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Study of Uranium-235 Fission Products in the WWR-SM Reactor Core

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Abstract. The article shows the necessity, as well as the technical and methodological feasibility of determining fission products in the water of the primary circuit and assessing the relative leakage of fuel assemblies of the reactor core. This is very important from the point of view of the further operation of a particular FA and determines the maximum allowable burnup depth in the FA. Timely determination of the relative leakage of fuel assemblies prevents the deterioration of the radiation situation at the facility, as well as the release of large activities into the atmosphere. And this, in turn, protects both personnel and a limited part of the population from increased exposure. Investigations of gaseous fission products in the upper reactor space were also carried out. The radiation monitoring system of the WWR-SM reactor constantly monitors the dose rate of water in the primary circuit and the dose rate of the upper reactor space. The system for monitoring radioactive emissions of the WWR-SM reactor is equipped with a point for continuous sampling of the gas-aerosol mixture entering the atmosphere. The technical support of the system for monitoring sources of release of radioactive substances from the WWR-SM reactor includes equipment that allows sampling for a filter package consisting of filtering and sorption-filtering materials.

INTRODUCTION

The high reliability and accident-free operation of the reactor is largely determined by the tightness of the fuel assemblies (FA). As a result of neutron irradiation, changes in the structure and physical and mechanical properties of materials of fuel elements (fuel rods) are observed: thermal conductivity, electrical resistance, elastic, mechanical and other properties.

As a result, the accumulation of fission products is accompanied by the appearance of internal overstresses in the material, an increase in gas pressure, which ultimately leads to the formation of cracks, swellings, and deformation of fuel elements. The tightness of the cladding is broken for damaged fuel assemblies, and radioactive gases and fission products penetrate into the coolant.

In this regard, the assessment of the relative leakage of fuel assemblies in the reactor core is a very urgent task.

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ACTIVITY OF NUCLIDES IN THE PRIMARY CIRCUIT WATER DURING REACTOR OPERATION

If the tightness of the fuel cladding is broken, fission products can enter the coolant, which is an additional source of coolant γ-radiation [1].

For the successful and safe operation of the reactor, it is also important to have information about the tightness of the fuel rods in the reactor core and in the spent fuel storage facility. The state of the fuel elements can be assessed on the basis of information on the specific activity of the fission products of uranium-235 in the coolant.

The release of fission products from the fuel composition into the coolant can be carried out in several ways [2]:

– due to recoil atoms during the fission of uranium-235 from the surface of the fuel cladding in contact with the coolant;

– due to the diffusion of fission products through the system of cavities and pores that appear in the fuel composition during the formation of microcracks.

During operation of the reactor, the activities of reference radionuclides in the coolant are measured. The assessment of the state of the reactor core is carried out using the values of the activities of reference nuclides [3].

Due to the short half-life ($T_{1/2}$=7.4 s) of ^{16}N as a radiation source (E_γ=6.13 MeV), it is of interest only during reactor operation at power; nuclides such as ^{13}N ($T_{1/2}$=10 minutes) and ^{18}F ($T_{1/2}$=109.771 minutes) can make a significant contribution to the γ-radiation dose rate at equipment in the first few hours after shutdown.

Table 1 shows the specific activities of nuclides in the water of the primary circuit during the operation of the reactor at nominal power, water samples in a volume of 5 ml were taken before the ion-exchange filter. 24 fuel assemblies with various burnups are loaded into the reactor core; the maximum burnup of uranium-235 does not exceed 60%.

