box plot (13.1 TBqy^{-1} to 40.4 TBqy^{-1}), shown in fig. 3. The remarkable fluctuation of tritium discharge can't be related to the release from fuel or secondary sources since the significant failures of fuel rods and secondary source rods in the cycle didn't happen. It is reasonably speculated that some parts of tritium generated in the primary loops during the operation in 2006

Table 4. Results of the statistic of maximum tritium discharge

Unit	Average [TBqy ⁻¹]			Maximum [TBqy ⁻¹]		
	Gaseous	Liquid	Total	Gaseous	Liquid	Total
<i>Group 1</i>						
A	3.8	18.2	22.0	4.8	32.4	37.1
B	3.0	22.2	25.2	4.8	35.3	39.7
C	2.1	21.5	23.6	2.6	31.8	39.3
D	2.2	22.6	24.8	2.5	35.3	37.0
All	2.8 0.7	21.1 1.7	23.9 1.2	4.8 1.1	35.3 1.6	39.7 1.2
<i>Group 2</i>						
E	3.9	26.9	30.8	5.4	36.5	44.0
F	3.9	29.3	33.2	4.6	37.7	41.9
G	2.6	28.8	31.3	3.6	39.1	41.9
H	2.5	27.6	30.1	3.2	37.1	38.8
All	3.2 0.7	28.1 0.9	31.4 1.1	5.4 0.8	39.1 1.0	44.0 1.8

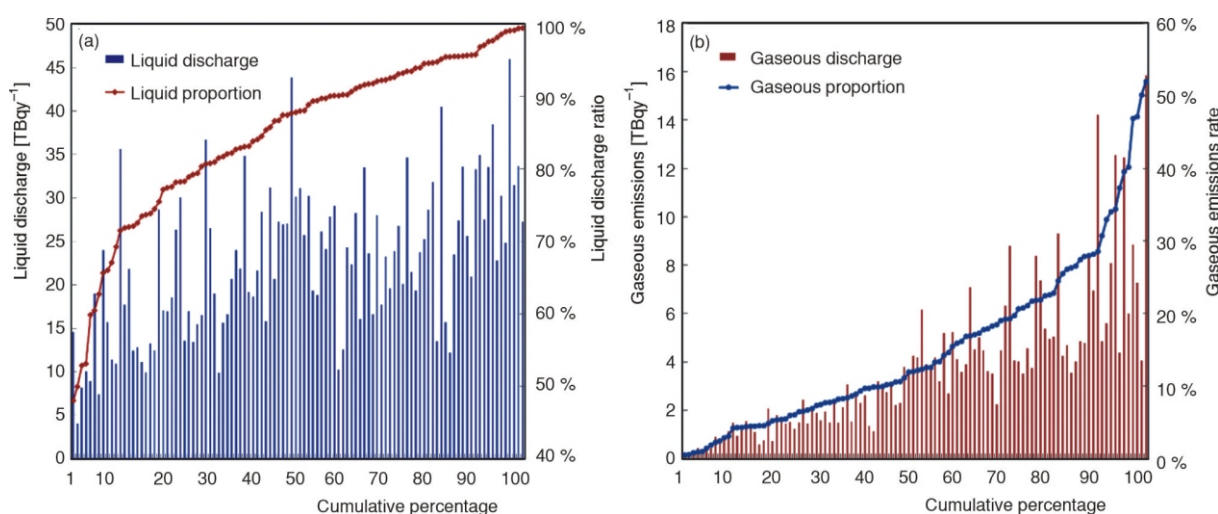


Figure 4. Distribution of the release proportion of tritium in the U.S. PWR: (a) liquid release, and (b) gaseous release

have been batch released before the overhaul in 2007. The liquid batch release occurs commonly during the operation of US PWR, but the batch release can be well controlled if the management of radioactive effluents has been optimized limited, *e. g.*, setting the strict limits of quarterly and monthly tritium discharge.

To reduce the impact of the liquid batch discharge to some certain extent, the rolling average of the historical tritium discharges in a calendar year of the predecessors has been analyzed when a significant outlier is found with a box plot. The statistical results of tritium discharges by rolling average are shown in tab. 4.

