Analysis of Calculation Model for Primary Coolant Fission Products

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Abstract

The sources of radioactive contamination of the primary coolant by fission products when the unit is operating at the rated power are as follows: defect fuel elements with gas leakiness and substantial damages, surface contamination of the outer surfaces of fuel claddings, superficial contamination of structural materials of fuel assemblies. Initially in the reactor operation (if there are no manufacturing defects in fuel elements), the contamination of the coolant by fission products is determined by the release into the reactor coolant circuit of fission fragments of Uranium-235 (due to their kinetic energy) that is present on the outer surfaces of fuel elements as contamination in their manufacturing. During normal operation of the reactor, the integrity of cladding may fail due to various processes of corrosion fatigue type. These processes result in, first of all, micro-fissures and then in large defects in the claddings, which is accompanied by an increase in the release of fission products from fuel elements into the primary coolant.

Keywords: reactor; calculation model; corrosion; primary coolant; fission products.

1. Introduction

The main sources of active substances at the nuclear power plant (NPP) are the fission products of Uranium-235 as a result of the neutron irradiation of the nuclear fuel, and activation of structural materials, corrosion products and chemical additives in primary coolant by neutron sources of the reactor plant [1]-[2].

In order to confine the spread of radioactive gases and aerosols and to prevent their release into the environment, the principle of defence-in-depth is implemented, which is based on using a system of barriers. These barriers are as follows [1]:

- fuel matrix;
- cladding;
- reactor coolant system boundary;
- containments;
- biological shielding.

The NPP has additional process systems and equipment in order to prevent spreading of radioactive materials at the NPP premises and into the environment.

Under normal operating conditions, all of the provided barriers and process systems are functional. According to operational procedure requirements, loss of integrity of barriers or failure of the protection system power operation of the NPP is prohibited.

For the entire range of normal operating conditions of the NPP (power operation, shutdown, maintenance, refuelling etc.), the operating limits and safe operation limits are established, which characterise the condition of the systems (components) and the NPP as a whole (reactor output, reactor coolant pressure, coolant temperature during

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reactor operation). Introduction of such limits enables us to guarantee the integrity of the barriers (the claddings, first of all), and thereby to prevent considerable release of fission products from the fuel into the reactor coolant system and further - to the plant areas where the main process equipment is located [1]-[4].

Little portion of all of the radioactive fission and activation products that are produced during the unit power operation can be released into the plant waste, and the more so into the environment. The portion of the fission products accumulated in the fuel that remains in the spent fuel assemblies is at least 99.9 % of their entire amount being produced.

2. Goal of research

The goal of research is assessment of radioactive waste production during normal operation of power unit in compliance with technical assignment for NPP for fuel element leakage corresponding to the operation limit.

3. Calculation model for primary coolant fission products

The diffusion model that describes migration and leak of radioactive fission products from Uranium dioxide and from the cladding under various irradiation conditions was used as the model for substantiating the contamination of the coolant with fission products.

Within this model, the following mechanisms for release of fission products from Uranium dioxide into the fuel-clad gap of the cladding are used [1]-[2]:

- release due to kinetic energy of fragments in fission of Uranium-235 (release due to recoil);
- release due to "knocking-out" fission products from the fuel surface by the fission fragment missiles (the knock-out-effect);
- release of fission products from the zone of structural changes (the high-temperature area of the fuel).

If the reactor core contains leaky fuel elements, the activity of some fission products in the primary coolant will be observed, especially after reactor power reduction. This increase in activity (the spike-effect) is caused by an additional release of gaseous and volatile fission products that are accumulated in the fuel-clad gap of leaky fuel elements. The most intensive release of fission products is observed in the course of decreasing 100 % power of the reactor (by more than 30 %) [1], [5].

