CArbon-14 Source Term

CAST

Report on graphite categories in the RBMK reactor (D5.3) Version 2

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CAST – Project Overview

The CAST project (CArbon-14 Source Term) aims to develop understanding of the potential release mechanisms of carbon-14 from radioactive waste materials under conditions relevant to waste packaging and disposal to underground geological disposal facilities. The project focuses on the release of carbon-14 as dissolved and gaseous species from irradiated metals (steels, Zircaloys), irradiated graphite and from ion-exchange materials as dissolved and gaseous species.

The CAST consortium brings together 33 partners with a range of skills and competencies in the management of radioactive wastes containing carbon-14, geological disposal research, safety case development and experimental work on gas generation. The consortium consists of national waste management organisations, research institutes, universities and commercial organisations.

The objectives of the CAST project are to gain new scientific understanding of the rate of re-lease of carbon-14 from the corrosion of irradiated steels and Zircaloys and from the leaching of ion-exchange resins and irradiated graphites under geological disposal conditions, its speciation and how these relate to carbon-14 inventory and aqueous conditions. These results will be evaluated in the context of national safety assessments and disseminated to interested stakeholders. The new understanding should be of relevance to national safety assessment stakeholders and will also provide an opportunity for training for early career researchers.

For more information, please visit the CAST website at:
http://www.projectcast.eu

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Report on graphite categories in the RBMK reactor

Executive Summary

For Ukraine, the main radiocarbon source is irradiated graphite from the Chernobyl Nuclear Power Plant. The ChNPP is a decommissioned nuclear power station about 14 km northwest of the city of Chernobyl, and 110 km north of Kyiv (Kiev). The ChNPP had four RBMK reactor units. The commissioning of the first reactor in 1977 was followed by reactor No. 2 (1978), No. 3 (1981), and No.4 (1983). Reactors No.3 and 4 were second generation units, whereas Nos.1 and 2 were first-generation units. RBMK is an acronym for "High Power Channel-type Reactor" of a class of graphite-moderated nuclear power reactor with individual fuel channels that uses ordinary water as its coolant and graphite as its moderator. The combination of graphite moderator and water coolant is found in no other type of nuclear reactor.

The approved «ChNPP Decommissioning Program» establishes a preferred decommissioning strategy.

This report presents information on graphite characterisation as is being undertaken as part of the ChNPP Decommissioning Program. Work considering chemical decontamination of graphite is reported, as is the overall graphite waste management approach.

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1 Introduction

For Ukraine, the main radiocarbon source is irradiated graphite from the Chernobyl Nuclear Power Plant. The ChNPP is a decommissioned nuclear power station about 14 km northwest of the city of Chernobyl, and 110 km north of Kyiv (Kiev). The ChNPP had four RBMK reactor units. The commissioning of the first reactor in 1977 was followed by reactor No. 2 (1978), No. 3 (1981), and No.4 (1983). Reactors No.3 and 4 were second generation units, whereas Nos.1 and 2 were first-generation units. RBMK is an acronym for "High Power Channel-type Reactor" of a class of graphite-moderated nuclear power reactor with individual fuel channels that uses ordinary water as its coolant and graphite as its moderator. The combination of graphite moderator and water coolant is found in no other type of nuclear reactor.

Parameters RBMK-1000	
Thermal power, MW	3840
Core diameter, m	11.80
Core height, m	7.0
Core volume, m^3	765
Mean specific power per core volume, MW/m	5.02
Mean specific power per fuel quantity, MW/t	20.8
Mean power per fuel rod length, kW/m	18.3

On September 9, 1982, a partial core meltdown occurred in Reactor No. 1 at the Chernobyl plant. The extent of the accident was not made public until several years later. The reactor was repaired and put back into operation within months. On Saturday, April 26, 1986, a disaster occurred at Reactor No. 4, which has been widely regarded as the worst accident in the history of nuclear power in the world.

The approved «ChNPP Decommissioning Program» [1] establishes preferred decommissioning strategy- deferred dismantling (SAFATOR). According to the National program of Chernobyl NPP decommissioning and Shelter object, decommissioning is to be carried out in few stages:

A. **Shut down (preparatory stage for decommissioning)** - stage during which nuclear fuel will be removed and transported in the Spent Fuel Storage Facility intended for long-term storage. The current stage - during which the major task, defining stage duration, is carried out – nuclear fuel should be removed from power units. Time for completion is not earlier than 2014.

