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CAST „CARBON-14 SOURCE TERM“

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„REPORT ON MODELLING OF C-14 INVENTORY IN RBMK REACTOR CORE“

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CArbon-14 Source Term

CAST

Report on modelling of C-14 inventory in RBMK reactor core (D5.17)

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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 release 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
Executive Summary

This report is one of the CAST Work Package 5 (WP5) deliverables and it presents results of Lithuanian Energy Institute (LEI) activities performed in the Task 5.2 “Characterisation of the C-14 inventory in i-graphites”.

The aim of this work is to perform the neutron activation modelling of Ignalina Nuclear Power Plant (Ignalina NPP) Unit 1 reactor RBMK-1500 graphite stack and determine C-14 inventory. In order to achieve this aim, new numerical models for neutron activation are developed and activity distribution of C-14 within whole reactor graphite stack is obtained. Then, combining this data with the available C-14 activity measurement results, estimation of C-14 inventory in the graphite stack is made.

Modelling of neutron flux was performed using MCNP 5 ver. 1.6 code. The newly developed model envelopes the whole reactor core with the graphite stack consisting of 2488 graphite columns and surrounding structures, in the space region of 21x21x15 m (21x21 m is the length and the width of a concrete reactor vault while 15 m is the height of the modelled reactor core including surrounding structures). The modelled neutron flux was grouped into 238 energy groups (the most detailed discrete neutron energy group structure used in SCALE codes system). The modelled neutron flux in the 238 groups structure then was used by COUPLE code from SCALE 6.1 codes system, for automated preparation of problem-specific cross-section data for ORIGEN-S code (also from SCALE 6.1 codes system). Having problem-specific cross-section data and using total neutron flux, the neutron activation modelling of the whole reactor RBMK-1500 graphite stack was
performed. The modelling results revealed the theoretically possible 3D distribution of the C-14 inventory in the reactor graphite stack, i.e. possible C-14 inventory heterogeneity.

A preliminary study to estimate experimental C-14 activity in several graphite sub-samples of the GR-280 grade graphite bushing from the temperature channel of the Ignalina NPP Unit 1 RBMK-1500 reactor revealed that the average value of the C-14 specific activity in this graphite was $1.67 \times 10^5$ Bq/g (study performed by Ignalina NPP and Nature Research Centre staff, Lithuania).

By having the modelled distributions of specific C-14 activity in each group of the graphite columns (in normalised units), by knowing the correspondence of measured specific C-14 activity to the exact point (location) of the modelled specific activity distribution, and by applying quantities and masses of the graphite columns in the respective group, the integral C-14 activity in the graphite stack was calculated.

As a main emphasis of this report it could be stated that, combining C-14 activity modelling and measurement results, the following numbers regarding C-14 inventory in the irradiated graphite stack of the Ignalina NPP Unit 1 RBMK-1500 reactor at the time of reactor final shutdown were obtained:

- Total C-14 activity in the graphite stack: $3.222 \times 10^{14}$ Bq;
- Total mass of the graphite stack: $1.700 \times 10^9$ g (1700 t);
- Average C-14 activity in the graphite stack: $1.895 \times 10^5$ Bq/g.

However, it should be also noted that the above numbers are only for the reactor RBMK-1500 graphite stack consisting of GR-280 grade graphite blocks. The remaining graphite components in the reactor (graphite rings, sleeves, rods and other graphite parts in minor quantities) are not accounted. For these parts, as a first attempt, the same average specific activity could be applied, which together with the mass of ~200 t results in $3.790 \times 10^{13}$ Bq of C-14. Finally, this sums to $3.601 \times 10^{14}$ Bq of C-14 in ~1900 t of the irradiated graphite of all types in one RBMK-1500 reactor.
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1 Introduction

This report is one of the CAST Work Package 5 (WP5) deliverables and it presents results of Lithuanian Energy Institute (LEI) activities performed in the Task 5.2 “Characterisation of the C-14 inventory in i-graphites”.

1.1 Aim of the work

The aim of this work is to perform the neutron activation modelling of Ignalina Nuclear Power Plant (Ignalina NPP) Unit 1 reactor RBMK-1500 graphite stack and determine the C-14 inventory. In order to achieve this aim, the new numerical models for neutron activation are developed and activity distribution of C-14 within the whole reactor graphite stack is obtained. Then, combining this data with the available C-14 activity measurement results, estimation of the total C-14 inventory in the graphite stack is made.