The flow diagram illustrating the migration of fission products from sintered Uranium dioxide-based fuel rods with zirconium cladding is shown in Fig.1.
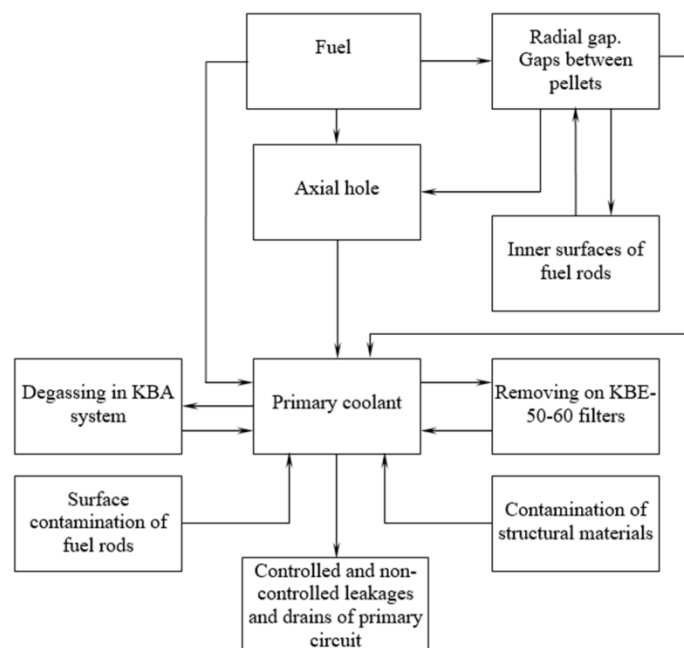

Fig.1. Fission products migration.

Maximum design activity of long living radionuclides during power operation of the plant is reached by the end of the third year of fuel assembly (FA) operation due to the maximum burn-up.

4. Design activity of fission products in the primary coolant

The activity of fission products in the primary coolant was calculated at the end of the three-year fuel cycle using the REL water cooled water moderated power reactor (WWER) computer code [6]. It has been conservatively assumed that the quantitative values of failed fuel elements are constant over the entire period of nuclear fuel cycle. The calculated values of the activity of fission products in the primary coolant are given in Table 1 [1], [6].

Table 1. Activity of fission products in the primary coolant.