TABLE 1. Specific activities of nuclides in the water of the primary circuit

№	Nuclide	Half-life $T_{1/2}$, hours	Sampling activity, µCi	Specific activity during sampling, µCi/l	Specific activity after 2 hours, µCi/l	Specific activity after 24 hours, µCi/l	Specific activity after one week, µCi/l
1	Nb-95	841,439	3,21E-03	6,43E-07	6,42E-07	6,30E-07	5,58E-07
2	Zr-95	1536,48	1,42E-03	2,84E-07	2,82E-07	2,80E-07	2,62E-07
3	Mo-99	66,02	3,69E-04	7,38E-08	7,22E-08	5,72E-08	1,26E-08
4	Ru-103	944,4	6,23E-04	1,25E-07	1,24E-07	1,22E-07	1,10E-07
5	Cd-109	11136	3,57E-03	7,14E-07	7,12E-07	7,00E-07	7,00E-07
6	I-131	192,96	4,59E-03	9,17E-07	9,10E-07	8,42E-07	5,00E-07
7	I-133	20,8	4,33E-02	8,66E-06	8,10E-06	3,88E-06	3,20E-08
8	Xe-135	9,11	8,02E-02	1,60E-05	1,36E-05	2,58E-06	4,52E-11
9	Cs-137	264463	4,01E-03	8,03E-07	8,02E-07	8,02E-07	8,02E-07
10	La-140	306,936	2,69E-03	5,38E-07	5,34E-07	5,08E-07	3,68E-07
11	Ce-141	780	5,42E-04	1,08E-07	1,08E-07	1,06E-07	9,20E-08
12	Ce-144	6823,2	3,21E-03	6,42E-07	6,40E-07	6,40E-07	6,30E-07
Total activity				**2,95E-05**	**2,65E-05**	**1,11E-05**	**4,07E-06**

The total activity of nuclides in all cases is less than the maximum allowable activity of water in the primary circuit - 2.5E-4 Ci/l.

Also, a water sample of the primary circuit was taken in a volume of 5 ml after the ion-exchange filter and the activity of nuclides was measured.

Table 2 shows the specific activities of nuclides in the water of the primary circuit during the operation of the reactor at nominal power, water samples in a volume of 5 ml were taken after the ion-exchange filter. 24 fuel assemblies with various burnups are loaded into the reactor core; the maximum burnup of uranium-235 does not exceed 60%.

TABLE 2. Specific activities of nuclides in the water of the primary circuit

№	Nuclide	Half-life $T_{1/2}$, hours	Sampling activity, μCi	Specific activity during sampling, μCi/l	Specific activity after 2 hours, μCi/l	Specific activity after 24 hours, μCi/l	Specific activity after one week, μCi/l
1	Nb-95	841,439	1,25E-04	2,51E-08	2,50E-08	2,46E-08	2,18E-08
2	Zr-95	1536,48	3,56E-04	7,12E-08	7,00E-08	7,00E-08	6,60E-08
3	Mo-99	66,02	4,18E-05	8,36E-09	8,18E-09	6,60E-09	1,43E-09
4	Ru-103	944,4	1,93E-05	3,86E-09	3,84E-09	3,80E-09	3,40E-09
5	Cd-109	11136	3,35E-04	6,70E-08	6,60E-08	6,60E-08	6,62E-08
6	I-131	192,96	2,35E-04	4,71E-08	4,66E-08	4,34E-08	2,56E-08
7	I-133	20,8	4,12E-04	8,25E-08	7,70E-08	3,96E-08	3,04E-10
8	Xe-135	9,11	4,22E-03	8,43E-07	7,24E-07	1,58E-07	2,36E-12
9	Cs-137	264463	2,32E-06	4,65E-10	4,64E-10	4,60E-10	4,60E-10
10	La-140	306,936	2,34E-04	4,67E-08	4,60E-08	4,40E-08	3,18E-08
11	Ce-141	780	3,66E-05	7,32E-09	7,20E-09	7,18E-09	6,30E-09
12	Ce-144	6823,2	4,52E-05	9,04E-09	9,04E-09	9,02E-09	8,80E-09
Total activity				**1,21E-06**	**1,08E-06**	**4,73E-07**	**2,32E-07**

The total activity of nuclides in all cases is less than the maximum allowable activity of water in the primary circuit - 2.5E-4 Ci/l.

TABLE 3. Lists the activities of nuclides in the upper reactor space

№	Nuclide	Half-life $T_{1/2}$, hours	Energy (KeV)	Activity during sampling, μCi	Specific activity during sampling, μCi/l
1	Kr-85M	4,48	402,57	3,70E-07	3,70E-10
2	Kr-89	0,05266	220,9	7,64E-06	7,64E-09
3	Xe-135	9,11	249,79	6,64E-05	6,64E-08
4	Cs-137	264463	605,06	5,42E-07	5,42E-10
Total activity					**7,49E-08**

The total activity of nuclides in all cases is less than the maximum allowable activity of nuclides in the air of the primary circuit - 7.5E-7 Ci/l.

The table shows that ion-exchange filters purify the water of the primary circuit well. With an increase in the activity of nuclides, more than control levels, the control of the tightness of the fuel assembly is carried out.

