Determining the optimal strategy of disposing irradiated graphite moderator

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I. Introduction

A large part of the nuclear reactors was commissioned in the 1970s and 1980s. Their service life was designed for 30-45 years, with the possibility of extension to 45-60 years. Thus, as of 2022, more than 200 power units were permanently shut down for further decommissioning, among which about 50 units are graphite moderated reactors [1].

Thereby appear complex engineer task on decommissioning this type of power units.

Dealing the final decision on irradiated graphite (*i-graphite*) treatment is complicated by several factors:

– presence of long-lived radionuclides in graphite pieces (foremost ^{14}C, ^{36}Cl);

– significant quantity of products - each power unit is up to 3,770 tons of this material;

– Most part of graphite radioactive waste is medium active isotopes (see Table 1 [2-6] for example).

Table 1 Composition of the graphite moderator of some reactors

Isotope	Specific Radioactivity, Bq/kg				
	Wylfa, United Kingdom	**JÜLICH, Germany**	**Vandellós I, Spain**	**Leningrad NPP, Russia**	**EI-2, Russia**
^{3}H	$5.46 \cdot 10^{8}$	$1.2 \cdot 10^{9}$	$2.75 \cdot 10^{8}$		$5.4 \cdot 10^{6}$
^{14}C	$2.21 \cdot 10^{7}$	$6.3 \cdot 10^{7}$	$5.62 \cdot 10^{7}$	$1.2 \cdot 10^{9}$	$1.4 \cdot 10^{9}$
^{36}Cl				$1 \cdot 10^{6}$	$1.1 \cdot 10^{5}$
^{60}Co	$1.23 \cdot 10^{8}$	$4.1 \cdot 10^{8}$	$1.34 \cdot 10^{7}$	$1.4 \cdot 10^{6}$	$3 \cdot 10^{6}$

The problem of disposal of spent reactor graphite is relevant for most countries with nuclear power plants. About 250 uranium-graphite reactors have been built in the world, and a significant amount of i-graphite has also been accumulated – about 250,000 tons. First of all, the problem of treatment of these materials is relevant for Great Britain – more than 77,000 tons [7], Russia – more than 50,000 tons, USA – more than 50,000 tons and France – more than 23,000 tons of graphite waste.

II. Properties of Irradiated Graphite

A key aspect in choosing a strategy of i-graphite treatment is the radioactivity values. It should be taken into account that the distribution of radioisotopes will be unique for each power unit (see Table 1). An unpleasant feature of the "main" isotope ^{14}C is a very long half-life – 5,730 years. Also, because of the especially radioactive ^{60}Co in the first decades after the shutdown higher safety precautions must be applied.

Generally, the longer the reactor has been in operation and the more powerful it is, the higher the total radioactivity. Meanwhile, other factors have an impact on the radiological inventory, among them are the initial purity of the reactor graphite [8] as well as the gas cooling the graphite pile. For example, when cooled with nitrogen, a noticeable portion of the irradiated carbon produced by the $^{14}N(n,p)^{14}C$ route (see Table 2).

Table 2 Comparison of ^{14}C generation rate in different reactors

Process\Reactor	^{14}C generation rate, Bq/W		
	Magnox	AGR	RBMK
$^{13}C\ (n,\gamma)\ ^{14}C$	4100	1300	3800~5500
$^{14}N\ (n,p)\ ^{14}C$	6700	2200	6400~9300
Total	10800	3500	10200~14800

In addition, during the operation of the reactors there is a partial destruction of graphite and spills are forming – a loose fraction of graphite, which also complicates the task.

III. Ways to Solve the Problem

Several different ways to solve the problem of irradiated graphite are now proposed:

A. Green Mound

In the green mound concept, the entire reactor would be buried underground (using containment varieties of clay, concrete, etc.). However, no materials and structures can remain intact and keep radioactive carbon from spreading until it decays.

B. Leave on station for a while

Temporarily leave the graphite in the reactor and wait until the activity decreases. Exposure for 30-50 or 80 years will indeed help to get rid of much of the alpha and gamma radiation, but the main source of radioactivity - the ^{14}C isotope will remain. Considering that the building of the power unit will need to be maintained for the entire period of exposure - from an economic point of view, the approach becomes questionable.

C. Remove and disposal

The most rational option is to remove the i-graphite from an NPP site with subsequent disposal. In addition, it is possible to pretreat graphite to change its radioactive waste class. This can be, for example, plasma firing of the outer layer, the use of special retention solutions and other methods. Part of the graphite (the most low-active) can be reused, for example, in geopolymer mixtures during cementation of containers with radioactive waste [9]. However, there may occur problems associated with graphite shredding – such as contamination of equipment, leakage of radioactivity, etc. Whereas the existing physical form of reactor graphite is the most compact and stable in terms of radionuclide migration.

As part of 'GRAPA' project, a general plan of i-graphite treatment and disposal was developed (see Figure 1) [10]. «Packing / Transportation» part of this plan requires suitable container. This approach is planned for implementation in the UK and France [11].