B. **Final shutdown and preservation of reactor installations.** During this stage preservation of reactors and most radioactively contaminated equipment (roughly till 2028) will be carried out.

C. **Safe enclosure of reactor installations** for the period during which natural decreasing of radioactivity up to the acceptable level should occur (roughly till 2045).

D. **Dismantling of reactor installations.** During this stage the equipment will be dismantled and site will be cleaned with the purpose to release in maximum restrictions and regulatory control (roughly till 2065)

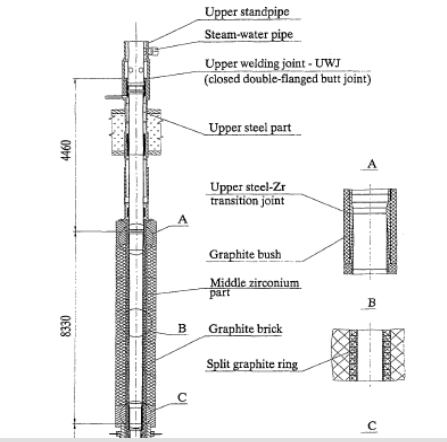
Today – there are significant delays in decommissioning the Chernobyl NPP.

2 Overview of graphite elements of RBMK-1000 reactor

Inside the hermetic enclosure of the reactor, mounted on the bottom plate is the graphite core consisting of 2488 vertical columns of graphite. The blocks are stacked inside the reactor vessel into a cylindrical core with a diameter and height of 14 by 8 meters. The moderator graphite blocks are manufactured from GR-280 graphite. The size of a blocks are 600 (500,300,200) mm high and by 250 mm x 250 mm square, with a central hole 114 mm in diameter through the longitudinal axis of the blocks for the fuel and control channels [2].

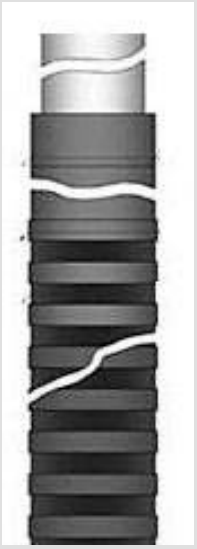
Installation of graphite stack (www.chnpp.gov.ua) Graphite blocks GR-280

The fuel channels consist of welded Zircaloy pressure tubes 80 mm in inner diameter with 4 mm thick walls, located through the channels in the center of the graphite moderator blocks. For protection from radiation, in the gap between the pipe channels are placed upper metal steel sleeves of 700 mm in height. The gap between the pipe and the channel path in the vicinity of the bottom metal is filled with graphite bushings. The pressure tube is held in the graphite stack channels with two alternating types of 20 mm high split graphite rings; one is in direct contact with the tube and has 1.5 mm clearance to the graphite stack, the other one is directly touching the graphite stack and has 1.3 mm clearance to the tube; this assembly reduces transfer of mechanical loads caused by neutron-induced swelling, thermal expansion of the blocks, and other factors to the pressure tube, while facilitating heat transfer from the graphite blocks.



Technological channels (TC) with graphite elements

The debris of split graphite ring



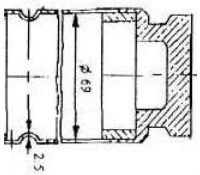
The split graphite rings are manufactured from a quality graphite GRP-2-125. About 80–85% of the core thermal power is removed by the fuel rod coolant channels, via the graphite rings. The small clearance between the pressure channel and the graphite block makes the graphite core susceptible to damage. If the pressure channel deforms, e.g. by too high internal pressure, the deformation or rupture can cause significant pressure loads to the graphite blocks and lead to their damage, and possibly propagate to neighbouring channels. In the reactor core unit "1" are 1692 FC[1] and 195 RCR[2] channels, in unit "2" there are 1693 FC and 195 RCR channels, and in unit "3" there are 1661 FC and 227 RCR channels. Most of the reactor control rods are inserted from above; 24 shortened rods are inserted from below and are used to augment the axial power distribution control of the core. With the exception of 12 automatic rods, the control rods have a 4.5 m long graphite section at the end, separated by a 1.25 m long telescope (which creates a water-filled space between the graphite and the absorber), and a boron carbide neutron absorber section.