Nuclide	Half-life, s	Volumetric activity of the primary coolant, Bq/m^3			
		Operational limit for damage to fuel elements		Safe operational limit	
		Rated operating conditions	Power reduction mode (Spike-effect)	Rated operating conditions	Power reduction mode (Spike-effect)
Br-84	$1.91 \cdot 10^3$	$4.73 \cdot 10^8$	$1.12 \cdot 10^{10}$	$2.02 \cdot 10^9$	$5.51 \cdot 10^{10}$
Kr-85m	$1.61 \cdot 10^4$	$1.78 \cdot 10^9$	$8.94 \cdot 10^9$	$8.65 \cdot 10^9$	$4.40 \cdot 10^{10}$
Kr-85	$3.38 \cdot 10^8$	$7.58 \cdot 10^5$	$2.12 \cdot 10^6$	$3.74 \cdot 10^6$	$1.05 \cdot 10^7$
Br-87	$5.55 \cdot 10^1$	$4.45 \cdot 10^8$	$2.70 \cdot 10^{10}$	$1.40 \cdot 10^9$	$1.32 \cdot 10^{11}$
Kr-87	$4.58 \cdot 10^3$	$2.70 \cdot 10^9$	$2.82 \cdot 10^{10}$	$1.26 \cdot 10^{10}$	$1.39 \cdot 10^{11}$
Kr-88	$1.02 \cdot 10^4$	$5.57 \cdot 10^9$	$3.52 \cdot 10^{10}$	$2.67 \cdot 10^{10}$	$1.74 \cdot 10^{11}$
Cs-137	$9.52 \cdot 10^8$	$1.88 \cdot 10^9$	$1.99 \cdot 10^9$	$9.22 \cdot 10^9$	$9.80 \cdot 10^9$
Xe-138	$8.48 \cdot 10^2$	$1.62 \cdot 10^9$	$3.38 \cdot 10^{10}$	$6.11 \cdot 10^9$	$1.65 \cdot 10^{11}$
Cs-138	$2.00 \cdot 10^3$	$2.46 \cdot 10^9$	$3.56 \cdot 10^{10}$	$1.04 \cdot 10^{10}$	$1.74 \cdot 10^{11}$
Ba-139	$5.09 \cdot 10^3$	$1.02 \cdot 10^9$	$1.02 \cdot 10^9$	$3.60 \cdot 10^9$	$3.60 \cdot 10^9$
Ba-140	$1.10 \cdot 10^6$	$1.32 \cdot 10^7$	$1.32 \cdot 10^7$	$3.65 \cdot 10^7$	$3.65 \cdot 10^7$
La-140	$1.45 \cdot 10^5$	$1.46 \cdot 10^6$	$1.46 \cdot 10^6$	$4.00 \cdot 10^6$	$4.00 \cdot 10^6$
Ce-141	$2.81 \cdot 10^6$	$1.85 \cdot 10^6$	$1.85 \cdot 10^6$	$3.95 \cdot 10^6$	$3.95 \cdot 10^6$
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Zr-95	$5.53 \cdot 10^6$	$1.67 \cdot 10^6$	$1.67 \cdot 10^6$	$2.32 \cdot 10^6$	$2.32 \cdot 10^6$
Nb-95	$3.02 \cdot 10^6$	$1.13 \cdot 10^4$	$1.13 \cdot 10^4$	$2.17 \cdot 10^4$	$2.17 \cdot 10^4$
Zr-97	$6.12 \cdot 10^4$	$1.11 \cdot 10^8$	$1.11 \cdot 10^8$	$1.52 \cdot 10^8$	$1.52 \cdot 10^8$
Nb-97	$4.33 \cdot 10^3$	$9.37 \cdot 10^7$	$9.37 \cdot 10^7$	$1.29 \cdot 10^8$	$1.29 \cdot 10^8$
Inert radio-active gases	-	$1.97 \cdot 10^{10}$	$2.23 \cdot 10^{11}$	$9.03 \cdot 10^{10}$	$1.10 \cdot 10^{12}$
Total Iodines	-	$1.39 \cdot 10^{10}$	$2.15 \cdot 10^{11}$	$6.53 \cdot 10^{10}$	$1.06 \cdot 10^{12}$
Total	-	$5.18 \cdot 10^{10}$	$6.54 \cdot 10^{11}$	$2.31 \cdot 10^{11}$	$3.20 \cdot 10^{12}$

With account for the data provided one can see that during spike effect the primary coolant activity increases only by gaseous (Kr, Xe) and volatile (Br, Rb, I) radionuclides. Activity of the other considered radionuclides stays at the same level [1], [2].

The design values of fission products in the coolant are maintained at a sufficiently low level by means of blowing down the coolant continuously through the ion-exchange filters of system KBE 50-60 and by subsequent degassing (degassing ratio – 0.17 1/h).

During cool down of reactor prior to scheduled refuelling, the fission product activity in the primary coolant lowers from Spike-effect for operating fuel rod damage limit to total activity of $4 \cdot 10^9$ Bq/m^3. During refuelling, no inert radioactive gases are present in the primary coolant since at reactor cool down its full degassing occurs.

Any scheduled shutdowns (for refuelling, maintenance) do not cause increase in releases to the environment together with excess water.

5. Activity of activation products

Activated products in the primary coolant and on the surfaces are produced as a result of the following [1-2]:

- activation and corrosion of materials of various components of reactor internals and the core;
- corrosion of materials outside the reactor core, with the corroded particles being activated when passing through the reactor core with the primary coolant.

In addition, sodium and potassium admixtures dissolved in the coolant are activated when passing through the core. The equilibrium activities of Na-24 and K-42 in the coolant (averaged over the fuel cycle) are $1.85 \cdot 10^9$ Bq/m^3 and $8.14 \cdot 10^9$ Bq/m^3, respectively. Based on operating experience at the reference plants with WWER and pressure water reactor (PWR), the balanced activity of C-14 in the coolant is $1.5 \cdot 10^1$ Bq/m^3 to $1.0 \cdot 10^2$ Bq/m^3. Since C-14 and Na-24 (in compliance with NRB-96 permissible nominal specific activity C-1 in water is $2.2 \cdot 10^6$ Bq/m^3) have low activities and are not of any danger to the personnel and the public, these nuclides are not considered in the further analysis.

