ON THE CHALLENGE ASSOCIATED WITH THE FINAL STAGE OF IRRADIATED GRAPHITE MANAGEMENT FROM WATER-GRAPHITE NPP REACTOR UNITS

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The paper discusses engineering, economic and legislative aspects influencing the ways of addressing the problem associated with the final stage of irradiated graphite management from water-graphite NPP reactor units in Russia. It presents some calculated and experimental data demonstrating the feasibility of introducing some corrective changes to the legislative and regulatory framework existing in the Russian Federation to enable near-surface disposal of irradiated graphite from power reactors.

Keywords: *radioactive waste, water-graphite reactors, decommissioning, irradiated graphite, radioactive waste management, legislative and regulatory rules.*

The decommissioning cost of NPP units being at the final stage of their life cycle, including the management of generated RW, directly affects the cost of products produced by nuclear power plants (electricity and heat).

To maintain the competitiveness of nuclear power in the electricity market, nuclear operators should constantly improve the efficiency of operations performed at all stages of NPP life cycle.

As it comes to the final stage, the latter suggests the reduction of relevant decommissioning costs. At the same time, according to different estimates, the share of RW management costs accounts for some 40—45 % from the total decommissioning cost per one NPP unit.

The above scattering is mainly explained by the differences in the design options implemented in pressurized water reactors (WWER-type reactor units) and high-power channel-type reactors (RBMK-type units) affecting the amount of RW generated from decommissioning.

In case of uranium-graphite reactors (UGR) of RBMK type, it is the final RW management stage involving the irradiated reactor graphite and replaceable parts of the graphite stack (bushings, rings, etc.), which is associated with their disposal, that accounts for the key element affecting the RW management cost.

An inventory of over 57,000 tons of reactor graphite has been accumulated to date in the Russian Federation [1], including:

- over 31,000 tons held in 13 production uranium-graphite reactors (PUGR);
- over 26,000 tons stored at NPP sites with UGR units (AM reactor of the world's first NPP, AMB-100 and AMB-200 reactors at Beloyarsk NPP, 4 EGP-6 reactors at Bilibino NPP and 11 RBMK-1000 reactors at Leningrad, Kursk and Smolensk NPPs) (Figure 1) [2].

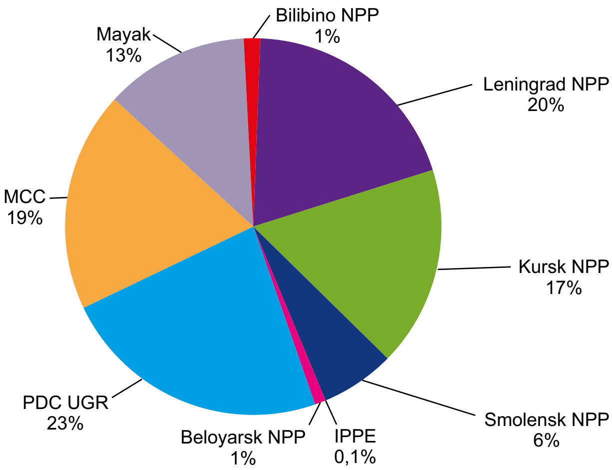

Figure. 1. The inventory of irradiated graphite accumulated at different sites, as a % of the total mass [2]

In the Russian Federation, the RW management procedure is regulated by the following federal laws: No. 170-FZ On Atomic Energy Use, No. 190-FZ On Radioactive Waste Management and on Amendments to Certain Legislative Acts of the Russian Federation and other regulations featuring some legal norms in the field of RW management, decommissioning of nuclear installations and storage facilities, as well as international conventions, norms and rules.