[1] "FC": fuel channels, which are used for fuel clusters.

[2] "RCR channels": regulating control rods channels.

	
Unit 4 ChNPP -Reactor hall, 1983 (www.chnpp.gov.ua)	RBMK 1000-Reactor hall

The role of the graphite section, known as "displacer", is to enhance the difference between the neutron flux attenuation levels of inserted and retracted rods, as the graphite displaces water that would otherwise act as a neutron absorber (although much more weakly than boron carbide; a control rod channel filled with graphite absorbs fewer neutrons than when filled with water, so the difference between inserted and retracted control rod is increased).



The graphite section, known as "displacer", 1986	The displacer with graphite GR-93

In contrast to the rest of the graphite, graphite displacer control rods - present in a separate group - were irradiated at a temperature of not more than 50^0C and may contain stored energy (Wigner energy). Filling RCR consists of boron carbide enriched isotope ^{10}B + dysprosium titanate. (Incidentally, graphite was present in the end portion of the control rods more than 26 years ago in the RBMK RCR, but after the Chernobyl accident this was dismantled out of all the blocks in a modernization task). When the control rod is fully retracted, the graphite displacer is located in the middle of the core height, with 1.25 m of water at each of its ends. There is a need for additional surveys of displacer graphite control rods and for the development of technology for dealing with them.

The reactor operates in a helium - nitrogen atmosphere (70-90% He, 10-30% N_2). The gas circulation almost does not remove graphite heat, it only creates the condition for its effective removal.

3 Characterization of graphite used of Chernobyl NPP

Nuclear graphites in former Soviet Union have been manufactured from a range of raw materials using different manufacturing processes, this includes differing impurity levels.

Original impurities in the reactor graphite include a large number (up to 30) of naturally occurring elements with the concentration 10^{-4}–10^{-6}% by mass, many of which form long-lived radionuclides through the (n, γ), (n, p), (n, α), (n, 2n), and (n, t)- reactions. In addition to this group of radionuclides, actinides and fission product occur in the graphite due to several causes, the main of which is delivery of fuel particles to the graphite stack resulting

from leaks of coolant through leaky joints (mainly through bottom steel–zirconium couplings) and from the presence of non-tight fuel assemblies in the reactor.

Three major contamination sources for the graphite are:

A. Neutron activation of impurities;

B. Delivery of radionuclides as a result of technological incidents during the reactor operation;

C. External contamination of the graphite by radionuclides of corrosive origin that is present in the cooling pond (CP) water.

3.1 *Characterizations the microstructure of the graphite*

In framework agreement with IAEA, non-irradiated samples of the following graphites, which have been stored since the reactors were constructed, are being investigated:

- split graphite rings from technological channels of the Chernobyl NPP reactor (GRP-2-125 graphite);
- samples from graphite stack (GR-280).

The samples of irradiated graphite rings were taken after the termination of irradiation (1991). The graphite rings were kept in water in the cooling pond near reactor [3, 4], as was graphite after the Unit 4 accident.

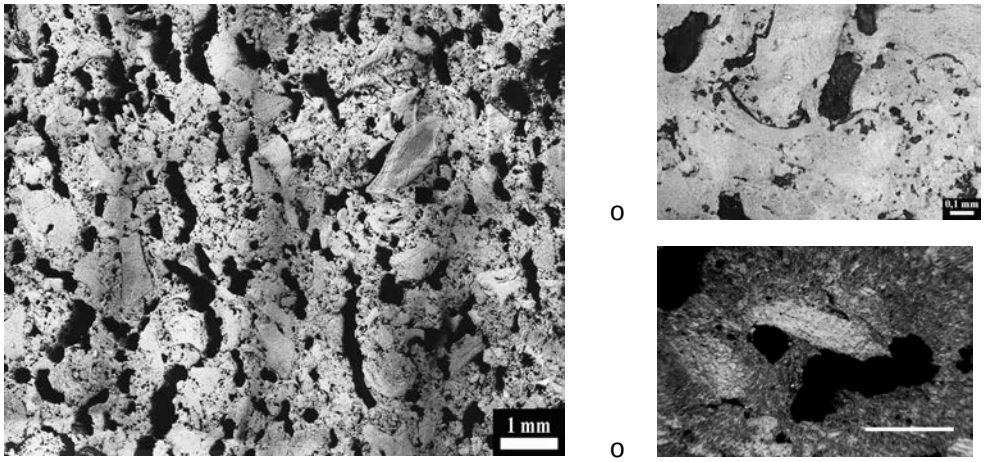
For characterization of graphite, the following techniques were used: optical microscopy, SEM and EDAX analysis; XRD analysis.

