STRATEGY FOR HANDLING AND TREATMENT OF INPP RBMK-1500 IRRADIATED GRAPHITE

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ABSTRACT

There are two RBMK-1500 water-cooled graphite-moderated channel-type power reactors at Ignalina NPP. After the final shutdown of the INPP, radioactive *i*-graphite dismantling, handling, conditioning, storage and disposal is an important part of the decommissioning activities.

The core of the INPP unit 1 and 2 contains about 3600 tons of *i*-graphite. Formation of activation products strongly depends on the contents of impurities, operational mode and concentration of impurities in the graphite.

The case study for INPP envisages the analysis of possibilities of graphite handling and treatment in the context of immediate decommissioning.

1. INTRODUCTION

The Ignalina NPP contains two RBMK-1500 type reactors. The core of the reactor is housed in 25m deep, 21x21m cross-section concrete vault. The core volume is dominated by a large cylindrical graphite stack. The graphite stack of the RBMK -1500 reactors serves several functions. The primary one is neutron moderation and reflection, but it also provides structural integrity and in the event of temporary malfunction, a relatively large heat capacity. The stack can be visualized as a vertical cylinder, made up of 2488 graphite columns, constructed from various types of graphite blocks. The blocks are rectangular parallelepipeds, with a base of 0.25x0.25m, and heights of 0.2, 0.3, 0.5 and 0.6m of which the 0.6 blocks are most common. The short blocks are used only in the top and bottom end reflectors, as required to provide a staggered fit to neighbouring columns. The total mass of columns graphite is about 1700 tons. The material GR-280 must meet stringent purity requirements and has a density of 1650kg.m^{-3}.

The four rows of columns at the outer edge make up the radial reflector, and a 0.5m thick layer at the top and bottom make up the end reflectors. The blocks possess a 0.114m diameter bore opening through the vertical axis. This provides a total of 2044 channels which are used for fuel clusters, reactivity regulating control rods and several types of instruments into core. In the remaining 444 columns located within the radial reflector the central holes are filled by graphite rods, increasing the density and neutron reflecting effectiveness of this part of the graphite stack.

In order to improve heat transfer from the graphite stack, the central segment of the fuel channel is surrounded by the 20 mm high and 11.5 mm thick split GRP-2-125 grade graphite rings. These rings are arranged next to one another in such a manner that one is in contact with the channel, and the other with the graphite stack block. The graphite stack, including its hermetically sealed cavity, is called the sealed reactor space. This space is filled with a circulating helium-nitrogen mixture (about 10% nitrogen) at an excess pressure of 0.5–2.0 kPa. Gas circulation improves heat removal from the graphite stacks, control rods and reflector, as well as protects stack from oxidation at high temperature conditions. In order to prevent loss of helium, the space surrounding the cylindrical graphite stack is filled with nitrogen at a pressure of about 0.29-0.98 kPa greater than that of the helium-nitrogen mixture.

2. ORIGIN OF GRAPHITE

Nuclear graphite GR-280 for the RBMK-1500 was manufactured from petroleum coke (КНПС-0.09mm-46%, 0.5mm-22% 05-1.2mm-18%, 1.2-2.3mm-14%) mixed with coal-based binder pitch heated and formed into bricks, and then baked at 1,000 °C for several days. To reduce porosity and increase density the bricks were impregnated with coal pitch at high temperature and pressure before a final bake at 2,500 °C. Individual bricks were then machined into the final required shapes.

It is important that graphite used in nuclear reactors contains a low level of impurities, particularly those which are effective neutron absorbers. The removal of boron from artificial graphite for nuclear purposes makes use of the high-temperature reaction between boron and chlorine to form volatile chlorine-containing compounds. Graphitized components are exposed to gaseous chlorine at temperatures of up to 2500^0C. Under these conditions any boron within the graphite reacts with chlorine to form boron trichloride, a volatile material which escapes from the graphite as a gas. As a result of this treatment the graphite may contain residual chlorine, but because of the high temperatures the amount is likely to be small. The results of measurements of stable chlorine in the graphite ring are 16ppm.

3. OPERATIONAL HISTORY OF INPP UNIT 1

During operation three gas cooling modes were used for the graphite stack:

(1) Dry air (where N_2 comprises about 80 %). This is for an auxiliary regime which is only used for a shutdown reactor.

(2) Nitrogen (100 % N_2). This is for an auxiliary regime which is used at the reactor power below 750 MW(e).

(3) Helium-nitrogen mixture (90 % He + 10 % N_2). This is for normal reactor power operation.

In 1984-1985 graphite stacks were cooled with the nitrogen gas. Over this period release of ^{14}C with cooling gas was 5GBq/MWt(el) year.

Since the middle of 1985 cooling was achieved by circulation of helium-nitrogen mixture (about 10% nitrogen). Flow rate - $400m^3.h^{-1}$, Tgraphite - 650^0C.