A number of studies performed in 2016—2020 under the Comprehensive Program for Irradiated Graphite Management from Uranium-Graphite Reactors at the Enterprises of the State Atomic Energy Corporation Rosatom [1, 3, 4] have demonstrated the potential for the industrial application of irradiated graphite decontamination or deep processing methods. However, according to further assessments performed with due account of the existing legal requirements, these were found to be technically and financially infeasible which was due to the high cost of relevant processes and equipment. In addition, the decontamination results in some secondary long-lived intermediate-level waste (ILW). Moreover, the resistance of such waste to external impacts (leaching, dissolution and oxidation) appears to be significantly lower than the one of the initial (primary) graphite RW.

Thus, the irradiated reactor graphite management method does not imply graphite decontamination. Therefore, it seems reasonable to consider irradiated graphite processing (treatment) only in case of nuclear fuel spill extraction from "damaged" graphite, i. e., to reduce the volume of retrievable waste pertaining to the 1st and 2nd RW classes.

According to [2], graphite from RBMK stacks (if nuclear fuel spills are absent) belongs to the 2nd class of retrievable solid RW and is subject to deep geological disposal (in DDF RW).

An approximate cost of deep geological disposal estimated for reactor graphite from an RBMK unit would include [2]:

- the cost of NZK-150-1.5P container with a SM-500 insert having an effective storage capacity of 1.15 m^3 — 225,000.00 RUB;
- the cost of graphite packing into NZK-150-1.5P container — 60,000.00 RUB;
- the disposal cost per 1 m^3 of RW packaging, i. e., gross volume — 684,733.50 RUB. (in 2020 prices) [5].

At the same time, the gross NZK-150-1.5P container volume accounts for 3.74 m^3.

In this case, the disposal cost per one NZK-150-1.5P container, excluding the transportation and intermediate storage cost, will amount to 2,845,903.29 RUB.

Taking into account the NZK-150-5P container filling factor, the weight of the graphite RW emplaced into it will be around 1.863 tons.

Therefore, a total of 13,420 NZK-150-1.5P containers will be needed to dispose of 26,000 tons of graphite RW and relevant costs associated with this option are estimated to amount to some 38.2 billion RUB.

The above estimates show how important and urgent is the task of reducing the management costs associated with reactor graphite generated from the decommissioning of NPP with UGR units operated by the Rosenergoatom Concern.

To minimize the cost of irradiated reactor graphite management, the feasibility and the environmental safety of an option providing for graphite RW disposal in a near-surface RWDF can be evaluated: it can potentially result in a several-fold reduction of its disposal cost, nevertheless, requiring the introduction of relevant amendments to some legislative acts.

The second way providing potential cost reduction is the development and the use of purpose-designed containers for reactor graphite: these are supposed to be similar in their designs to NZK type containers and fitted with some removable inserts of a new design. Since the beta emitting ^{14}C is considered as the main contaminant present in irradiated reactor graphite, the thickness of the protective container walls can be reduced, which will increase the net volume of the disposed waste while keeping the same gross volume (the NZK container volume) and reduce the container manufacturing cost.

The above proposals do not contradict the statement issued by the State Atomic Energy Corporation Rosatom based on the progress achieved in the industry program implementation [6]. Thus, to optimize the design solutions proposed for reactor graphite pre-disposal treatment, it is necessary:

a) to arrange for issue-specific studies of graphite from all NPPs reactors with UGR units to specify the distribution of ^{14}C and ^{36}Cl in graphite stacks, their integral amount, leaching rate from the matrix during the entire time period while the graphite waste potentially remains hazardous;

b) to precise the inventory of RW subject to disposal (according to RW classes) implying the assessment of reactor graphite waste inventory distribution by "new RW classes" (entombment option appears to be not acceptable for NPP decommissioning purposes [7]);

c) to search for and develop efficient packaging designs and to optimize the certification procedure for reactor graphite.

Considering the irradiated reactor graphite inventory, it seems feasible to establish a separate RWDF for its disposal. However, in this case, it seems more feasible to dispose of this graphite waste in near-surface disposal facilities, rather than in the deep ones.

The above suggestions take into account the following:

1. The immediate dismantlement option was adopted by Rosenergoatom Concern as a basic one for NPP unit decommissioning purposes [7].

