Approaches to Deal with Irradiated Graphite in Russia – Proposal for New IAEA CRP on Graphite Waste Management - 12364

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Abstract

The problems of spent reactor graphite are being shown, the options of its disposal is considered. Burning method is selected as the most efficient and waste-free.

It is made a comparison of amounts of ^{14}C that entering the environment in a natural way during the operation of nuclear power plants (NPPs) and as a result of the proposed burning of spent reactor graphite. It is shown the possibility of burning graphite with the arrival of ^{14}C into the atmosphere within the maximum allowable emissions.

State of problem

Countries having the advanced nuclear technology are working out the various options for using spent graphite of uranium-graphite reactors with their decommissioning.

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At present, our country has two blocks of Beloyarsk plant that are stopped and number of industrial uranium-graphite reactors (Reactors for plutonium production). 11 units of RBMK reactors are working each has about 1,850 tons of graphite blocks and other parts of the active zones.

The total quantity of irradiated graphite of Russian reactors is about 50 000 tons (world-wide about 250,000 tons). A feature of the graphite is:

- presence of spent nuclear fuel, i.e. contamination of the fissile material and fission fragments (Table 1);
- accumulation of unstable isotopes (^{14}C, ^{36}Cl, T, etc.) in the graphite as a result of activation of the impurities of graphite by neutron in reactor (contamination of ^{14}C in the reactor graphite are presented in Table 2).

Table 1. Amount of the fissile materials in graphite reactor

Reactor	Nuclear fuel weight (U) in a stack, kg
AMB -100 (Beloyarsk NPP)	**70-150**
I-1 (Siberian CC)	**7,9**
I-2 (Siberian CC)	**3,6**
ADE-3 (Siberian CC)	**8**
AV-1,AV-2, AV-3 («MAYAK»)	**3-4**
IR-A1 («MAYAK»)	**2,75**

Table 2. Concentration of ^{14}C in the irradiated reactor graphite

Reactor	Specific activity ^{14}C, 10^7 Bq /kg
AMB-100 (Beloyarsk NPP)	**6,6 – 11,4**
RBMK-1000 (Chernobyl NPP)	**1,1**
IR-A1 («MAYAK»)	**1,8**
I-1 (Siberian CC)	**$1{,}9 \cdot 10^2$**
I-2 (Siberian CC)	**$1{,}1 \cdot 10^2$**
ADE-3 (Siberian CC)	**$1{,}0 \cdot 10^2$**
Sleeves (Siberian CC)	**4,4**

Ways to solve problems

It is considered various options for the treatment of spent reactor graphite. One option is to isolate the reactor at the location after extraction of the fuel without the removal of the graphite stack.

The graphite stack kept in the reactor space. The radiation-contaminated graphite blocks and the rests of the destroyed fuel are inside. Advantages of the method – there is no costs for the technological processes of reprocessing of

radioactively contaminated graphite. Disadvantages - the operating costs for the duration of conservation, monitor, prevent the possible release of radionuclides beyond the isolated volume (the creation of additional engineered barriers).

This method does not contradict to Russian legislation in the field of radioactive waste and the concept of long-term controlled storage "in place" is justified for the decommissioning of reactors for plutonium production.

Another option for the treatment of graphite during the decommissioning of the uranium-graphite reactors is the graphite stack removal and its placement in containers for further storage or disposal.

At current levels of contamination by long-lived radioactive isotopes the irradiated graphite should be buried underground. The deep of burial and the selection of the geological environment are determined by estimates of the possible migration of radionuclides beyond the cemetery.

It should be recognized [1] that the spent reactor graphite should be classified, and some of it classified as LLW so may be reprocessed in order to reduce the volume, for example, by oxidation providing allowable emissions of $^{14}CO_2$.

In the case of the dismantling of the graphite stack, containerization, and disposal of reactor graphite it is a strong requirement of separation the fissile material of the graphite mass.

Table 3 lists the most discussed methods of purification of irradiated graphite from fissile materials and shows advantages and disadvantages.

Table 3. Separation technologies of fuel spillage from graphite

	Advantages	**Disadvantages**
Processing of graphite with nitric acid to fuel dissolve		- the necessity of washing the treated graphite with water and subsequent drying, as a result – formation of the secondary liquid radioactive waste; - volume of i-graphite remains unchanged
Chlorination of fuel and chlorides distillation		- the necessity of chlorination of the entire volume of graphite; - the necessity of use of the complex high-temperature large-size equipment (approximately 6 - 15 times larger than the firebox for burning); - the necessity of creation

		of a complex gas cleaning system; - corrosion problems due to presence of chlorides; - volume of i-graphite remains unchanged.
The oxidation of graphite in molten salts	- transferring the fuel spillage in a form suitable for further conditioning. - partial capture of aerosols and ^{14}C by salt melt; - elimination of the entire volume of graphite	- low rate of oxidation of graphite due to the screening of the graphite by salt melt; - formation of an additional waste contaminated salt; - discharge of ^{14}C into the atmosphere
Burning of graphite in an air flow	- elimination of the entire volume of graphite; - the use of combustion equipment significantly smaller volume compared to the equipment using the salt melts; - fuel allocation into a compact phase.	- the necessity to create an efficient gas cleaning system; - ^{14}C release into the atmosphere.

Graphite burning in air flow is also attracting the attention of researchers in different countries [2]. Advantages: the elimination of almost the entire mass of the graphite with the use of furnace equipment is much smaller in volume in comparison with the volumes of equipment using salt melts, the fuel allocation into the compact phase. Disadvantage: the need to create a complex effective cleaning gas system from radionuclides.

Direct burning requires two options for the emissions treatment:

- prevent the release of ^{14}C;
- organize the reprocessing ensuring maximum concentration of ^{14}C in emission.