After maintenance of the Unit 1, every year the graphite stack was dried, and oxygen was removed within three day by $400m^3$/h flow rate of the nitrogen gas coolant until $CO+CO_2$ concentration went down to the required limits.

During 22 years of operation, the Unit 1 did not have any major incidents with rupture of fuel channels or water leakages into the graphite stack.

4. EXPERIMENTAL PROCEDURE

In order to analyze the condition of the graphite stack the following activities have been carried out:

- Programme of Graphite Sampling from the Graphite Stack has been developed;

- Equipment for remote graphite sampling has been designed and manufactured by INPP personnel;

- 4 CPS channels and 16 process (technological) channels have been retrieved for sampling purposes;

Sampling of GR-280 graphite stack – **completed, September 2013**
2012 - 40 specimens (graphite bushings and samples from columns without 4 CPS channels).
2013 - 150 specimens (samples from columns without 14 fuel channels).

- Gamma-spectrometric studies of samples have been carried out;

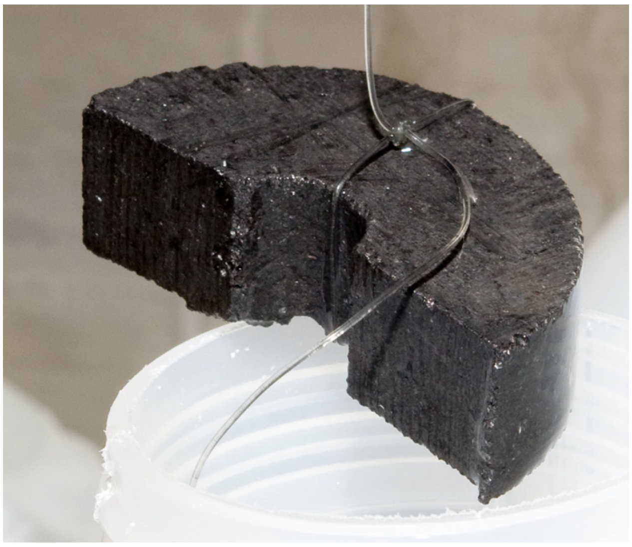
Core samples obtained from the graphite blocks РБМ-К5.Сб.05 contain the following nuclides with maximum values:

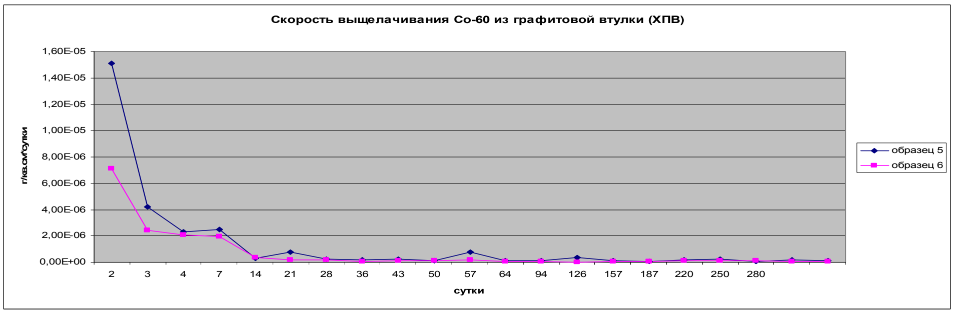
nuclide	*Co-60*	*Cs-137*	*Ba-133*	*Eu-154*	*Eu-155*
Max Bq/g Υ-activity	*6E+4*	*3E+2*	*2.6 E+3*	*1.3 E+4*	*7.9 E+3*

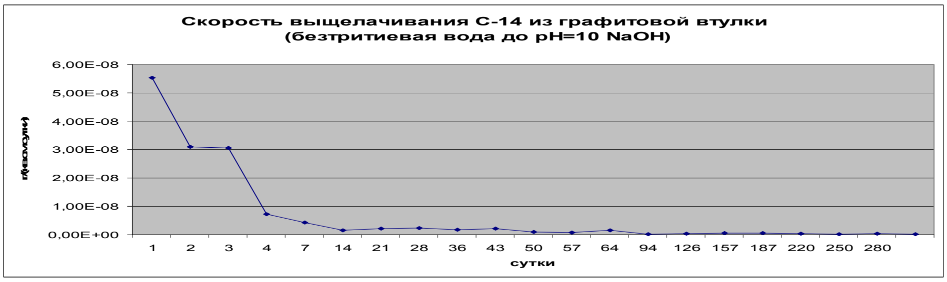
Studies of GRP-2-125 grade graphite rings have been carried out.

nuclide	***Inlet content Bq/g***	***Outlet content Bq/g***	***Outlet/inlet***
C-14	***2.6E+05***	***3.9E+06***	***15***
Co-60	***1.4E+4***	***3.6E+5***	***25***
Cs-137	***2.7E+2***	***2.3E+3***	***8***
C-14/Co-60	***18***	***11***	

- Tests on leaching of Co-60, C-14 from the reference bushing of the temperature channel made of GR-280 have been carried out.