2. Estimates performed by NRC Kurchatov Institute [8] have demonstrated that the dose rate from graphite stacks will decrease to the transport criterion level as soon as after 10 years of cooling (Figure 2), i.e., reach the upper limit established for the dose rate (no more than 2 mSv/h on the surface) allowing the transportation of graphite RW packages.

The basic NPP decommissioning option adopted by the Rosenergoatom Concern suggests that under the best-case scenario NPP graphite stack dismantlement will start 10—15 years after the final shutdown providing a decrease in the radiation dose rate emitted by graphite by up to 1 mSv/h.

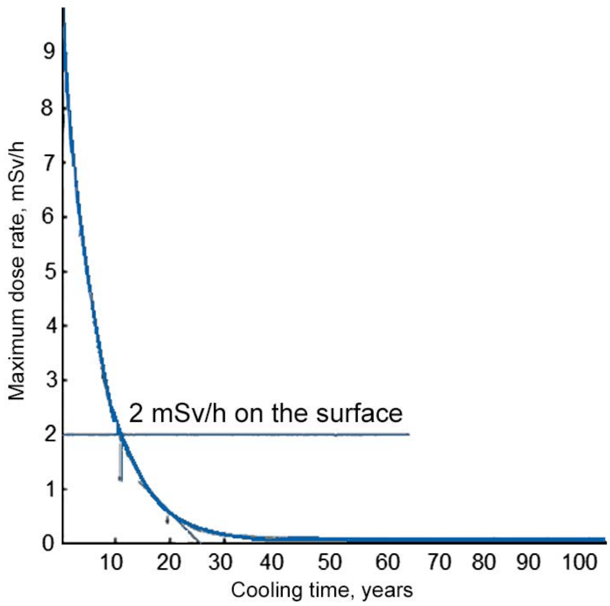

Figure 2. Equivalent dose rate from a graphite block depending on its cooling time

This will provide the use of containers designed specifically for graphite RW transportation with the conditioned graphite being disposed of in removable inserts allowing for some important reduction in the gross volume of disposed RW.

In 2015, PUGR EI-2 decommissioning project was implemented according to the entombment option at the site No. 2 of JSC PDC UGR [9]. To ensure the final disposal of the graphite stack in the reactor volume, internal cavities were backfilled with barrier mixtures based on natural clay. The material used to produce additional safety barriers was selected by the Institute of Physical Chemistry of the Russian Academy of Sciences according to the following criteria [10, 11]: low water permeability; low diffusion coefficients; high radionuclide sorption capacity; stability of properties over the entire time period while the RW potentially remains hazardous; absence of components capable of increasing the radionuclide mobility during barrier operation; stability of barrier properties assuming the availability of rocks with different water saturation levels; environmental safety; adequate load bearing capacity; availability; economic acceptability.

Environmental safety assessment focused on the above graphite disposal option was performed accounting for two scenarios describing the radionuclide migration: assuming normal evolution of the system and a most probable alternative scenario assuming the formation of a 1 m^2 crack.

The studies performed indicate that the rocks enclosing the PUGR shafts will maintain their protective barrier functions for at least 10,000 years.

Operation of a monitoring system available at EI-2 RW storage facility (SF RW EI-2) was analyzed revealing [12] no statistically significant impact of SF RW EI-2 on the environment in 2016—2020. At the same time, it should be noted that it was the initial period after SF RW EI-2 establishment that was considered as the most critical one in terms of negative consequences and their identification.

Reactor graphite (GR-280 grade) constituting to the stack of RBMK-type reactors operated in Russia and in the former USSR is practically identical to the PUGR graphite (GR-220 graphite grade). Differences between them are minimal considering the types of the raw material, production and treatment methods, density, structure (crystalline and porous), impurity composition, physical and mechanical properties and other characteristics [13]. Therefore, the accumulation degree and its specific aspects, as well as the spatial distribution of radionuclides in graphite depend on the specific aspects of RBMK and PUGR operation.