The research was carried out to investigate the structure and morphology of the surface layer of the graphite stack GR-280 and fine graphite composite GR-2-125, which has a polycrystalline structure with crystals held together by a binder phase.

The X-ray powder diffraction investigation (Cu K_α X-ray source) was the most readily available technique to measure the crystal structure of graphite. Silicon standard was used to determine the peak position and peak broadening. It was shown that the interlayer spacing

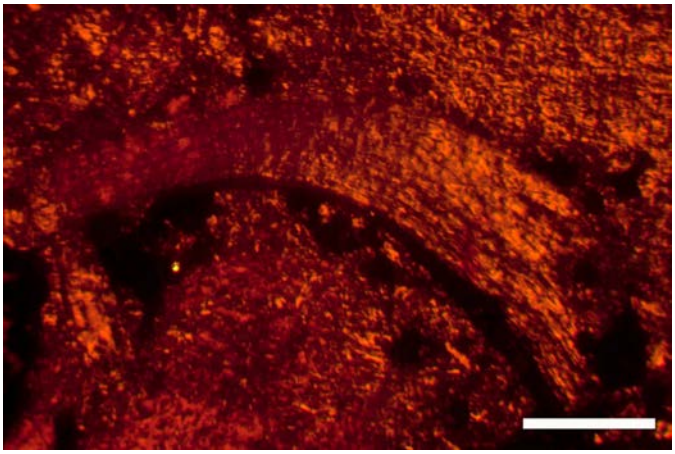
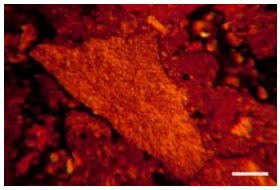
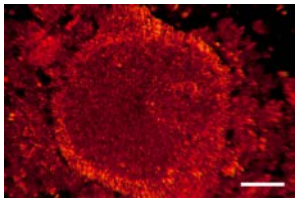
(d_{00l}) expands in the treated graphite. It was also established that the X-ray patterns indicated typical reflections characteristic of graphite, but the extent of their broadening was different. Crystallographic parameters has been calculated from the Scherer equation using the (00l) to Lc and (hk0) to La.

Based on diffraction picture, estimates were made of the distance between planes and the magnitude of the coherent scattering (OCD). The reflex broadening of the diffraction peak (00l) corresponds to the size of OCD ≈ 350Å. The diffraction picture of the powder sample indicates that, in fact, only one well-defined broad diffraction peak exists. The shape of this diffraction peak indicates the presence of two fractions with different dispersion and crystallinity properties in the sample. It is quite difficult to obtain high quality data for graphite just by using X-ray powder diffraction. The results of experiments were presented at the XX International conference on physics of radiation phenomena and radiation material science [5]



- The optical microscopy GR-280 graphite: the reflected light, Nicols is (II).

The optical microscopy GR-280 graphite was investigation microstructural characterizations of the materials, the fraction (crystalline) and pore size. The samples polished thin section and the reflected light with Nicols (II and +).

o  o  o 

oThe optical microscopy GR-280 graphite. The reflected light, Nicols is **(+).**

Filling:

Grain size of the filler is 0.5-1.5 mm, irregularly angled shape. The internal structures are homogeneous or banded more coarse aggregate. The crystal size is 10-20 microns. The bulk of the binder is fine-grained crystallites of about 1 micron.

