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Determination of escape rate coefficients of fission products from the defective fuel rod with large defects in PWR

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ABSTRACT

During normal operation, some parts of the fission product in the defective fuel rods can release into the primary loops in PWR and the escape rate coefficients are widely used to assess quantitatively the release behaviors of fission products in the industry. The escape rate coefficients have been standardized and have been validated by some drilling experiments before the 1970s. In the paper, the model to determine the escape rate coefficients of fission products has been established and the typical escape rate coefficients of noble gas and iodine have been deduced based on the measured radiochemical data in one operating PWR. The result shows that the apparent escape rate coefficients vary with the release-to-birth and decay constants for different fission products of the same element. In addition, it is found that the escape rate coefficients from the defective rod with large defects are much higher than the standard escape rate coefficients, i.e., averagely 4.4 times and 1.8 times for noble gas and iodine respectively. The enhanced release of fission products from the severe secondary hydriding of several defective fuel rods in one cycle may lead to the potential risk of the temporary shutdown of the operating reactors.

1. Introduction

During the normal operation of a nuclear power plant, a large number of radioactive fission products can be generated and accumulated in the fuel rods. Once fuel rods fail, a part of the noble gases and the volatile fission products will be released into the primary coolant, resulting in an increase of the radiation dose, radioactive waste and discharges to the environment eventually. Therefore, it is strictly required to maintain the integrity of fuel elements for operational states and to ensure that any damage to fuel elements is kept to a minimum for design basis accidents in the industry [1,2]. However, it is not possible to avoid the failure of a small number of fuel rods in the core during the entire lifetime of a nuclear power plant. In the early stage of the nuclear power plant, 0.1%–1% failure rate of fuel rods occurred in reactors [3]. With the improvement of the fuel design and manufacturing technology in the past decades, the average fuel rod failure rate in pressurized water reactors has decreased and remained as low as 3.7E-5 from 2006 to 2015 across the world [4].

In the early stage of the nuclear power plant, the escape rate coefficient has been introduced to represent the release rate of fission product from the defective rod quantitatively in the irradiation programs. The escape rate coefficient is defined as the fraction of accumulated fission products escaping from one defective fuel rod into the primary loops per unit time. Considering that the defect of fuel that occurred in a UO_2 rod was most likely a small hole from weld porosity or weld contamination in the manufacturing process in the 1950s, some fuel rods with small holes drilled through the cladding were irradiated in the Nuclear Research Reactor and Material Testing Reactor initiated at the Bettis plant. In the irradiation programs, the effect on UO_2 melting of fuel clearance in the rod was developed, and the relationship between the linear heat rate and the fission product release was determined. The escape rate coefficients have been derived from these tests and then used in the early prototype of power reactors, such as the Shippingport Atomic Power Station [5,6]. Later supplemental tests were conducted in the NRX reactor in Canada to determine the effect of rod length on the release of fission gas and iodine from defective fuel rods [7]. The envelopment of the escape rate coefficients has been validated by drilling experiments in Saxton reactor [8]. These experimental results show that the escape rate coefficient will increase exponentially with the linear power of the defective fuel rods. The escape rate coefficients of fission products at linear power of about 280 W/cm (8.5 kW/ft), listed in Table 1, have been standardized by U.S. Atomic Energy Commission [9] and

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Table 1
Standard escape rate coefficients for fission products.

Nuclide	Escape rate coefficients (s^{-1})
Xe, Kr	6.5E-08
I, Br, Rb, Cs	1.3E-08
Mo	2.0E-09
Te	1.0E-09
Sr, Ba	1.0E-11
Y, Zr, Nb, Tc, Ru, La, Ce	1.6E-12

henceforth widely considered to be enough conservative for the commercial pressurized water reactor with ~180 W/cm average linear power. In the design of the Generation II reactor, the operation limits of radioactivity in the primary loops in the radiochemical specification have been determined based on the design basis with the specific fraction of power from failed fuel (1% in the early industry and 0.25% since the early1980s) and the standardized escape rate coefficients of noble gas and iodine. They have been still used in the design of the Generation III reactors, e.g., AP1000 reactor [10], U.S. EPR reactor [11], APR-1400 reactor [12].