4.1. Analysis of Results

The Final Decommissioning Plan presents the following data, Bq/g:

Nuclide	Graphite blocks in reactor core,Bq/g	Graphite sleeves in reactor core	Fuel channels in reactor core
C^{14}	6.3×10^{4}	5.5×10^{4}	3.0×10^{5}
Cl^{36}	2.6×10^{3}		1.3×10^{4}
Co^{60}	8.1×10^{4}	7.5×10^{4}	1.5×10^{9}

Specific activity is distributed depending on:

- Location in the CORE
- Type of item and material (thin ring or block,GR-280 or GRP-2-125)

- Mode of operation (T, composition of cooling mixture)

Comparison has demonstrated a higher rate of C-14 in the surface (up to 2 mm) layer of graphite rings. It is explained by the presence of 10% nitrogen in the cooling gas.

Concentration of Cl-36 in the irradiated graphite turned out to be almost 10 times lower that the expected one. It is probably linked to the decrease in concentration of stable chlorine as well as to the release of activated chlorine-36 during operation.

In the process of tests on leaching, Cl-36 release in the first day reaches 10%, and in 7 days it is lower than MDA.

Leaching for Co-60 is relatively high and in 14 days it reaches 3%, which is explained by its chemical properties. Over 1 year, up to 15% of initial activity is released. When pH gets higher, Co-60 leaching gets lower.

Major C-14 release lasts for 14 days, when organic phase on the surfaces and pores gets dissolved, and later it dramatically falls down, since release is preconditioned by properties of the graphite matrix.

It has been noted that C-14 release depends on pH environment – in more alkaline environment C-14 release is significantly higher than in the neutral environment.

5. STRATEGY FOR GRAPHITE WASTE MANAGEMENT

Based on the immediate dismantling strategy, the design for dismantling of process channels and CPS channels in the R-1 phase is being developed. Graphite rings and bushings will be separated and placed into 200-litre drums. 8 drums will be placed into a reinforced concrete protective storage container. For temporary storage (for the period of up 50 years), containers will be moved to the Temporary Storage Building. Amount of rings and bushings from 2 units – 200 tonnes.

It is suggested that graphite blocks should be retrieved as one piece and placed into FIBS containers without treatment. FIBS containers will be placed into reinforced concrete containers for temporary storage.

There is no final decision on the long-lived intermediate and low level waste (ILW&LLW) disposal option or disposal container in Lithuania. According to proposed generic repository concept for RBMK-1500 spent nuclear fuel (SNF) disposal in the crystalline rocks in Lithuania, the long-lived intermediate level waste (ILW) could be disposed of at the same repository at certain distance from

SNF emplacement tunnels [1]. The far field will consist of crystalline host rocks and a cover of sedimentary rocks of different hydrogeological properties (forming aquifers and aquitards). In the south of Lithuania, crystalline rocks occur at the depths ranging from 210 m to 700 m, while in the most of Lithuania the depth of the basement exceeds 700 m, reaching 2300 m in the west.

In case of graphite geological disposal, some modelling has been performed by Lithuanian Energy Institute (LEI) within the frame of EC Project on 'Treatment and Disposal of Irradiated Graphite and other Carbonaceous Waste (CARBOWASTE)' under the 7th EURATOM FP (FP7-211333) [2]. Within this study, a cementitious grout (NRVB backfill) was assumed as proposed in the Nirex concept (United Kingdom) to fill void regions within the tunnels after the emplacement of the long-lived waste. The relation between treatment and disposal on repository performance was analysed by the way of radionuclides transport modelling from geological repository, as the radionuclides eventually released from the irradiated graphite waste could be transported to the surface and lead to human exposure. Modelling of C-14 migration in the near field and far field was performed with the source term based on LEI modelling results on RBMK-1500 inventory, illustrative rates for representation of possible differences on non-treated/treated graphite and conceptual models developed. The importance of waste leaching rate was studied within the context of different performance of engineered barrier (in terms of sorption, limited solubility) and considering possible graphite encapsulation in cementitious material. It was concluded that the importance of waste leaching rate depends on several aspects: on the performance of the backfill and natural barrier system (on the scope of its impact on the attenuation of the radionuclide flux). The impact of the options (treatment *versus* non-treatment of graphite) on the C-14 flux to geosphere is not straightforward. While reasoning of option of treatment/non-treatment the inventory, leaching rates, barrier performance and transport conditions need to be considered.

6. CONCLUSIONS

Ways of handling of graphite originating from dismantling of the units have been identified for the period of interim storage. Treatment and final disposal possibilities and options will be studied in the near future.

REFERENCES

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