Table 1. Fraction size of graphite

Fraction, size, μ	Graphite, GR-280	Graphite, GR-93
	Weight, %	Weight,%
>90	46	55
>500	22	19
>1200 - <500	18	26
>2200 - <1200	14	-

Adsorption of gases in the reactor graphite in the course of the reactor operation may to a large extent determine its activity [6], the major contribution to accumulation of ^{14}C comes

from the reaction on nitrogen $^{14}N(n, p)= ^{14}C$, and nitrogen is mainly accumulated through its adsorption during the reactor operation. Tritium in graphite arises from the reaction $^{14}N + n = ^{12}C + ^{3}T$. If this issue is ignored, wrong conclusions can be drawn and, accordingly, mistakes can be made in planning the removal of the Chernobyl NPP power units from service and solving the arising problems. All these uncertainties require serious experimental studies.

Therefore, only experimental determination of the isotope content and induced activity may yield real information on the radioactive status of the graphite. Some experimental measurements of C-14 inventory in the RBMK-1000 reactor graphite were performed by Ukrainian institute's Academy of Sciences (Institute for Nuclear Research, Institute Environmental Geochemistry) and Russian Research Centre Kurchatov Institute within the framework of a contract with Chernobyl Nuclear Plant.

In 2003, studies were conducted of the stored energy (Wigner energy) for graphite rings reactor Unit 2. The average temperature during the operation of graphite rings GRP-2-125 is 310^0C, and the total neutron fluence ~ 3E +22 neutrons/cm^2. It was established experimentally that the stored energy is 54-125 J/g for different rings, and the rate of accumulation does not exceed 0.376 J/g deg.

Due to the fact that the average temperature of the graphite stack during operation was about 500^0C, substantial amounts of Wigner energy are not accumulated in the graphite.

Experimental studies have shown that the total $A_{specific}$ for graphite rings is 10^{+4}-10^{+5}Bq/g, and is determined primarily by radionuclides: ^{14}C (55%), ^{3}H (4%), ^{36}Cl (5.5%). Some samples also recorded significant activity of ^{60}Co. The obtained data can be applied, to a first approximation, to the graphite GRP-2-125 reactor units number 1 and number 3, taking into account their effective working time on power/energy production per unit.

For the first time integrated b-spectrometric and radiometric investigations, α- and X-ray spectrometry of GRP-2-125 graphite from ChNPP Unit 2 were carried out. An analysis of obtained data has shown that it is necessary to carry out the comparison with experimental and calculated results of graphite activity research only on the basis of radioactive nuclide

^{14}C from irradiated reactor graphite. Nuclides ^{3}H and ^{36}Cl are distributed uniformly among the samples. Others nuclides are distributed inhomogeneously. Inhomogeneous distribution was detected for ^{60}Co, ^{90}Sr and ^{137}Cs only in one sample. Presence of ^{137}Cs and $^{154,155}Eu$ indicated a contamination of fissions products. Activity of irradiated GRP-2-125 graphite was defined of ^{14}C content as well as of content of radioactive nuclides of impurity and technological origin (^{3}H, ^{36}Cl, ^{55}Fe, ^{60}Co, ^{63}Ni, ^{93}mNb, ^{94}Nb, ^{133}Ba, ^{134}Cs) and fuel fission products (^{90}Sr, ^{137}Cs, ^{154}Eu, ^{155}Eu) [1]. Studies sampled graphite rings from Chernobyl RBMK-1000 reactor unit number 2. Technological channels number's V299, V358, V427, G209 and G75 (table 2) were investigated; these are sited in the "plateau" of the reactor and at the close of operation had approximately the same energy production - about 7 GW/day.

Table 2. Isotope content of the GRP-2-125 graphite rings.

Samples	Specific concentration, Bq/g			Total activity, Bq g^{-1}
	3H	14C	36Cl	
B 427- 603	690	4080	990	5760
B 427- 962	1230	12100	938	14268
B 427- 1062	490	8520	699	9709
B 427- 1145	1050	28000	1126	30176
G 75- 603	620	10840	1084	12544
G 75- 962	610	4000	1102	5712
G 75- 1062	1740	13280	943	15963
G 75- 1145	605	5600	1243	7448
G 209- 603	680	14400	1159	16239
G 209- 962	520	7240	1093	8853
G 209- 1062	820	14600	995	16415
G 209- 1145	750	10720	1384	12854
V 299- 603	710	8520	990	10220
V 299- 962	450	10600	1004	12054
V 299- 1145	980	9080	1478	11538
V 299- 1062	650	5360	1417	7427
V 358- 603	1290	18000	821	20111
V 358- 962	780	6320	929	8029
V 358- 1062	890	10400	1131	12421
V 358- 1145	420	8960	1084	10464