1.2 Description of the RBMK-1500 reactor

Ignalina NPP contains two power Units with RBMK-1500 reactor in each. While in operation, these reactors were the most powerful and most advanced versions of all RBMK reactors. Since the end of 2004 y, Unit 1 is permanently shut-down (its start-up dates back to the end of 1983 y). After being more than 10 years in a decommissioning stage, a number of systems and equipment of Unit 1 were radiologically characterised and dismantled. However, these works were performed for relatively low contamination systems, while the reactor itself is intact (although defueled) and not radiologically characterised. The highest volume of reactor structures is attributed to the graphite core, so radiological characterisation of it still is an open issue, affecting dismantling and temporal storage strategies as well as final disposal routes (especially C-14 inventory).

A general view of the RBMK reactor is presented in Figure 1, while the cross-section of it is presented in Figure 2. The RBMK-1500 is a graphite-moderated, water-cooled, channel-type boiling water reactor having a thermal power generation capacity of 4800 MW, which was decreased to 4200 MW after the Chernobyl accident.
The reactor is housed in a 25 m deep, 21x21 m wide concrete vault. The core volume is dominated by a large cylindrical graphite stack (6, Figure 2). The graphite stack is located in a hermetically sealed cavity, consisting of cylindrical wall (7, Figure 2) and top and bottom metal plates (2 and 8, Figure 2), which is filled with a helium-nitrogen mixture (nitrogen ~60 % by mass) preventing graphite oxidation and improving heat transfer from the graphite to the fuel channels. This also excludes the radiolytic corrosion of graphite that is common for gas-cooled (CO₂) graphite reactors, thus the mass loss of graphite is not specific for the RBMK reactors.

The stack can be visualized as a vertical cylinder of 8 m height and 14 m diameter, made up of 2488 graphite columns arranged next to each other without keying. The outer four rows
of columns of the stack make up the radial reflector (RR), and a 0.5 m thick layer at the top and bottom make up the end (top and bottom, respectively) reflectors. The total mass of graphite blocks in a whole graphite stack is about 1700 tonnes.

Figure 2: Vertical cross-section of the RBMK-1500 reactor vault (reproduced from [2])

Every graphite column is constructed from graphite blocks of different height, see Figure 3. The blocks are rectangular parallelepipeds, with a base of 0.25x0.25 m, and heights of 0.2, 0.3, 0.5 and 0.6 m of which the 0.6 m blocks are most common. The short blocks are used only in the top and bottom ends of the column, as required to provide a staggered fit to the neighbouring columns.
The blocks possess a 0.114 m diameter bore opening through the vertical axis. This provides a space for a total of 2052 channels which are used for placing Fuel Assemblies (FA) (so called Fuel Channels – FC), reactivity regulating control rods and several types of other instrumentation (so called Control and Protection System channels – CPS channels) into the core and cooling of radial reflector (so called Radial Reflector Cooling channels – RRC channels). The blocks of the remaining 436 columns located within the radial reflector have the central bore openings of 0.089 m diameter and this space is filled by graphite rods (6, Figure 4), providing keying of the blocks within the column, increasing the density and
neutron reflecting effectiveness of this part of the graphite stack. Graphite rods are manufactured from the different grade graphite compared to the blocks (GRP-1-280 is the grade of graphite used for rods while GR-280 is the grade used for blocks), however the total mass of the graphite rods in a whole graphite stack is much lower, i.e. ~60 tonnes.

**Figure 4: Segment of the reactor core with the part of graphite stack [2]**

Within the reactor core, FC and CPS channels are positioned by the help of special graphite rings and sleeves. In order to improve heat transfer from the graphite stack, the central segment of FC is surrounded by the 0.02 m height and 0.0115 m thickness split rings of GRP-2-125 grade graphite (17, Figure 2). 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. Above and below graphite rings section, there are also GRP-2-125 grade graphite sleeves of different shape and size placed. The arrangement of graphite rings and sleeves for CPS channels is somewhat different. The total mass of the graphite rings and sleeves of FC and CPS channels is ~120 tonnes in the reactor.

CPS channels could be equipped with control rods of 4 different types. Figure 5 presents schematic view of all types of control rods in their limiting positions in respect to the active core, i.e. fully inserted and fully withdrawn.