6. Model for calculating the activity of corrosion products

The formation of corrosion products in the components of the reactor coolant system was analyzed using the COTRAN computer code [1], which allows us to assess the specific activity of corrosion products in the coolant, the activity on the filters in the system for purifying the reactor coolant, and the surface corrosion activity at different parts of the reactor coolant system equipment during steady-state operation of the power unit:

- the hot pipeline;
- the hot steam generator (SG) collector;
- the SG tubing;
- the cold pipeline;
- the cold SG collector;
- the reactor coolant pump (RCP);
- the protecting tube system.

The algorithm of the COTRAN code is developed using a physico-chemical model that takes into account the behaviour of soluble and dispersed phases of corrosion products in the primary circuit. The basic mechanisms of the model that are employed in the program are as follows [1], [2]:

- production of non-radioactive corrosion products in the primary system as a result of corrosion of the structural materials;
- formation of particles in the locations of the reactor coolant system, where the saturation concentration of soluble corrosion products is higher than the solubility multiplication value for the given thermodynamic conditions;
- formation of external oxide layer of corrosion products due to crystallization of the soluble phase, particle sedimentation, dissolving and erosion of deposit layer;
- presence of ion exchange among the coolant, particles and the deposit layer.

7. Design parameters employed in calculation of corrosion product activity in the primary coolant and at reactor plant surfaces

Core characteristics and process parameters forming the contamination level of coolant by corrosion products during normal operation and anticipated operational occurrences are given in Table 2 [1], [6]-[10].

Table 2. List of input data for calculation of corrosion activity in primary coolant and at reactor plant surfaces.

Parameters	Values
The ratio of the core surface to the surface of the reactor equipment flowed over by the reactor coolant	0.0531
Duration of the scheduled refuelling, h	720
Concentration of ammonia in the coolant, mg/kg	15
Concentration of hydrogen in the coolant, ml/kg	$30 \cdot 10^{-9}$
Concentration of H_3BO_3 at the beginning of campaign, g/kg	7
Nuclide activation rate, 1/s:	
^{58}Fe (n, γ) ^{59}Fe	$3.13 \cdot 10^{-11}$
^{58}Ni (n, p) ^{58}Co	$1.48 \cdot 10^{-11}$
^{50}Cr (n, γ) ^{51}Cr	$2.64 \cdot 10^{-10}$
^{54}Fe (n, p) ^{54}Mn	$1.09 \cdot 10^{-11}$
^{59}Co (n, γ) ^{60}Co	$1.26 \cdot 10^{-9}$
Burn-up rate of active nuclides, 1/s:	
- Fe-59	0
- Co-58	$1.09 \cdot 10^{-7}$
- Cr-51	0
- Mn-54	0
- Co-59	0

Equilibrium content of corrosion products in the primary coolant and at surfaces of primary equipment as calculated by the code COTRAN for long power operation of unit is given correspondingly in Table 3 and Table 4 [1].

Table 3. Activity of corrosion products in the primary coolant.

Activity of corrosion products, Bq/m^3						
Radionuclide	Fe-59	Co-58	Cr-51	Mn-54	Co-60	Total
Half-life, s	$3.90 \cdot 10^6$	$6.12 \cdot 10^6$	$2.39 \cdot 10^6$	$2.70 \cdot 10^7$	$1.66 \cdot 10^8$	-
Total	$2.78 \cdot 10^5$	$8.83 \cdot 10^5$	$2.17 \cdot 10^5$	$1.06 \cdot 10^5$	$6.28 \cdot 10^5$	$2.11 \cdot 10^6$

Table 4. Activity of corrosion products on the reactor coolant system surfaces.