Considering the impact produced on the processes associated with the accumulation of radionuclides in RBMK graphite, the following points differ when it comes to PUGR and RBMK reactors:

- somewhat higher thermal neutron fluxes are typical for RBMK reactors, while their service life until the final shutdown is practically the same (as compared to PUGR ADE-2, -4, -5). Nevertheless, the form of neutron flux volume distribution in individual graphite blocks and the entire stack only slightly differs from the one being considered typical for PUGR units;
- RBMK graphite stacks were operated in a nitrogen-helium atmosphere (with a nitrogen fraction of 10 %), whereas PUGR stacks were operated in a high-purity nitrogen atmosphere: as compared to PUGR reactors, the contribution of the $^{14}N(n,\gamma)^{14}C$ reaction in gaseous nitrogen reduces to the formation of ^{14}C in case of RBMK reactor units;
- RBMK is characterized by lower radionuclide contamination of graphite associated with the irradiation of materials present in the fuel composition, since the number and the potential scale of incidents involving the ingress of fuel composition into the stack are typically lower when it comes to the operation of this reactor type, as well as the lighter are the potential consequences, the way they progress and the needs for their elimination;
- elements of the RBMK core such as pressure channels and cladding of fuel elements are made of zirconium alloy (with niobium being viewed as a basic additive), whereas the PUGR core is made of aluminum alloy suggesting that an increased content of radionuclides can be potentially observed in the near-surface layer of the graphite blocks. These are found in corrosion products of these structural materials (^{60}Co and Eu isotopes in case of PUGR; ^{60}Co and Zr, Nb isotopes in case of RBMK units). It should be noted that, despite the fact that ^{94}Nb ($T_{1/2} = 2.03 \cdot 10^4$ years) and ^{93}Zr ($T_{1/2} = 1.5 \cdot 10^6$ years) isotopes are considered as long-lived ones, their specific activity in graphite will not weight in on the retrievable RW categorization results. Moreover, these isotopes can be effectively contained by relevant safety barriers;
- RBMK graphite in certain sections of the graphite stack is operated at a higher temperature than the corresponding sections of the PUGR stack, which affects the degree of structural radiation-thermal changes and, accordingly, those features of radionuclide distribution and fixation that depend on relevant structural features (mainly porous structure, geometry and orientation of grains, etc.). This factor can affect the content and fixation strength of "non-impurity" radionuclides (^{14}C formed on gaseous nitrogen, activated corrosion products of structural materials, isotopes of "fuel" origin). In particular, the ^{14}C isotope formed from impurity nitrogen at higher irradiation temperatures is less susceptible to leaching due to the greater strength of fixation in the subsurface layers of pores and individual crystallites [14].

Considering the influence of the above differences on the radionuclide accumulation, one can conclude [2] that by the final shutdown of an RBMK unit, RBMK graphite blocks will be characterized by lower ^{14}C content (as estimated, by 1.5—2 times) as compared with the PUGR ones. This can be explained by the fact that, in comparison with the PUGR units (in particular, with ADE-2, -4, -5 operated for over 40 years), a decrease in the contribution of $^{14}N(n,\gamma)^{14}C$ reaction on gaseous nitrogen dominates over the positive contribution of higher RBMK neutron flux to the dynamics of ^{14}C formation. For RBMK, a tendency towards a slightly higher ^{60}Co content should be expected. The specific activity in this case, will tend to have the same order of magnitude as compared with the PUGR units. Moreover, there are no prerequisites for some fundamental differences considering the volume distribution of these radionuclides in individual RBMK and PUGR graphite blocks and stacks.

In case of RBMK graphite blocks, a slightly larger variation in ^{14}C content (relative to the average one) is considered possible, since, in comparison with the PUGR units, relative contribution of its formation from gaseous nitrogen decreases. Therefore, relative contribution of its formation from impurity nitrogen characterized by some important content variation in unirradiated graphite increases.