The article [7] deals with the results of the comparative analysis of specific activity and nuclide composition of graphite elements of ChNPP Unit 2 reactor channels (graphite of split rings and sleeves - GRP-2-125**)**. The comparative analysis was performed using the design data on ChNPP Unit 1 graphite as well as the experimental data on radioactive contamination of Russian industrial reactors graphite stack. From the analysis results it was

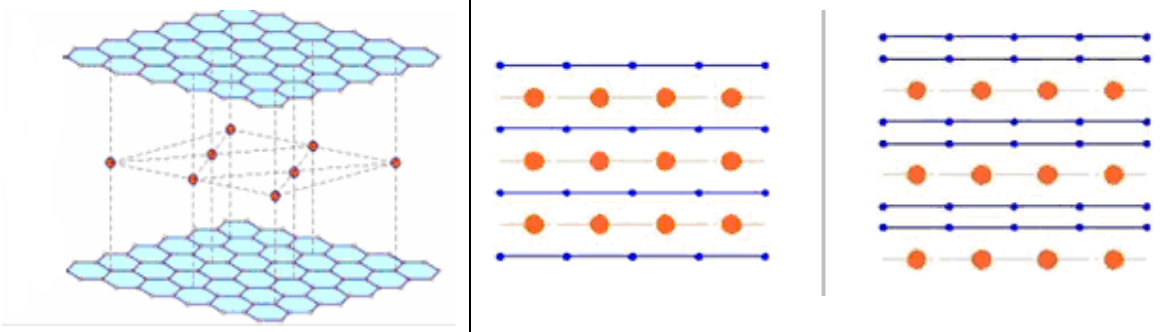
concluded that the design value of specific activity of main nuclides determining the radioactive contamination of the GRP-2-125 graphite was over-estimated in comparison with the experimental data. According to the estimations [8, 9], after the shutdown of the RBMK reactor the graphite stack and graphite elements of channels may account for as much as 80% of the total amount of radioactive waste. However, the estimates are to a large extent dependent on the impurity content of the graphite.

4 Chemical decontamination of graphite

The aim of the present research is to develop a technique for the decontamination of graphite using an intercalation process in an acid/ketone mix of solutions. The acid treatment of graphite is carried out by the wet process in which the graphite is immersed in a strongly oxidizing solution prepared by mixing a concentrated sulfuric acid with oxidizing agents.

Decontamination of Chernobyl NPP graphite is being investigated using an intercalation process. As a second step ("exfoliation"), cohesion between graphene layers is disrupted through ultrasound treatment. Intercalation compounds are formed by the insertion of atomic or molecular layers of a guest chemical species between layers in graphite material. The intercalation process occurs in highly anisotropic layered structures where the *intra* planar binding forces are very strong and the *inter* planar binding forces are very weak. When these conditions prevail, entire layers of 'guest' species can be inserted between layers of the graphite. For example, several hundred different chemical species are known to intercalate into non-nuclear graphite. Intercalation can be accomplished with various organic molecules (such as amines, pyridine, hydrazine, n,n-dimethylformamide and triethylmethylammonium methylsulfate. The introduction of each intercalate layer into the graphite adds a substantial strain energy as the crystal expands to accommodate the intercalate layer. The resulting interlayer intercalate repulsion favours the insertion of a minimum number of intercalate layers, consistent with a given average intercalate concentration. Thus for a given intercalate concentration, the minimal energy state corresponds to a close-packed in-plane intercalate arrangement with the largest possible

separation between a minimum number of intercalant layers. In practice it is possible to prepare single-staged graphite intercalation compounds (GICs) with only small (1-5%) admixtures of secondary-staged regions [10].