Fuel failure in operating reactors has been highly focused in the industry, especially for the utilities of reactors and it is necessary to predict the degree of the fuel failure before the examinations during the shutdown. Some comprehensive models of fuel-failure monitoring have been developed and integrated into some software in past decades and summarized as follows [13,14], DIADEME, PROFIP and the updated OSCAR based on the release-to-birth ratios of noble gas and iodine in the primary coolant and iodine release during a transient) for PWRs by the French Atomic Energy Commission and Framatome [15–17]), MERLIN by Electricite de France [18,19], CADE and FPA based on iodine and noble gas isotopes by Westinghouse for PWR [20,21], CHIRON by the Electrical Research Institute for PWRs and BWRs [22,23], ROTP-CA for the WWERs [24], Visual_DETECT for CANDU reactors [13,25]. In this paper, one model to determine the escape rate coefficients of fission products has been established and the typical escape rate coefficients of noble gas and iodine have been deduced based on the measured radiochemical data in one fuel cycle with large defects in one operating PWR. The envelopment of the standard escape rate coefficients for the commercial pressurized water reactor has been evaluated and explained from the release mechanism of fission products in the study.

2. The theoretical model

The fission products in the primary coolant in PWR originate from the direct release of contaminated actinides and the release of defective fuel rods. The former is governed by the recoil mechanism and is more significant for short-lived fission products. The latter is controlled by the diffusion mechanism and is more significant for the longer-lived fission products. It is necessary to differentiate them in the measured radiochemical data in the primary loops during unit operation.

The contaminated actinides include uranium impurities from zirconium ingot, tramp uranium on the fuel out-surface during manufacture, and the disseminated actinides from the failed rods in the current or previous cycle. For recoil release, it is usually assumed that 50% of the fission products generated by neutron irradiation of contaminated actinides have been released into the primary coolant if angular isotropy for fission fragments has been assumed. The release of fission products from recoil can be described by the following kinetic equation.

$$\frac{dN_i^{TU}}{dt} = \frac{1}{2} \cdot M^{TU} \cdot F^{TU} \cdot Y - (\lambda_i + \beta) \cdot N_i^{TU} \tag{1}$$

Where,

N_i^{TU} = population of fission products generated by the contaminated actinides in the primary loops (atoms).
t = operation time of reactor (sec).
M^{TU} = mass of the contaminated actinides in the outer surface of fuel assemblies (gram).
F^{TU} = fission rate per contaminated actinides for all the assemblies in the core (fission/sec/gram).
Y_i = accumulative yield of fission product i (1/fission).
λ_i = decay constant of fission product i (1/sec).
β = equivalent purification coefficient (1/sec) which is defined as $Q \cdot \eta_i / M$. Here Q is the letdown flowrate, η_i is the decontamination efficiency of purification system for fission product i and M is the mass of the water in the primary loops.

When the release of fission products reaches equilibrium, the following equation can be got.

$$N_i^{TU} = \frac{1}{2} \cdot \frac{M^{TU} \cdot F^{TU} \cdot Y_i}{(\lambda_i + \beta)} \tag{2}$$

The release-to-birth ratio R/B for the recoil mechanism is defined as the ratio of atoms of fission products by recoil released in the primary loops to the total atoms of fission products by fission reaction of all the contaminated actinides. It represents the degree of contamination state in the primary loops and can be expressed as follows.

$$(\mathrm{R/B})_i = \frac{1}{2} \cdot \frac{M^{TU}}{(\lambda_i + \beta)} \tag{3}$$

The release of the fission product from the failed fuel rods is dominated mainly by the diffusion mechanism essentially. The migration can be considered as two continuous steps, i.e., from the fuel matrix to the gap and then to the primary loops, and it is necessary to determine the empirical diffusion coefficients for the two processes based on the relationship between the measured fission products concentrations and post-irradiation examinations [26–29]. However, when the migration of two processes has been regarded as one combined rate process with the apparent escape rate coefficient from the fuel and gap, the overall release of fission products from the fuel pellet and gap to the primary circuit can be described by the first-order kinetic equations. The terms of neutron activation and leakage of the primary loop have been neglected in the model.