Until 2021, the largest experimental database on the radionuclide inventory of graphite from RBMK units being at the final stage of their operation (over 40 years) has been gained only considering the gamma-emitting radionuclides [15]. Since 2021, JSC PDC UGR assisted by some experts from the Institute of Physical Chemistry of the Russian Academy of Sciences named after A. N. Frumkin has been running larger-scale studies focused on the radionuclide content (including ^{14}C, ^{36}Cl, actinides) and their leaching involving some statistically representative arrays of samples from LNPP reactor units.

In [15], experimental data on gamma-emitting radionuclide inventory were presented based on the analyzed graphite samples taken along the height of the graphite stack cells from the Leningrad NPP reactor units (namely, units 1 and 2 of LNPP). It was shown that ^{60}Co isotope can be considered as the main dose-contributing radionuclide. For the most energy intensive areas of the graphite stacks, the variation in its content ranges from $1 \cdot 10^4$ to $7 \cdot 10^4$ Bq/g. Data on ^{60}Co inventory contained in the RBMK-1000 graphite from the Leningrad NPP (GR-280 grade) showed good agreement with the data on the PUGR graphite (GR-220 grade), taking into account the difference in thermal neutron fluxes, reactor operation time and the PUGR cool down after the final shutdown.

Until the beginning of 2021, the content of ^{14}C, ^{36}Cl in RBMK graphite was measured only for samples accounting for no coordinate reference within the stack volume. The experimentally measured specific activities of ^{14}C [2, 16] in the graphite samples taken from Unit 2 of Leningrad NPP were found to be ranging from $0.5 \cdot 10^6$ to $1.2 \cdot 10^6$ Bq/g. For ^{14}C contained in PUGR graphite, the maximum specific activity was found to be equal to ~ $2.5 \cdot 10^6$ Bq/g with the maximum value averaged over a "plateau" amounting to ~ $2.0 \cdot 10^6$ Bq/g. Thus, it can be stated that the experimental data on ^{14}C considering the graphite from Unit 2 of the Leningrad NPP demonstrates the correctness of the above estimates.

The specific activity of ^{36}Cl was experimentally measured for three weighed portions of graphite chips generated from the cutting of graphite blocks from LNPP Unit 2 reactor stack. These were found to be equal to ~ (500—570) Bq/g, which also falls within the range considered typical for PUGR units (up to $2 \cdot 10^3$ Bq/g) [3].

In 2017—2018, studied was the leaching of the key long-lived radionuclides from irradiated graphite. Irradiated PUGR graphite (block-type — grade GR-220, sleeve-type — grade GR-76) and samples of crushed RBMK-1000 block graphite taken from unit 2 of the Leningrad NPP (grade GR-280) generated due to maintenance performed to restore the resource characteristics of the stack were studied. The research showed [14, 17] that during ~ 1.5 years, the parameters characterizing the leaching dynamics for monolith samples of block PUGR graphite and crushed samples of block RBMK-1000 graphite slightly differ both as regards ^{14}C and ^{36}Cl (Figure 3).

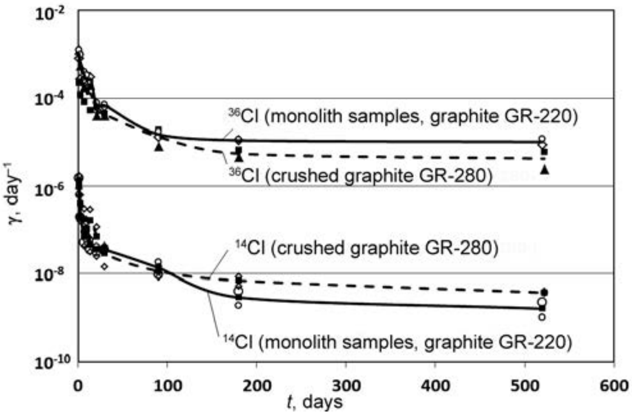

Figure 3. Dynamics of changes in the fraction of ^{14}C and ^{36}Cl leached out from the irradiated block graphite samples as per unit of time [14]

γ shown in Figure 3 characterizes the fraction of a radionuclide leached out from the irradiated block graphite samples per unit of time and can be viewed as a ratio between the activity of a radionuclide released into a contact solution per unit of time and the total activity of the radionuclide contained in the graphite (sample, fragment, integral part) present in the contact solution. This indicator introduced based on the studies performed [14, 17] can be used to estimate the resistance of irradiated graphite samples to radionuclide leaching.