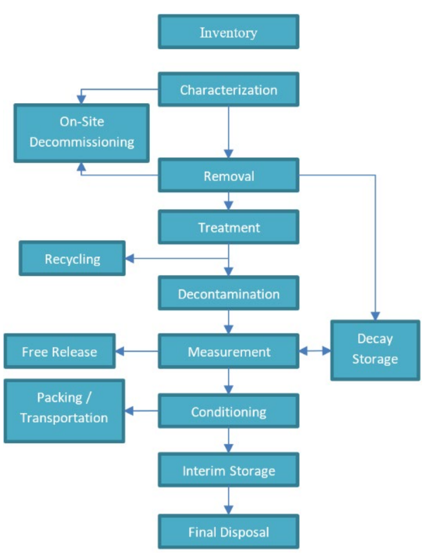

Figure 1: Target-Oriented Process Chain forming the Basis of the 'GRAPA' Project

IV. Concept of Packing and Removing i-graphite from an NPP Site

To ensure safe handling of graphite, after its extraction, it is necessary to determine how it will be stored and moved. GRAPA's plan envisages storage at each stage, and movement between "checkpoints" is through the use of transport packaging, which should preferably be "multipurpose" - it should be possible to use the container both for transport and for storage or burial of i-graphite.

We calculated ionizing radiation dose for single averaged i-graphite block (with implementation of a 10-year ageing time) (see Figure 2).

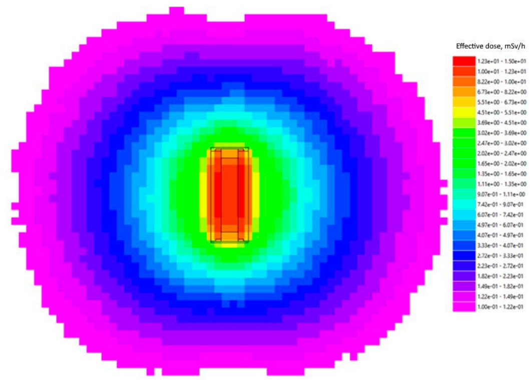

Figure 2: Ionizing radiation from single i-graphite block. Display range is 0.1 (pink) – 15 mSv/h (red)

In the first decades, the main dose is generated by the following nuclides: ^{137}Cs, ^{137m}Ba and ^{60}Co as γ-emitters and ^{90}Sr, ^{3}H and ^{14}C as β-emitters. Figure 3 shows the calculation of activity decline for graphite blocks with increased activity from the reactors mentioned in Table 1.

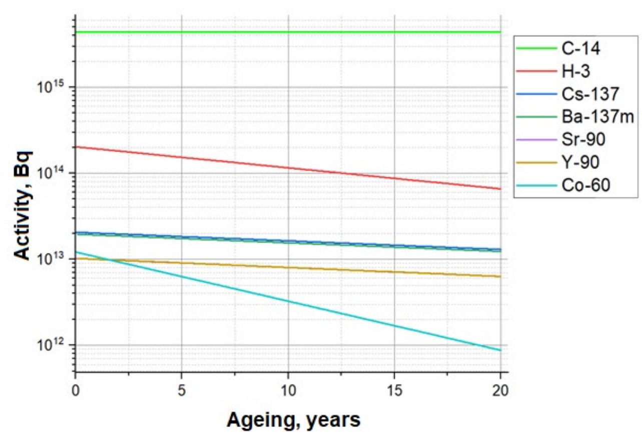

Figure 3: Activity of the main radionuclides in the period of 20 years after the reactor shutdown.

Based on the available data on the radionuclide composition of the irradiated graphite from different NPPs, the authors of this study updated the available calculation data of radiological protection [12] by modeling TUK-Graphite using modern software tools implementing the Monte-Carlo method - SCALE

(MAVRIC module) and PHITS in cooperation with the Laboratory of Nuclear Physics Research «Khlopin Radium Institute» (see Figure 4).

must remain airtight after being in a fire zone with a temperature of 800 ˚C for 30 minutes. When a container heats up, the pressure in its inner space filled with i-graphite block, concrete and air rises, which can lead to deformation of the walls and lid, and to decompaction of the container.