$$\frac{dN_i^F}{dt} = F^F \cdot Y_i - (\lambda_i + \varepsilon_i) \cdot N_i^F \tag{4}$$

$$\frac{dN_i^P}{dt} = \varepsilon_i \cdot N_i^F - (\lambda_i + \beta) \cdot N_i^P \tag{5}$$

Where,

N_i^F = population of fission products accumulated in fuel pellets and the gap (atoms).
N_i^P = population of fission products in the primary loop (atoms).
F^F = fission rate corresponding to the local linear power of the failed fuel rod (fission/sec).
Y_i = accumulative yield of fission product i (1/fission).
ε_i = escape rate coefficient of fission products from the fuel rod to the primary loop (1/sec).

It is noted that the apparent escape rate coefficient is time-

dependent and also varies with the defect size, e.g., the following expression of the escape rate coefficient for the fuel failure at initial time t_0 [30].

$$\varepsilon(t) = \varepsilon_0 \bullet \exp[-\xi \bullet (t - t_0)] + \varepsilon_0 \bullet [\omega \bullet (t - t_0)] \tag{6}$$

The first term to the right of Eq. (5) represents the initial burst and subsequent exponential fall-down of fission products through the defect for an initial period and the second term may represent the effect of a linear increase in the defect size of fission products. In Eq. (6), ε_0 is the initial escape rate coefficient at time t_0, ξ *and* ω represents the rate constant of change in the first term and the second term, respectively. However, assuming the quasi-steady state for the release of fission product during subsequent time-spans, it is appropriate that the escape rate coefficients are relatively stabilized during one long period in the paper. When the equilibrium is reached, the atoms inventory in the primary loop and escape rate coefficient can be got as follows.

$$N_i^P = \frac{F^F \cdot Y_i \cdot \varepsilon_i}{(\lambda_i + \varepsilon_i) \cdot (\lambda_i + \beta_i)} \cdot \varepsilon_i \tag{7}$$

$$\varepsilon_i = \lambda_i \cdot \frac{1}{F^F \cdot Y_i \Big/ \left(N_i^P \cdot (\lambda_i + \beta)\right) - 1} \tag{8}$$

The population of measured fission products in the primary loops N_i^M includes the recoil release and the diffusion mechanism when the fuel rods fail. Therefore, it can be expressed as follows.

$$N_i^M = \frac{1}{2} \cdot \frac{M^{TU} \cdot F^{TU} \cdot Y_i}{(\lambda_i + \beta)} + \frac{F^F \cdot Y_i \cdot \varepsilon_i}{(\lambda_i + \varepsilon_i) \cdot (\lambda_i + \beta_i)} \tag{9}$$

The release-to-birth ratio R/B for the diffusion mechanism is defined as the ratio of atoms of fission products released from the defective fuel rod in the primary loops to the total atoms of fission products generated by fission reaction in fuel rod.

$$(\mathrm{R/B})_i = \frac{N_i^P \cdot (\lambda_i + \beta)}{F^F \cdot Y_i} \tag{10}$$

$$\varepsilon_i = \lambda_i \cdot \frac{(\mathrm{R/B})_i}{1 - (\mathrm{R/B})_i} \tag{11}$$

It is noted that and the escape rate coefficient ε_i and the corresponding release-to-birth ratio $(\mathrm{R/B})_i$ in the paper are only used to characterize the release of fission products from the failed fuel rod to the coolant, and so any fission products from tramp uranium and the disseminated actinides from the failed fuel rod should not been considered in Eq (10) and Eq (11).