It should be noted that when the safety of PUGR EI-2 RW storage facility was demonstrated, conservatively overestimated leaching rates were used as initial data in predictive calculations on radionuclide migration as compared to the recent studies [14, 17], i. e., these were overestimated approximately by two orders of magnitude in case of ^{14}C and one order of magnitude in case of ^{36}Cl.

RWDF evolution scenario was studied based on the case study of a concept developed for a SF located in the vicinity of the Sosnoviy Bor city close to the site of FSUE RosRAO's Leningrad branch [18]: compliance with the safety requirements specified concerning the RWDF impact (laid down in the scenario) on the environment and population has been demonstrated.

Under the present study, the calculated hydrogeological model is based on the actual hydrogeological conditions observed at the DF site. Certified computer code GeRa/V1 was used to assess the basic evolution scenario. Initial data on graphite activity with respect to ^{14}C and ^{36}Cl corresponded to the values predicted for RBMK units (see above) with the amount of graphite subject to disposal taken equal to 60,000 tons (conservatively overestimated). The same conservative margin was applied as regards the parameters characterizing the leaching rate as the one used in case of PUGR EI-2 SF RW.

The simulation involving 60,000 tons of irradiated graphite intended for disposal has shown that under the considered conditions, the specific activity of radiologically important radionuclides in the groundwater measured at the monitoring station would be less than 0.1 HC, which corresponds to the dose criterion of 10 μSv/year assuming drinking water supply (even in case of significantly overestimated parameters associated with the radionuclide leaching intensity from graphite).

The above results showing the differences in radionuclide accumulation in RBMK and PUGR graphite, as well as the data of experimental and model studies allow us to conclude that, in terms of its characteristics (isotopic composition, radionuclide content, leaching parameters), the potential hazard level of RBMK graphite will not be higher than the one of the PUGR graphite.

Therefore, these data may serve a basis allowing to demonstrate the feasibility and the environmental safety of a disposal option suggesting that the irradiated graphite removed from decommissioned UGR NPP units is disposed of in near-surface final RW disposal facilities.

Some other countries are also studying the prospects of irradiated graphite disposal. In particular, an average disposal depth (at least 30 m from the earth's surface) was proposed under the reactor graphite disposal concept developed in the UK [2]. The UK disposal concept involves a shaft lined with concrete with its wall and a base plate made of reinforced concrete acting as elements of a multi-barrier safety system. The cemented waste packed into containers will be disposed of in the mine and the remaining gaps will be backfilled with a cement mortar suggesting that a single monolith is obtained. The mine chamber will be backfilled as well: thus, the waste will be isolated by tens of meters from the ground level. A mound will be constructed on the surface to reduce water seepage into the repository.

In France, options suggesting near-surface disposal of irradiated graphite are also being considered in detail. However, both in France and in other Western countries, graphite waste is categorized as low-level waste potentially containing long-lived radionuclides (with the specific activity of 10^2—10^6 Bq/g), which is quite different from the Russian case. According to IAEA provisions discussing the RW classification system [19], a near-surface disposal option is recommended for this category of RW.

Stricter requirements are nevertheless effective in the Russian Federation: assuming the same activity characteristics, approximately 81—88 % of graphite inventory [2] belongs to the category of intermediate-level waste (ILW). Therefore, according to article 12, part 2 of the Federal Law of July 11, 2011 No. 190-FZ [20], the power reactor graphite shall be disposed of in deep disposal facilities. Summarizing the above discussion, it can be noted that:

1. Assuming the available RW conditioning methods, the cost of irradiated reactor graphite disposal in a deep RW disposal facility appears to be high.