Figure 5: Layout of CPS rods position along the core of RBMK-1500 reactor [4]
Manual Control Rods of type 1 (MCR1) are control rods of initial design used in RBMK-1500 reactors to control the radial field of energy distribution. Later part of the MCR1 rods were replaced by the modified control rods (MCR2). Fast Acting Scram Rods (FASR) are used in the emergency situations to rapidly cease the nuclear chain reaction. Shortened Absorber Rods (SAR) are used to control the axial field of energy distribution, however in opposite to the other 3 types of rods, these are inserted to the active core from the bottom. Furthermore, some of CPS channels are equipped with the Axial Power Density Monitoring System sensors (PDMS-A) and Fission Chambers for reactivity monitoring purposes.

Although reactor graphite columns could be separated into different groups according to the various criteria, by considering the equipment placed inside the columns all 2488 pieces (pcs.) of reactor RBMK-1500 columns can be grouped in a structure as follows:

- Graphite columns with channel tubes (2052 pcs.), of which:
  - Fuel channel (1661 pcs.);
  - RRC channel (156 pcs.);
  - CPS channel (235 pcs.), of which:
    - containing MCR1 control rod (51 pcs.);
    - containing MCR2 control rod (96 pcs.);
    - containing FASR control rod (24 pcs.);
    - containing SAR control rod (40 pcs.);
    - containing PDMS-A sensor (20 pcs.);
    - containing Fission Chamber (4 pcs.).
- Graphite columns without channel tubes (436 pcs.), of which:
  - RR column containing graphite rods (436 pcs.).

The layout of the graphite columns in the RBMK-1500 reactor at Ignalina NPP Unit 1, based on the above-presented grouping structure, is shown in Figure 6.

It should be noted that there is a small amount of other graphite components present in the reactor core, like graphite bushings for temperature monitoring channels located in the corner of 4 neighbouring graphite columns or graphite displacers in the control rods, however their mass is much lower even than the mass of the RR graphite rods.
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Figure 6: Layout of graphite columns in RBMK-1500 reactor at Ignalina NPP Unit 1
2 C-14 inventory modelling

The methodology for modelling of C-14 inventory in the irradiated RBMK-1500 graphite for CAST project, in principle, is the same as used for EC 7th FP CARBOWASTE project [5, 6], i.e. initially, the energy and spatial distribution of neutron flux is modelled in the analysed system and later, the results of this modelling are used for neutron activation modelling. However, the modelling approach, developed models and computer codes used in the current study differ a lot compared to those of the CARBOWASTE project.

The approach user for CARBOWASTE project was based on the modelling of neutron flux using MCNP 5 ver. 1.2 code in one RBMK-1500 reactor cell containing FC (graphite column with constructions above and below it) with the model dimensions of 0.25x0.25x15 m. The modelled neutron flux was grouped into 3 energy groups (thermal, resonance and fast) and represented only neutron flux in the vicinity of plateau region of the reactor core for the graphite columns containing FC. The modelled thermal flux was then directly used for the graphite activation modelling using ORIGEN-S code from SCALE 5 codes system, while estimated resonance and fast neutron flux was used for determination of ORIGEN-S spectral parameters (updating of respective group cross-section data).

For the CAST project, modelling of the neutron flux was performed using MCNP 5 ver. 1.6 code [7]. The newly developed model envelopes the whole reactor core with the graphite stack consisting of 2488 graphite columns and surrounding structures, in the space region of 21x21x15 m (21x21 m is the length and the width of a concrete reactor vault while 15 m is the height of the modelled reactor core including surrounding structures). The modelled neutron flux was grouped into 238 energy groups (the most detailed neutron energy group structure used in SCALE codes system). The modelled neutron flux in these 238 groups structure then was used by COUPLE code from SCALE 6.1 codes system [8] for automated preparation of problem-specific cross-section data for ORIGEN-S code (also from SCALE 6.1 codes system). Having problem-specific cross-section data and using total neutron flux, the neutron activation modelling of the reactor RBMK-1500 graphite stack was performed.

The following subsections 2.1 and 2.2 of section 2 provide more information on the neutron flux and neutron activation modelling.
2.1 Neutron flux modelling

A general scheme of neutron flux modelling in the Ignalina NPP Unit 1 RBMK-1500 reactor graphite core used in this work is presented in Figure 7. The neutron flux modelling requires the input of reactor operation parameters, geometrics with specific material compositions, cross-sections for neutron transport and appropriate neutron transport modelling code.