The relationship between R/B and decay constants of fission products is important to assess the status of fuel integrity and the plot of release-to-birth ratio (R/B) of measured noble gas and iodine isotopes against their decay constant during steady operation is widely used in the industry [31]. When fission occurs in contaminated actinides, fission products are released directly into the coolant with no waiting time for decay. The release-to-birth ratio R/B for fission products from recoil release is independent of nuclide half-life and it exhibits horizontal slopes between R/B versus decay constants. However, if fuel failure happens, a more release of long-lived isotopes than short-lived ones into the primary coolant owing to the delay during the diffusion process from the fuel pellets, the pellet–cladding gap and the primary, and therefore it exhibits a sloping straight line between R/B versus decay constants. The relationship between R/B and decay constants for contaminated

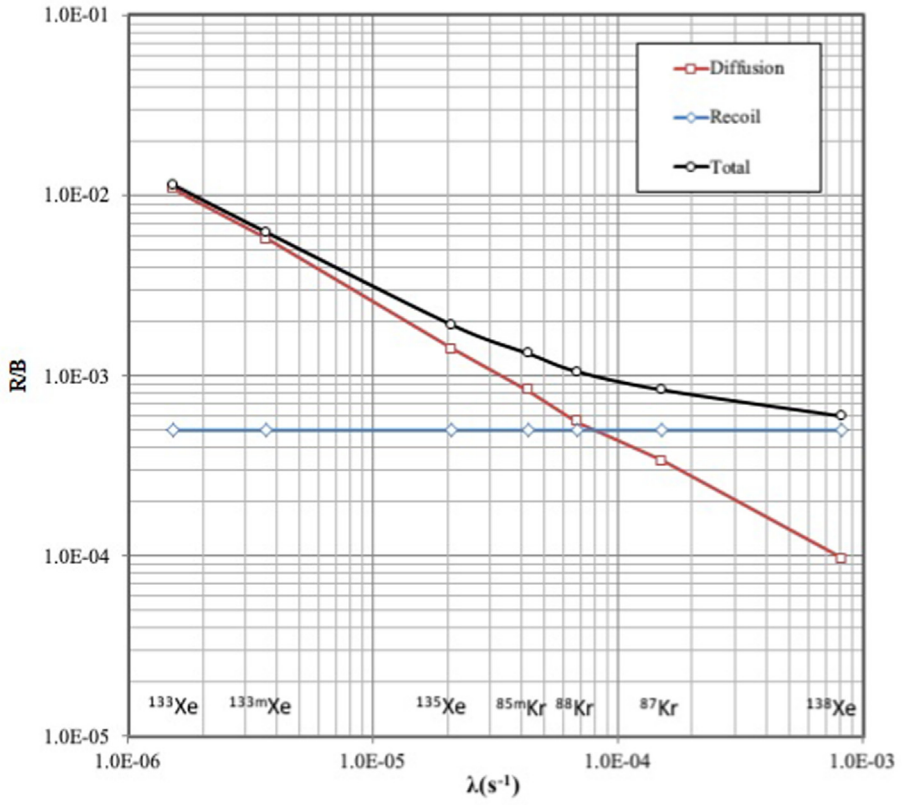

Fig. 1. Typical curves of R/B versus λ of noble gas in the primary loops.

Table 2
The typical characteristics of the 1000 MWe PWR.

Parameter	Value
Reactor	
Core Power	2895 MW
Electricity output	1089 MWth
RCP pressure	15.5 MPa
RCP flow rate	3 × 23790
Total water mass in RCP	~180 ton
Assemblies in core	157
Fuel and fuel management	
Fuel type	AFA 3G AA
Fuel cladding	M5 alloy
Active height	365.76 cm
Average linear power	186 W/cm
Assembly configuration	17 × 17
Rods per assembly	264
^{235}U enrichment	4.2 wt%
Nominal letdown flow rate	13.6 t/h
Operation	
Nominal letdown flow rate	13.6 t/h
Inlet coolant temperature	293.0 °C
Cycle length	~300 days
Decontamination efficiency of resin	≥90% for iodine and 0% for noble gas

actinides and failed fuel rods has been illustrated in Fig. 1. In the study, the least-squares regression has been used to determine the respective contribution of fission products from contaminated actinides and failed fuel rods. The extent of defect sizes of failed fuel rods can be identified according to the slope of log(R/B) and logλ, e.g., −0.5 for large failure and −1.0 for medium or small failure for noble gas [28].