Optimized tritium production and discharge

The release proportion of tritium has been analyzed based on the measured data in the predecessors and the cumulative percentage of the proportion of the liquid and gaseous release of tritium has been shown in fig. 4. The proportion of liquid tritium release varies

from 48 % to 99 %, which has been remarkably affected by the liquid batch release. The release proportions with two-year rolling data in these reactors are stabilized at 88 % for liquid release and 12 % for gaseous release respectively.

Based on the statistical analysis of the total tritium discharges, the annual tritium discharges in the predecessors of group 2 are 31.4 TBqy⁻¹ for the best-estimated condition (or the expected condition) and 45.8 TBqy⁻¹ for the conservative condition. The liquid and gaseous tritium discharges have been summarized in tab. 5 and are proposed as the optimized production and discharge of tritium in the AP1000 reactor taking into consideration the similarity between

Table 5. Proposed production and discharge of tritium in the AP1000 reactor

Condition	Tritium [TBqy ⁻¹]		
	Production	Gaseous discharge	Liquid discharge
Best-estimated	31.4	3.8	27.6
Conservative	45.8	5.5	40.3

predecessors of group 2 and the AP1000 reactor. In the comparison of tritium production by theoretical calculation in tab. 1, the results are much more reliable since they are derived from direct statistics of historical discharge data from similar predecessors and do not use assumptions of diffusion proportion from indirect sources. The value of tritium production at the best-estimated condition by the Westinghouse approach has been validated but the part of tritium generated by the fuel rod (ternary fission) actually originates from the secondary neutron sources. In comparison to tritium production at the best-estimated condition, the increments of the conservative production of tritium in AP1000 mainly result from the management of remarkable batch release, not from the higher diffusion from fuel rods. In addition, the conservative value of annual tritium discharge can also be further optimized according to the planning and management of radioactive liquid discharge by future utilities.

CONCLUSION

In the paper, the tritium production of the AP1000 reactor by the Westinghouse approach has been introduced and the assumptions used in the approach have been reviewed and challenged according to advances in research on the diffusion of tritium from indirect sources. The historical tritium discharges in the referenceable PWR have been collected and the statistical analysis shows that the release fraction of tritium from zirconium alloy is very low and fuel rods can't be one of the most important origins for the tritium in the primary loops, but the release of tritium from secondary sources is significant for the AP1000 reactor. In addition, it is found that the significant fluctuations of tritium discharges are remarkably affected by liquid batch release, especially before or during the overhauls in the U.S. PWR. Based on the statistical results, one optimized tritium production and discharge have been proposed based on the historical tritium discharge data from the predecessors with very similar core designs of the AP1000 reactor. It will help recognize the tritium generation in the primary loops and optimize the management of radioactive discharges for the utilities of the AP1000 reactor in the future.

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Ћи ВАНГ

ПРОЦЕНА СТВАРАЊА И ЦУРЕЊА ТРИЦИЈУМА ИЗ РЕАКТОРА АП1000 НА ОСНОВУ ИСТОРИЈЕ ПОДАТАКА У САД О ЦУРЕЊУ ИЗ РЕАКТОРА СА ВОДОМ ПОД ПРИТИСКОМ

Током нормалног рада реактора са водом под притиском, трицијум доприноси више од 95 % укупној радиоактивности свих радионуклида испуштених у животну средину и препознат је као кључни радионуклид у пројектовању и раду реактора. У раду је представљена производња трицијума у реактору АП1000 вестингхаусовског приступа и ревидиране су фракције дифузије трицијума из горивих шипки по овом моделу, у сагласности са напретком истраживања дифузије трицијума из легура цирконијума. Историје података о цурењу трицијума из претходних реактора са веома сличним дизајном језгра као АП1000, прикупљени су и сортирани. Према статистичким резултатима, просечно цурење трицијума приближно одговара трицијуму насталом неутронском активацијом борне киселине и литијум хидроксида и показује да се фракција ослобађања трицијума кроз кошуљице горива са легуром цирконијума може занемарити при најбољој процени стања. Ослобађање трицијума из секундарних неутронских извора потврђено је поређењем историје цурења трицијума из претходних реактора са и без секундарних извора. Поред тога, указано је да на локалне максималне емисије трицијума из америчких реактора са водом под притиском значајно утиче количина испуштања течности, посебно пре или током ремонта. То помаже препознавању стварања трицијума у примарним петљама и оптимизује управљање радиоактивним емисијама реактора АП1000 у будућности.

Кључне речи: трицијум, цурење, АП1000, реактор са водом под притиском