Figure 7: General scheme of neutron flux modelling in RBMK-1500 reactor graphite core at Ignalina NPP Unit 1

The spatial and energy distribution of the neutron flux within the reactor RBMK-1500 graphite stack was modelled using MCNP 5 ver. 1.6 computer code [7], employing ENDF/B-VII nuclear data library (mainly). Numerous data of the RBMK-1500 reactor were used while developing the MCNP 5 model. These data were: geometrics (dimensions) of the reactor core internals and surroundings, compositions and densities of the materials, reactor operation parameters and others. For example, the axial water density profile in FC was accounted assuming that the water (for FC it is actually a water-steam mixture, to be precise) density is 0.75, 0.50 and 0.25 g/cm$^3$ below, in and above the active core respectively, while in the CPS and RRC channels it was assumed to be 1.00 g/cm$^3$. All the data were based on the information presented in technical documentation [2-4, 9]. The Ignalina NPP Unit 1 reactor RBMK-1500 model was developed assuming that:

- Every control rod in the CPS channel (of 211 CPS channels with control rods in total) is in its full withdrawn position (see Figure 5 for details);
Every FC (of 1661 FC in total) is loaded with FA containing UO\textsubscript{2} fuel of average burnup of \(~10\) MWd/kgU, with initial 2.4 % U-235 enrichment and 0.41 % burnable Er\textsubscript{2}O\textsubscript{3} absorber content.

The parameters of all reactor components defined in the model (temperatures, densities, dimensions, etc.) were set to match the conditions of the reactor working at the nominal 4200 MW thermal power generation capacity.

Vertical and horizontal cross-sections of the Ignalina NPP Unit 1 RBMK-1500 reactor model, developed with MCNP 5 code, are presented in Figure 8 and Figure 9 respectively.

This developed model of the reactor RBMK-1500 is suitable for modelling of the neutron flux in all graphite columns of the stack. However, this approach is still conservative, as all the control rods in the CPS channels are modelled in their full withdrawn positions, i.e. neutron flux in the active core is not affected by the presence of CPS control rod absorbing part, which is located just above (for SAR rods just below) the active core, and is even favoured by the presence of the control rod graphite displacers in the active core.

In order to evaluate the differences of neutron flux in the graphite columns containing different equipment and located in a different reactor part, the neutron flux was evaluated separately for the graphite columns:

- with fuel channel (580 pcs.) in the central part of the active core;
- with fuel channel (601 pcs.) in the 1\textsuperscript{st} peripheral part of the active core;
- with fuel channel (480 pcs.) in the 2\textsuperscript{nd} peripheral part of the active core;
- RR column containing graphite rods (436 pcs.);
- with RRC channel (156 pcs.);
- with CPS channel containing MCR1 control rod (51 pcs.);
- with CPS channel containing MCR2 control rod (96 pcs.);
- with CPS channel containing SAR control rod (40 pcs.);
- with CPS channel containing FASR control rod (24 pcs.);
- with CPS channel containing PDMS-A sensor (20 pcs.);
- with CPS channel containing fission chamber (4 pcs.).
Figure 8: Vertical cross-section of modelled RBMK-1500 reactor using MCNP
Figure 9: Horizontal cross-section of modelled RBMK-1500 reactor using MCNP
The obtained neutron flux in every group of graphite columns was averaged over the whole cross-section of columns in the lateral direction and over the heights of 20 segments in the axial direction. The heights of 14 segments in the active core were 50 cm each, while the heights of the remaining 6 segments (3 in the top and 3 in the bottom reflector region) were 16.67 cm, see Figure 10. In this way the spatial neutron flux distribution in the whole stack was obtained (11 groups of graphite columns divided into 20 segments in axial direction). These flux distributions then were used for the evaluation of the spatial distribution of the induced specific activity of C-14 in the graphite stack of the Ignalina NPP Unit 1 reactor.

![Figure 10: Segmentation of graphite column in axial direction](image)

The neutron flux modelled with the MCNP code was grouped into 238 energy groups structure. Such grouping of neutron flux was selected intentionally as a 238 neutron energy groups structure is the one for which the most detailed multigroup cross-section data in the SCALE codes package is available.
2.2 Neutron activation modelling

A general scheme of neutron activation modelling in the Ignalina NPP Unit 1 RBMK-1500 reactor graphite core used in this work is presented in Figure 11. The neutron activation modelling (C-14 specific activity modelling) requires the input of reactor operation history, cross-sections data and appropriate neutron activation modelling code.

The ORIGEN-S computer code from the SCALE 6.1 codes system [8] was used for the neutron activation modelling. The COUPLE code (also from SCALE 6.1 codes system) was used to collapse the multigroup cross-sections using the spectrum (in a 238 energy group structure) obtained from the MCNP modelling and to prepare the resulting weighted cross-sections to produce a binary ORIGEN-S library that is specific for the current problem.