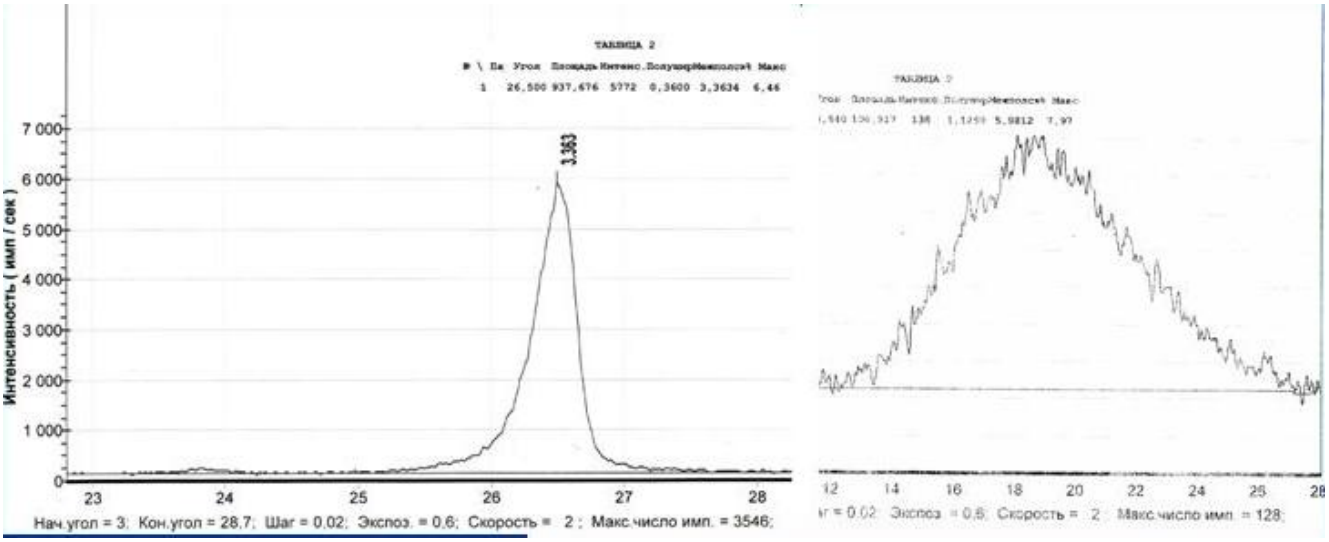


Scheme crystal lattice of intercalated graphite

4.1 *Experimental programme*

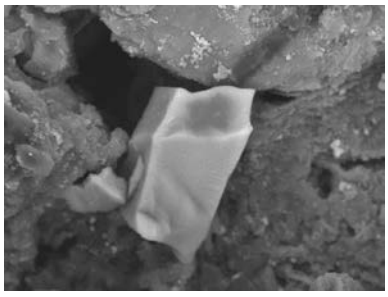
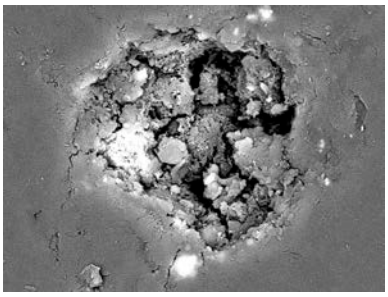
Graphite pieces were immersed in different ketones. The acid solution was prepared by mixing concentrated sulphuric acid and hydrogen peroxide (36%) in a ratio of 3:1. Chemical intercalation reaction and hydrolysis has a significant impact on the structural characteristics of the IG, the degree of graphitization is an increase in the amount of broken C-C bonds. The oxidation treatment markedly increased lattice spacing IG. The simplest method for stage characterization of intercalation compounds is x-ray diffraction based on (00l) reflections.

The aim of the present research is to development decontamination of graphite using an intercalation process in a mix of solutions acid/ketone. The samples used in this work were crushed and ground.



The 001 diffraction in GI, gradually disappeared with an increasing reaction time, implying the expansion of graphite layers.

The classic graphite intercalated compounds are prepared by the wet process by adding intercalation of sulfuric acid in the presence of an oxidant (HNO_3, $KMnO_4$, H_2O_2). Intercalation between layers of the graphite lattice should result in an increase the interlayer distance of graphite structure. The acid treatment of graphite was carried out by the wet process in which the graphite is immersed in a strongly oxidizing solution prepared by mixing a concentrated sulfuric acid with an oxidizing agent. Resulting surface morphology exfoliation structure shows numerous and complicated open canals.

o  o 

o Figure showing the surface morphology of graphite after treatment.