Structural component	Activity of corrosion products, Bq/m^2				
	Radionuclide				
	Fe-59	Co-58	Cr-51	Mn-54	Co-60
Protective tube system	$1.00 \cdot 10^7$	$1.22 \cdot 10^8$	$7.47 \cdot 10^7$	$9.29 \cdot 10^7$	$5.99 \cdot 10^8$
Hot pipeline of main circulating pipeline (MCP)	$1.29 \cdot 10^7$	$1.52 \cdot 10^8$	$1.04 \cdot 10^8$	$1.29 \cdot 10^8$	$7.59 \cdot 10^8$
Hot SG collector	$2.69 \cdot 10^7$	$3.00 \cdot 10^8$	$2.50 \cdot 10^8$	$3.08 \cdot 10^8$	$1.54 \cdot 10^9$
SG tubing	$3.70 \cdot 10^6$	$3.13 \cdot 10^8$	$1.79 \cdot 10^7$	$2.29 \cdot 10^7$	$2.17 \cdot 10^8$
Cold SG collector	$2.97 \cdot 10^7$	$2.33 \cdot 10^8$	$1.56 \cdot 10^8$	$1.95 \cdot 10^8$	$1.92 \cdot 10^9$
Cold pipeline of MCP	$1.62 \cdot 10^7$	$1.42 \cdot 10^8$	$7.55 \cdot 10^7$	$9.62 \cdot 10^7$	$9.92 \cdot 10^8$
RCP	$5.07 \cdot 10^7$	$3.96 \cdot 10^8$	$3.26 \cdot 10^8$	$4.11 \cdot 10^8$	$3.96 \cdot 10^9$
Reactor pressure vessel	$1.33 \cdot 10^7$	$1.23 \cdot 10^8$	$5.92 \cdot 10^7$	$7.62 \cdot 10^7$	$7.96 \cdot 10^8$

As given in Table 4, activity of corrosion products on the primary system surfaces washed with the primary coolant is calculated under assumption that the cobalt content in the steel of the SG, the reactor coolant pipelines and the protective tube system is maximum 0.0915 %.

8. Conclusion

The article gave an assessment of radioactive waste production during normal operation of power unit in compliance with technical assignment for NPP for fuel element leakage corresponding to the operation limit.

It is shown that the sources of radioactive contamination of the primary coolant by fission products when the unit is operating at the rated power are defect fuel elements with gas leakiness and substantial damages, surface contamination of the outer surfaces of fuel claddings, superficial contamination of structural materials of fuel assemblies.

It has been proven that initially in the reactor operation, the contamination of the coolant by fission products is determined by the release into the reactor coolant circuit of fission fragments of Uranium-235 that is present on the outer surfaces of fuel elements as contamination in their manufacturing. The maximum allowed surface contamination of the fuel should be not more than 10^{-8} kg/m^2 for U-235.

It is shown that during normal operation of the reactor, the integrity of cladding may fail due to various processes of corrosion fatigue type. These processes result in, first of all, micro-fissures and then in large defects in the claddings, which is accompanied by an increase in the release of fission products from fuel elements into the primary coolant.

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Аналіз моделі розрахунку продуктів поділу в теплоносії першого контуру

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Анотація

Джерелами радіоактивного забруднення теплоносія першого контуру продуктами поділу при роботі блоку на номінальній потужності є дефектні тепловиділяючі елементи з негерметичністю та суттєвими пошкодженнями, поверхневе забруднення зовнішніх поверхонь оболонок ТВЕЛів, поверхневе забруднення конструкційних матеріалів паливних збірок. На початковому етапі роботи реактора (за відсутності виробничих дефектів тепловиділяючих елементів) забруднення теплоносія продуктами поділу визначається виходом у контур осколків поділу урану-235 (за рахунок їх кінетичної енергії), який присутній на зовнішніх поверхнях паливних елементів в якості забруднення при їх виготовленні. При нормальній роботі реактора цілісність оболонки може порушуватися внаслідок різних процесів корозійної втоми. Ці процеси призводять до появи спочатку мікротріщин, а потім великих дефектів оболонок, що супроводжується збільшенням надходження продуктів поділу з ТВЕЛів у теплоносій першого контуру.

Ключові слова: реактор; модель розрахунку; корозія; теплоносій першого контуру; продукти поділу.