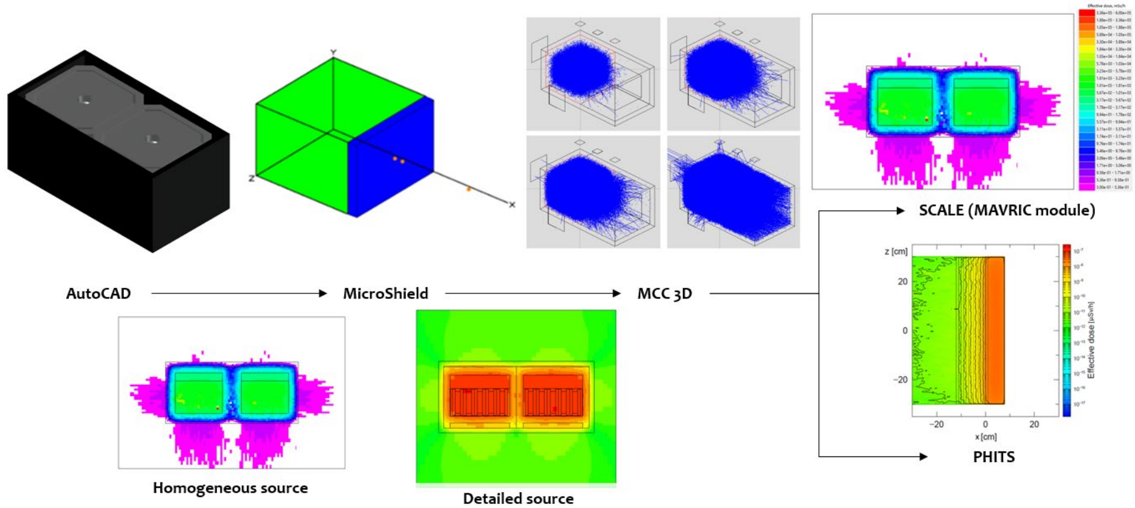

Figure 1: Transition from CAD model to radiation safety justification.

Based on the simulations carried out by the authors of this work, the following conclusions can be made:

— β-radiation does not have a significant impact on the design of the container;

— the vulnerable place for radiation safety reasons is the bottom of the container;

— in the case of graphite transportation, the maximum exposure radiation in the vulnerable area of the container and at a distance of 1 meter from the surface does not exceed 2 mSv/h if blocks are placed correctly.

Cast iron with nodular graphite was chosen as material for the container on account of cheaper production in comparison with steel (see Figure 5). Simple technological process of casting turns out to be cheaper than multi-stage assembly of steel sheets. Also, cast iron is not afraid of corrosion, combines durability and good mechanical properties (tensile strength and ductility), does not lose strength at low temperatures.

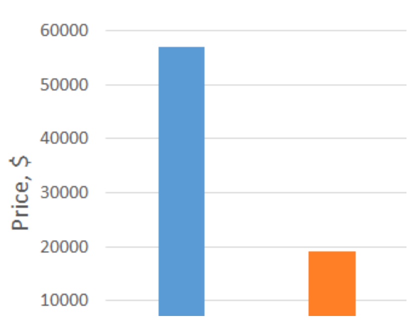

Figure 5: Comparison prices of containers for containers of different materials.

According to the IAEA Regulations [13], in addition to satisfying the requirements for radiation protection, the package

Simulation was done by research staff of the Peter the Great St.Petersburg Polytechnic University using ANSYS engineering software. Modeling of such situation for the conceptual container showed that the maximum pressure would not exceed 435 kPa at a temperature of 475 ˚C, and the tightness of the container would not be compromised (see Figure 6) [14].

Based on results, the suitable container was modeled (see Figure 7).

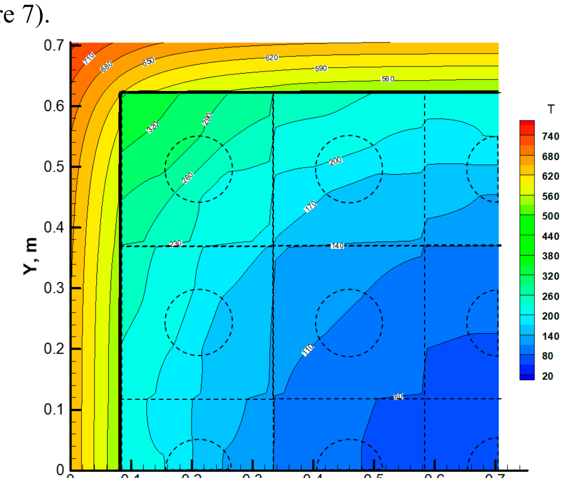

Figure 6: Temperature field in the cross-section of the container 30 minutes after the start of fire

Figure 2: Overall view of two versions of container.

V. Conclusion

At present, a sufficient number of unresolved problems have accumulated in the field of decommissioning of nuclear power plants and the management of radioactive waste and spent nuclear fuel. The final stage of the life cycle of nuclear facilities is not fully provided with effective and safe solutions. The fulfillment of processes and creation of technologies that will allow to ensure effective transition from the accumulated problems to the possibility of their timely solution is not only an interesting task, but also an urgent one for the next few years.

The right choice of strategy for the disposal of irradiated graphite will significantly reduce not only environmental but also economic risks. Removal and disposal of the most active part of the graphite moderator in the event of decommissioning of high-power uranium-graphite reactors will reduce the decommissioning time and, therefore, reduce the costs associated with the long-term preservation of a large nuclear facility under surveillance.

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