3. Results

When the failure of a lot of fuel rods happens in one fuel cycle, some averaged characteristics of all defective rods can be evaluated and it is difficult to evaluate the status of each failed fuel rod. However, it provides the opportunity to assess the applicability of the standard escape rate coefficients used in the industry if there is only one defective fuel rod in most cycles. One typical case with fuel failure of large defects in one 1000 MWe PWR unit has been presented to help the understanding of the release of fission products from the defective fuel rod. The typical reactor design and fuel design of the PWR and operating parameters of the cycle have been listed in Table 2.

The fission products in the primary loops have been monitored routinely by grab sampling and HPGe spectrometer twice one week, and the frequency of sampling will increase to once a day in case of occurrences of severe fuel failure. The volume activities of the measured noble gas and iodine in the primary coolant have been present in Fig. 2. In the first week of operation, the activities of noble gas and iodine in the coolant increased rapidly by at least five hundred times, which indicates the fuel failure happened.

With the unit operation, the release of fission products in the fuel gap of the failed fuel rod gradually decreased, but the amount of disseminated actinides in the primary coolant gradually increased. The released fission product from the defective fuel rod and fission product from the disseminated actinides in the primary loops have been discriminated by the least-squares method and the results of regressions between log(R/B) and logλ for the released fission product of noble gas from the defective fuel rod have been shown in Fig. 3. The range of slope (−0.62, −0.85) indicates the defect size is very large. The trend of increasing ^{134}I activity indicates the dissemination of fissile material from the defective rod in the cycle and large defects exist. At the end of the cycle, about 80% of fission products with short lives ^{134}I and ^{138}Xe in the primary coolant are generated by the fission of disseminated actinides in the out-surface of fuel rods but not directly released from the failed rod.

The maximum escape rate coefficients and the averaged escape rate coefficients weighted by the operation periods have been evaluated and presented in Table 3. It shows that the apparent escape rate coefficients vary for different fission products of the same element. The result reveals that the escape rate coefficients from the defective rod with large defects are much higher than the standard escape rate coefficients widely used in the industry, i.e., averagely 4.4 times and 1.8 times for noble gas and iodine respectively. According to Eq. (11), the escape rate coefficient of the fission product depends on the product of its decay constant and R/B from the defective fuel rods. In the comparison of fission products with shorter half-life, the decay constant of fission products with longer half-life is less, but the R/B value is higher since more atoms will decay during the migration process from a defective rod and fewer atoms can exist in the primary loops. Therefore, there aren't any fixed decreasing or increasing trends between the escape rate coefficients and decay constants for the fission products. In addition, it is noted that the weighted average escape rate coefficient of I-134 is slightly less than that of other radio-iodine and it may not actually characterize the overall diffusivity of iodine from the fuel matrix and the gap since most of I-134 in the primary loops is mainly generated by recoil mechanism. The uncertainty of the results is slightly less than 10% which results from the measurement of the volume activities of noble gas and iodine in the primary loops in the cycle.

The in-mast sipping examination and the visual inspection

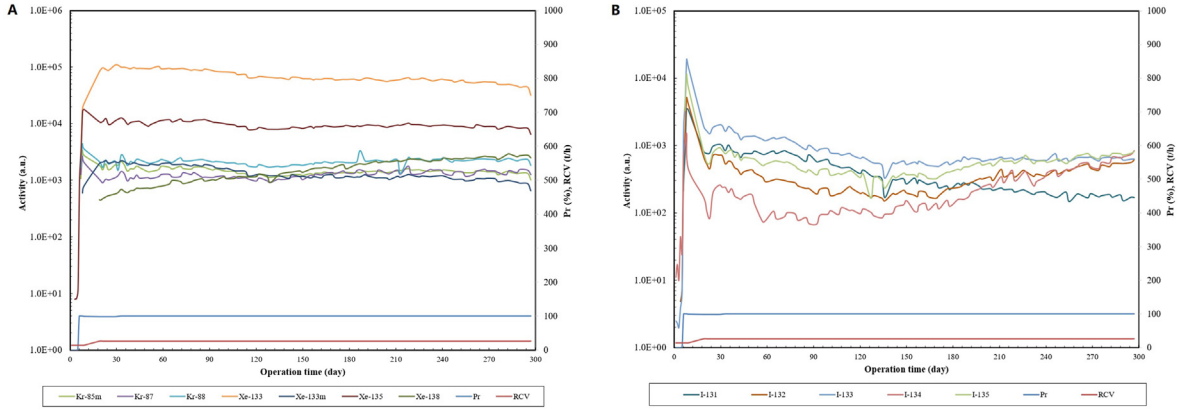

Fig. 2. The trend of measured fission product in the primary loops: (**A**) noble gas, and (**B**) iodine.