2. Assuming the natural cooldown, in 10—15 years since the final shutdown of NPP units with water-graphite reactors, the radiological hazard of graphite will be mainly associated with long-lived ^{14}C and ^{36}Cl (β-emitters). Due to their decay, the decreased contribution of these dose-forming radionuclides (mainly ^{60}Co) will allow some important reduction in the radiation protection requirements established for the graphite RW packaging and further handling of waste containers up to their disposal in the repository. Thus, the option suggesting the use of lighter class containers with removable inserts appears to be quite feasible.

3. Differences in the processes associated with radionuclide accumulation in graphite PUGR and RBMK stacks were evaluated allowing to state that they do not produce any fundamental effect on the isotopic composition of the key radionuclides, as well as on the specific aspects of their accumulation and spatial distribution in RBMK graphite as compared with the PUGR one (except for the so-called radionuclides of a “fuel” origin). Minor differences (less than one order of magnitude) may appear in the characteristic values describing the amount of some individual radionuclides, which depends on the differences in the irradiation conditions and the variation in the structural characteristics of graphite over the graphite stack volume.

The parameters characterizing the dynamics of radionuclide leaching processes from irradiated PUGR and RBMK block graphite minorly differ both in case of ^{14}C and ^{36}Cl.

4. Conclusions presented in p. 3 along with the forecasted evolution of a conceptual RWDF proposed in the vicinity of the Sosnoviy Bor city, as well as some positive practical experience of JSC PDC UGR gained during scientific and technical, engineering, design development, safety demonstration and construction of a near-surface RWDF

during PUGR EI-2 decommissioning can be used as a basis to demonstrate the feasibility of the proposed near-surface disposal option for power UGR graphite. In this case, relevant costs can be reduced by more than 4 times.

5. Development of near-surface RWDF for graphite disposal purposes appears to be in line with relevant IAEA recommendations and the efforts implemented abroad.

6. Given the global scale of the challenge associated with finding some final solution for the reactor graphite disposal (a total inventory of over 184 thousand tons, excluding the one accumulated in the Russian Federation), methods, technologies and other technical developments resulted from the development of a near-surface RWDF for reactor graphite in Russia can stir some commercial interest abroad.

7. At present time, the disposal of irradiated power reactor graphite in near-surface RWDF contradicts the Decree of the Government of the Russian Federation No. 1069 and the provisions of the Federal Law of July 11, 2011 No. 190-FZ [20, 21], according to which long-lived ILW shall be disposed of in deep disposal facilities for RW.

To introduce the corresponding amendments to the above legislative acts, a feasibility study should be performed and a positive environmental safety statement on the possibility of reactor graphite disposal in a near-surface RWDF should be issued. The above papers should be used thereupon to develop the corresponding proposals, including the engineering specification for the development of a near-surface RWDF and the equipment required for reactor graphite conditioning and disposal.

Conclusion and suggestions

The following proposals were considered to reduce the cost of irradiated reactor graphite disposal:

1. To provide more specific data on the radiation characteristics of irradiated power reactor graphite to avoid conservative (exaggerated) estimates of its specific activity.

2. Environmental and feasibility study of a design option suggesting irradiated graphite container disposal in a near-surface RWDF should be performed assuming subsequent introduction of the required amendments to relevant provisions of the currently effective legislative acts and other regulations.

3. A new transport-packing container fitted with removable inserts of a larger useful capacity, but nevertheless being similar in its overall dimensions to the NZK type containers should be developed and certified allowing to reduce the gross volume of containers intended for graphite waste disposal.

We believe that all of the above measures and their implementation during the decommissioning of NPPs with UGR units will significantly reduce both the reactor graphite disposal costs and the total costs associated with the management of the generated RW.

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