TABLE 4. Compares the activities of nuclides in the water of the primary circuit before and after the filter

№	Nuclide	Specific activity of nuclides before the filter, µCi/l	Specific activity of nuclides after the filter, µCi/l	Activity ratios of nuclides before and after the filter
1	Nb-95	6,43E-07	2,51E-08	2,56E+01
2	Zr-95	2,84E-07	7,12E-08	3,99E+00
3	Mo-99	7,38E-08	8,36E-09	8,83E+00
4	Ru-103	1,25E-07	3,86E-09	3,23E+01
5	Cd-109	7,14E-07	6,70E-08	1,07E+01
6	I-131	9,17E-07	4,71E-08	1,95E+01
7	I-133	8,66E-06	8,25E-08	1,05E+02
8	Xe-135	1,60E-05	8,43E-07	1,90E+01
9	Cs-137	8,03E-07	4,65E-10	1,73E+03
10	La-140	5,38E-07	4,67E-08	1,15E+01
11	Ce-141	1,08E-07	7,32E-09	1,48E+01
12	Ce-144	6,42E-07	9,04E-09	7,10E+01
Total activity		**2,95E-05**	**1,21E-06**	**2,44E+01**

CONTROL LEVEL OF ACTIVITY OF NUCLIDES

As a control level of nuclide activities, the calculated activity (Table 5) from uranium contamination from the surface of 24 fuel assemblies loaded into the reactor core was taken and the component, according to the technical passport, is 10^{-8} g/cm^2 for one fuel assemblies. Thus, based on the surface area of one fuel assembly - 1.37 m^2, the coolant for 24 fuel assemblies should contain 3.288×10^{-4} g of uranium, which corresponds to a concentration of 2.35×10^{-8} g/l.

TABLE 5. Calculated activity from uranium contamination

Nuclide	Activity, µCi	Specific activity, µCi/l	Nuclide	Activity, µCi	Specific activity, µCi/l
^{88}Kr	3,10E+09	5,59E-06	^{135}I	1,35E+10	2,43E-05
^{88}Rb	3,10E+09	5,59E-06	^{135}Xe	1,39E+10	2,50E-05
^{99}Mo	8,50E+09	1,53E-05	^{137}Cs	1,21E+07	2,18E-08
^{103}Rh	6,00E+08	1,08E-06	^{138}Cs	1,32E+10	2,38E-05
^{129m}Te	2,60E+07	4,68E-08	^{141}Ba	1,32E+10	2,38E-05
^{130}I	4,40E+09	7,93E-06	^{141}Ce	4,00E+09	7,21E-06
^{131}I	3,40E+09	6,13E-06	^{143}Ce	1,26E+10	2,27E-05
^{132}I	8,80E+09	1,59E-05	^{147}Pm	7,00E+07	1,26E-07
^{133}I	1,52E+10	2,74E-05	^{155}Eu	1,50E+06	2,70E-09
^{134}I	1,72E+10	3,10E-05	^{156}Eu	1,70E+07	3,06E-08

FA TIGHTNESS CONTROL

ATC stand is designed to determine the tightness of the fuel assembly cladding unloaded from the core. FA is loaded into a special canister, which is filled with distilled water, which is infused for 24 hours. Next, spectrometric measurements of water samples taken from the canister are carried out.

After holding for 24 hours, 10 water samples (0.5 l each) are taken from an ATC special canister. The dose rates of these samples are measured using the FH-40G device. Samples having higher doses are then taken to measure their activities using a GC detector. Measurements of sample activities are carried out on a gamma ray spectrometer consisting of a high-purity germanium detector HPGe.

To determine the concentration of radioactive activation products of inert gases and uranium-235 fission products in air, a filter package consisting of filtering and sorption-filtering materials was used. To do this, the investigated air of a certain volume from the ATC stand was pumped through a filter package, then the radioactivity was measured on a γ spectrometric setup.

To control the tightness of claddings of the IRT-4M type fuel assemblies with various burnups, measurements of the activities of nuclides in samples of infused water and a filter package were carried out. The total activity of fuel assemblies increases with increasing uranium-235 burnup, as it should be [4].

Figure 2 shows the activities of nuclides in samples of infused water depending on the burnup of uranium-235 in fuel assemblies.