The first option is the most traditional way – it is capturing all CO_2, for example, in the form of barium carbonate and increase the mass of solid waste in 30 times. Exotic methods (isotope extraction, static centrifugation, separation of the gas stream, laser ionization and electrostatic separation) are ineffective and expensive [2].

To solve the problem of the second version it requires an update information on the concentration of ^{14}C in specific batches of graphite and the calculation of dispersed emissions evolving during burning.

Entering ways of ^{14}C into the air

Radiocarbon ^{14}C is continuously formed by the interaction of neutrons of cosmic origin with nitrogen of the upper atmosphere by the reaction $^{14}N(n, p)^{14}C$ at a speed of $1{,}48 \cdot 10^{15}$ Bq/ year. Exchange pool (the atmosphere, biosphere) in the equilibrium state contains 10^{19} Bq ^{14}C. The residence time of ^{14}C in the atmosphere is 8-10 years. During this time the most of the ^{14}C goes into the deeper layers of the ocean and in sediments [2].

Approximate average of annual emissions of ^{14}C from all working nuclear reactors in the world is $1{,}85 \cdot 10^{14}$ Bq [2, p.137].

During burning the spent graphite stack of RBMK-1000 reactor it will come (assessing it is expected the contamination of graphite on ^{14}C – $3{,}7 \cdot 10^{7}$ Bq /kg) ~ $7{,}4 \cdot 10^{13}$ Ci into the atmosphere, and the burning of graphite of 11 RBMK reactors it will come ~ $8{,}1 \cdot 10^{14}$ Ci in the atmosphere.

Table 4 shows the comparison of different ^{14}C streams.

Table 4. Sources of ^{14}C income

Source	Income, 10^{14} Bq/ year	Cosmic income, %	^{14}C part in exchange pool, %	Equilibrium activity ^{14}C in exchange pool, Bq	^{14}C activity in deep ocean, Bq
Cosmic radiation	14,8	100	$1{,}4 \cdot 10^{-2}$	10^{19}	$1{,}2 \cdot 10^{20}$
All NPPs	1,85	12,5	$1{,}8 \cdot 10^{-3}$		
Graphite burning of one RBMK per 1 year	0,74	5	$7{,}1 \cdot 10^{-4}$		
Graphite burning of 11 RBMK per 1 year	8,14	55	$7{,}8 \cdot 10^{-3}$		

The $^{14}CO_2$ emissions contribution formed during burning of spent reactor graphite into effective dose of radiation

The limiting effective dose of radiation for the population is 1 mSv / year [3]. The effective dose due to ^{14}C naturally occurring is 0.012 mSv / year [4]. It should be added 0,001-0,070 mSv/year to this dose - the actual annual doses of ^{14}C to the population in the area of NPPs [5], for a maximum total is 0.082 mSv/year. When the alleged burning of RBMK graphite reactor with an average of ^{14}C $3{,}7 \cdot 10^{7}$ Bq /kg per year the effective dose for the population living in the area of graphite burning will be 0.028 mSv/year.

Estimates of reachability allowable ^{14}C emissions

Burning graphite is necessary to ensure the maximum permissible ^{14}C concentration released into the atmosphere in flue gases formed during particular spent reactor graphite burning (Table 5).

Table 5. Russian radiation safety standards for ^{14}C

The maximum acceptable concentration in air	Bq/m3
For the staff	$1,3 \cdot 10^{6}$
For population	55
Natural background	$3,65 \cdot 10^{-2}$

It is necessary to estimate the local increase of ^{14}C concentration in the atmosphere by burning irradiated RBMK reactors graphite (Table 6).

Table 6. ^{14}C concentration in the near-ground air layer from the source with the power of $3,7 \cdot 10^{10}$ Bq/day, located on the high of 100m (calculation for the medium-range weather conditions**)**

The distance from the source, km	1	2	4	6	8	10
Activity of technogenic ^{14}C / The activity of natural background ^{14}C	1,76	2,65	1,76	1,20	0,88	0,69

Using the Table 6 data of we can estimate the quantity of irradiated graphite (to determine the activity of ^{14}C - $3,7 \cdot 10^{7}$ Bq /kg), which can be burned under the conditions of ^{14}C emission limit values (high pipe through which burning products release - 100-120m):

- for areas where there is no agricultural production – 50 tones per day;
- for areas where there is agricultural production – 4 tones per day.

Conclusion

This paper analyzes the different ways of spent reactor graphite treatment. It is shown the possibility of its reprocessing by burning method in the air flow. It is estimated the effect of this technology to the overall radiation environment and compared its contribution to the general background radiation due to cosmic radiation and NPPs emission. It is estimated the maximum permissible speeds of burning reactor graphite (for example, RBMK graphite) for areas with different conditions of agricultural activities.

List of references

1. Romenkov A. The achieved results. The treatment of radioactive graphite during decommissioning of uranium-graphite reactor using technology of the oxidation of graphite in molten salts. REA (Rosenergoatom) 2011, № 3, c. 32-35.
2. Rublevsky V.P., Yatsenko V.N., EG Chanyshev (ed. Kochetkov O.A.) The role of carbon-14 in the technogenic of man irradiation. - M: 2004.–197 c.
3. Radiation Safety Standards (NRB - 99/2009) SanPiN 2.6.1.2593-09. M. 2009.
4. United Nations. Sources and effects of ionizing radiation. United Nations Scientific Committee on the Effects of Atomic Radiation, 2000 Report to the General Assembly. Annex B: Exposures from natural radiation sources. United Nations, New York, 2000.

5. biometrica.tomsk.ru > ftp/medicine/jablock.htm