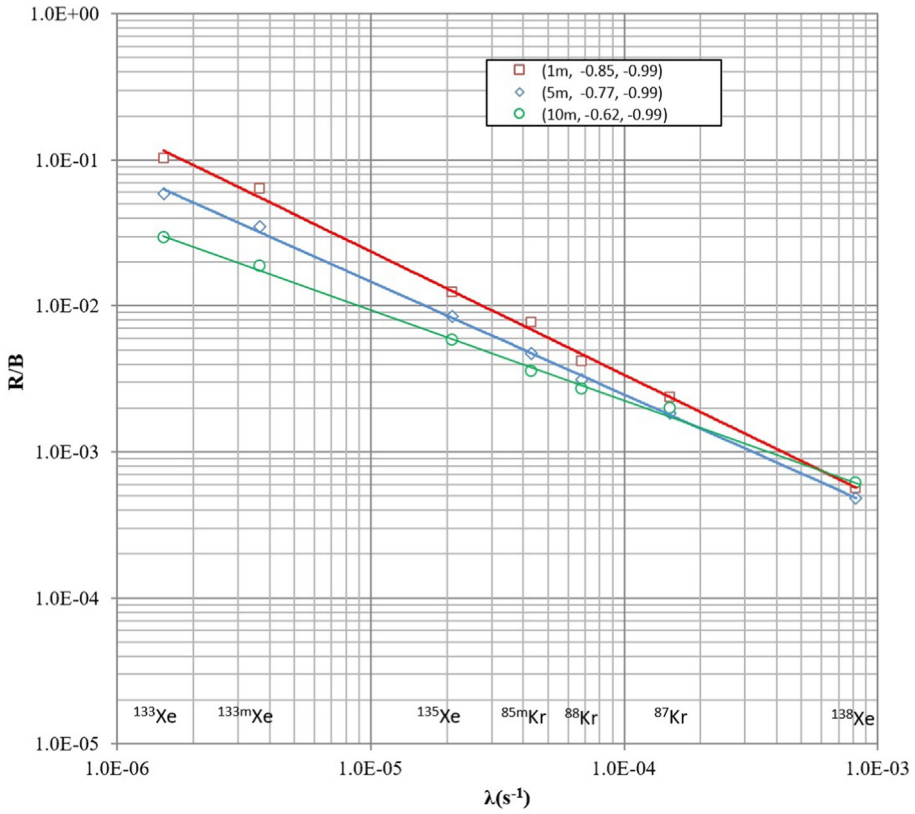

Fig. 3. The regression relation of R/B and λ for noble gases from failed fuel rods. Natation (x, y, z): x is the operation time (month) after fuel failure, y and z are the slopes and R^2 for regressions between log(R/B) and logλ respectively.

show one fuel assembly irradiated in the core for the first time is suspected and one failed fuel rod with linear power around 242 W/cm has been recognized by ultrasonic test (UT). After storage in the spent fuel pool for six years, the failed fuel rod was transported to the hot cell and post-irradiation examinations (PIE) confirmed that six defects exist on the failed fuel rod and the maximum defect size reached the order of centimeters in diameter. In addition, the results of gamma scanning of the defective rod have confirmed that several grams of disseminated fissile material has been lost in the failed rod due to secondary degradation.

4. Discussions

The enhanced release of noble gas and iodine in the cycle is accompanied by the occurrences of secondary degradation of the defective fuel rod. Secondary degradation often can induce different modes of failure, e.g., guillotine rupture and axial splitting of fuel cladding, and the dissemination of the fuel matrix in the defective fuel rods. The unusual grain growth and unexpected restructuring under oxidizing environment are remarkably accompanied by the stoichiometric change of the pellet into UO_{2+x} when secondary degradation occurs severely. The fission gas release of UO_{2+x} fuel is enhanced owing to the increased diffusivity due to stoichiometry excess [32] and the higher temperature caused by degraded fuel thermal conductivity [33,34]. Consequently, the escape rate coefficient of fission products, especially for noble gases, will increase from the defective fuel rods correspondingly and lead to the high volume activities of fission products in the primary loops during operation.