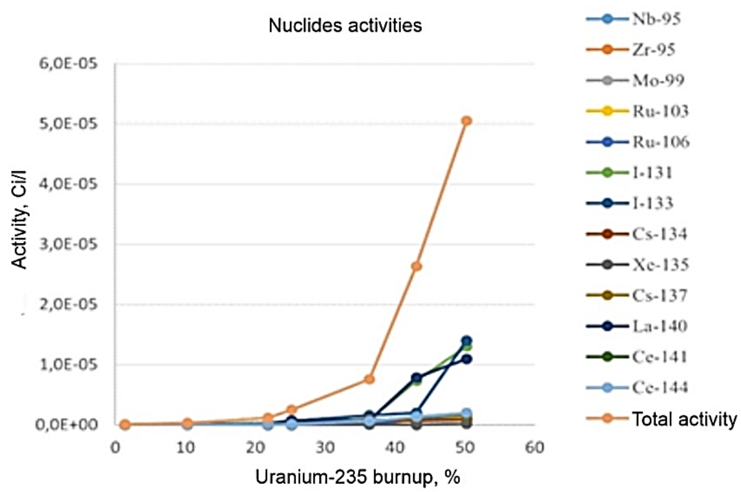

FIGURE 1. Activity of nuclides in samples of infused water depending on the uranium-235 burnup in fuel assemblies

The activities of nuclides (Ci/l) in the air depending on the uranium-235 burnup in fuel assemblies during sampling are given in Table 6.

TABLE 6. The activities of nuclides (Ci/l) in the air depending on the uranium-235 burnup

Uranium-235 burnup, %	**1,38**	**10,3**	**21,8**	**25,2**	**36,3**	**43,05**	**50,21**
Kr-85M	5,15E-11	4,1E-10	2,2E-10	4,1E-10	7,4E-09	7,5E-09	1,0E-08
Kr-89	8,65E-10	5,2E-10	2,9E-09	3,2E-09	7,1E-09	6,4E-09	1,0E-08
Xe-135	6,53E-10	4,2E-09	1,1E-08	1,5E-08	5,0E-08	6,6E-08	8,8E-08
Cs-137	3,65E-13	3,4E-12	3,7E-10	5,2E-10	6,4E-11	2,7E-10	5,6E-10
Total activity	**1,6E-09**	**5,2E-09**	**1,4E-08**	**1,9E-08**	**6,5E-08**	**8,0E-08**	**1,1E-07**

Knowing the regularity of cesium-137 activity in samples of infused water, depending on the uranium-235 burnout in fuel assemblies, allowed us to determine the uranium-235 burnt-out mass in fuel assemblies [5].

Table 7 shows the results of the cesium-137 activity measurements and, accordingly, the mass and burnup of uranium-235 in fuel assemblies.

TABLE 7. Results of the Cesium-137 activity measurements

№ FA	Initial mass ^{235}U	^{137}Cs activity on the day of unloading fuel assemblies from the core, Ci	Mass of ^{137}Cs, g	Measured mass of burnt-out ^{235}U, g	Burnup ^{235}U, %	Estimated mass of burnt-out ^{235}U, g	Measurement errors, %
1	314,1	515	5,95	96,04	31,57	99,16	3,25
2	305,3	652	7,54	121,59	41,1	125,48	3,20
3	304,4	887	10,26	165,42	56,1	170,77	3,24
4	301,6	915	10,58	170,64	58,42	176,19	3,25
5	308,1	949	10,97	176,98	59,3	182,7	3,23
6	305,5	950	10,98	177,17	59,9	182,99	3,29
7	300,2	936	10,82	174,56	60,02	180,18	3,22
8	310,8	975	11,27	181,83	60,4	187,72	3,24
9	303,3	956	11,05	178,29	60,7	184,1	3,26
10	303,2	980	11,33	182,76	62,22	188,65	3,22
11	306,7	997	11,53	185,93	62,6	191,99	3,26
12	314,8	1180	13,64	220,06	71,94	226,47	2,91

CONCLUSION

All results of the measurements performed show that no fuel leakage was observed, and the activity of all nuclides was lower than the allowable maximum activity for the fuel assembly, and the behavior of the fuel was expected. IRT-4M fuel assemblies, which were tested during the experiments, are in good condition, and the method used shows good results. The behavior of the activity of fuel assemblies as burnup increases is consistent with the well-known behavior of the activity of spent nuclear fuel.

All measured fuel assemblies were visually observed as far as possible. None of the measured fuel assemblies showed cracks, ruptures, blistering or other damage. This indicates the safe operation of fuel assemblies in the reactor core by operators.

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