Surface of original samples maintain a much smoother surface than oxidized samples. Irregularities in a smooth surface increase when the oxidation is progressing, and are easy to observe.

Leaching experiments examine the release of C-14 from graphite after treatment. The results show that the 14C radioactivity leached from graphite without an oxide layer is less compared to 14C leached from the same samples graphite with an oxide graphene layer. The oxide graphene layer with 14C has a potential for volatilization.

5 Graphite waste management

At the Chernobyl NPP, a considerable amount of equipment and special items were used during the Power Units' operation. Currently, they are stored in cooling pools, technological shafts, and in a reactors core of Units 1-3. A total amount of these special items (long-length waste) is about 26.000 units. According to the design dimensions of these special items, they are from 6 to 22 meters in length and up to 145 mm in diameter. Technological channels include 19 pieces of the graphite sleeves and 163 graphite rings.

A 'preferred' decommissioning strategy for Chernobyl RBMK Units 1 – 3 envisages safe storage until around 2045 with dismantling complete by 2064.

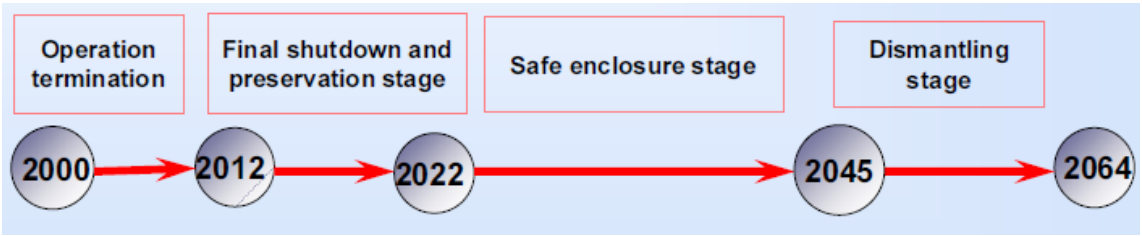


Table4. The waste inventory of irradiated graphite waste of Chernobyl NPP

Group	Volume, m^3	Weight, t	$A_{Specific}$. of ^{14}C Bq/g	Graphite type
Graphite stack	3732	5280	~ E+4 ÷ E+5	GR-280
Graphite rings and bushings	328	372	~ E+4 ÷ E+5	GRP-2-125
Graphite displacer control rods	22	35		GR-93

The commencement of grinding of long special items will form the first significant amount of graphite waste. By its radiation characteristics, waste from this source will be referred to the LLW-ILW. In addressing this problem at Chernobyl, account has to be taken of the fact that today in the world there is no unequivocal decision on how to deal with the contaminated graphite; tentatively-proposed technology for processing graphite is expensive, unproven and unprofitable. Therefore, Chernobyl is currently focused on developing long-term reliable technology and safe storage of radioactively contaminated graphite.

6 Conclusions

In compliance with the requirements of the regulatory documents, the composition and activities of radionuclides accumulated in structural materials and structures during the operation of the NPP power unit must be evaluated before their removal. The first to be examined will be the main sources of radionuclides, which are the structures of the reactor, especially the graphite stack and other graphite elements.

No specific waste-acceptance criteria have been defined. However, work is in progress on this issue by SNRCU in compliance with international standards as expressed in ICRP 60 which defines acceptable public and operator doses. Irradiated reactor graphite management determined by the Law of Ukraine "On Radioactive Waste Management» N 255/95-VR 30 June 1995 on the HLW and LLW

Issues to be addressed include:

- Uncertainty with terms of establishing a national geological repository, which may necessitate e.g. the construction of an interim storage facility;
- A lack of acceptance criteria for the disposal of graphite waste and appropriate container fleet, which in the future may result in a requirement to repackage i-graphite waste in storage;

- Verification techniques and equipment for the characterization of graphite waste are needed;
- There is a need for additional surveys of displacer graphite control rods to be undertaken and for the development of technology to deal with them;
- Handling graphite waste of the "Shelter".

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