According to the operation experience across the world, the secondary degradation of defective fuel rods is inevitable during long-term operation of reactors, e.g., occurrences in French PWR [35], UK PWR [36], Germany PWR [37] and Chinese PWR [38]. However, the enhanced release of fission products due to secondary degradation has not been paid enough attention in the design of source terms in most pressurized water reactors. During the operation of some commercial PWRs, the fission products volume activities of noble gas and iodine in the primary loops have been strictly limited to some extent to avoid the deterioration of fuel failure, e.g., the limit of total noble gas of 500 MBq/t within 48 h and 1000 MBq/t within 8 h for shutdown in the many PWR fleets [39]. However, the credible model and the measured radiochemical data in the paper indicate the conservatism of the standard escape rate

Table 3
Comparison of the calculated and the standard escape rate coefficients.

Nuclide	Half-life	Escape rate coefficients(s^{-1})			B/A	C/A
		Standard(A)	Maximum(B)	Averaged(C)		
Kr-85 m	4.48 h		4.4E-07	3.3E-07	6.8	5.1
Kr-87	76.4 m		5.1E-07	4.2E-07	7.9	6.5
Kr-88	2.83 h		3.1E-07	2.4E-07	4.8	3.7
Xe-133	5.25 d	6.5E-08	2.0E-07	1.3E-07	3.0	1.9
Xe-133m	2.20 d		3.1E-07	2.0E-07	4.8	3.1
Xe-135	9.14 h		2.7E-07	2.1E-07	4.1	3.2
Xe-138	14.1 m		5.6E-07	4.6E-07	8.7	7.0
Noble gas	–		5.6E-07	2.8E-07	8.7	4.4
I-131	8.03 d		5.3E-08	2.1E-08	4.1	1.6
I-132	2.30 h		1.1E-07	3.8E-08	8.3	2.9
I-133	20.8 h	1.3E-08	5.9E-08	2.4E-08	4.5	1.8
I-134	52.5 m		2.9E-08	1.1E-08	2.2	0.9
I-135	6.58 h		4.6E-08	2.3E-08	3.6	1.8
Iodine	–		1.1E-07	2.4E-07	8.3	1.8

coefficients has been challenged for the defective rod with large defects during the operation of the reactor. The severe secondary degradation of several defective fuel rods in one cycle, much less than the design basis with 0.25% fuel failure assumption, may result in high radiochemical levels in the primary loops and thus lead to the temporary shutdown of the reactors, which is more serious than expected and will cause the reduction of reactor economics.

In the current manufacture of fuel assemblies in commercial pressurized water reactors, the fuel assemblies must be cleaned several times and the tramp uranium at the outer surface of rods has decreased significantly. The transuranic radionuclides in radioactive solid wastes are generated predominately from the subsequent release of disseminated actinides due to secondary degradation. Therefore, it may be necessary to introduce the effect of fuel failure with large defects in the design of modern nuclear power plants.

5. Summaries

In the study, the evolution of the standard escape rate coefficient in PWR has been introduced and one model has been developed to determine the escape rate coefficients of fission products from defective fuel rods. The escape rate coefficients of noble gas and iodine have been determined based on the measured radiochemical data in one cycle with one defective fuel rod with large defects in one PWR and the results are much higher than the standard escape rate coefficients used for several decades in the industry. The enhanced release of fission products results from the severe secondary hydriding and the potential risk of the temporary shutdown of the operating reactors is proposed. It is necessary to review the relationship between the escape rate coefficients of noble gas and iodine and the characteristics of fuel defects systematically and it will help to identify the status of fuel failure according to the measured radiochemical data more effectively and accurately. The assessment of escape rate coefficients will comprehensively be in the future.

Additional requirements

No additional requirements.

Author contributions

The author listed has made a direct and intellectual contribution to the work and approved it for publication.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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