As it has been previously discussed, the neutron activation and spatial distribution of induced activities were estimated for the graphite blocks (GR-280 grade graphite) of the RBMK-1500 reactor.

The main points of C-14 induced activity modelling in the Ignalina NPP Unit 1 reactor RBMK-1500 graphite stack were as follows:
Total modelled neutron flux was used for the activation modelling of GR-280 grade graphite blocks (11 groups of graphite columns divided into 20 segments in axial direction);

Modelled neutron flux spectrum (in a 238 energy groups structure) was used for the problem-dependent cross-section library production;

Ignalina NPP Unit 1 reactor RBMK-1500 operation history, presented in Figure 12, was used;

Decay period of 300 years was assumed and analysed;

GR-280 grade graphite composition with the maximal values of all impurities [5, 6] (including 70 ppm N) and naturally occurring isotopic compositions was assumed.

Figure 12: Ignalina NPP Unit 1 reactor RBMK-1500 operation history

In regard to the operation history of Unit 1 reactor (see Figure 12), it was assumed that for the first 17 periods (17 years), the reactor operates at different regimes continuously without shutdowns and that during each period the operation regime remains constant. The remaining 4 years of operation were divided into 25 shorter periods, taking into account all reactor shutdowns for more detailed representation.

It should be noted that activation modelling was performed only for reactor graphite stack, i.e. 2488 graphite columns consisting of GR-280 grade graphite bricks. Graphite rods in radial reflector and rings/sleeves of CPS and fuel channels were not analysed, however their quantity (mass) is much lower than that of graphite blocks, as was indicated before.
3 Modelling results

Neutron flux and neutron activation modelling results are presented in subsections 3.1–3.11 below. Although modelled neutron flux is only an intermediate result used for the estimation of induced C-14 activity in the irradiated graphite, presenting it gives an idea of the correlation between modelled flux and respective C-14 activity. The modelled flux represents reactor operation at 4200 MW thermal power. Furthermore, the neutron flux presented here was collapsed to the 3 energy groups from the 238 energy group structure, as initially modelled with MCNP:

- Thermal neutrons, with energies up to 0.625 eV;
- Resonance neutrons, with energies in range of 0.625 eV – 1 MeV;
- Fast neutrons, with energies above 1 MeV.

The grouping of neutrons into such energy groups was made only for the easier comparison of the modelled neutron flux between the different groups of graphite columns and the possibility of direct comparison with the results of the earlier estimations, e.g. [5, 6].

The modelled specific activities of C-14 are presented for 4 time periods, namely – at the time of Reactor Final Shutdown (RFS), 50 year after RFS, 150 year after RFS and 300 year after RFS. This timeline was chosen arbitrary; for C-14 such time period has minor impact as C-14 is a long-lived radionuclide, however for the other short-lived radionuclides that are out of scope of the CAST project, it allows to demonstrate activity decrease during graphite waste management period from RFS up to the initial phase after (possible) graphite disposal in a geological disposal facility.

Additionally, the presented modelled neutron flux, as well as C-14 specific activities, are normalized to the highest total neutron flux and C-14 activity, respectively. The absolute values of the modelled neutron flux and C-14 specific activity are presented only for the graphite columns containing FC from the central part of the reactor, where maximal flux and activity is obtained. For the rest of the graphite column groups, only the normalised distributions are presented, allowing easy comparison of the modelled results in respect to the graphite columns containing FC from the central part of the reactor (maximal results).
For convenience, the centre of the graphite stack (column) is assumed to be at zero mark and the axial distance from the centre of the graphite stack is given in the ordinate axis.

### 3.1 Graphite columns containing fuel channels in reactor centre

The results of the neutron flux and C-14 specific activity modelling in the graphite columns with FC located in the central part of the reactor (see Figure 13), are presented respectively in Figure 14 and Figure 15.

**Figure 13:** Layout of graphite columns containing fuel channels in RBMK-1500 reactor central part at Ignalina NPP Unit 1
The graphite columns with FC are regularly distributed over the whole cross-section of the central part of the active core of the reactor, as given in Figure 13. These FC contain a fuel assembly that consists of two fuel bundles. The fuel assemblies were explicitly described in the MCNP model, so this accordingly affected the neutron flux distribution, see Figure 14.

The results of the neutron flux modelling show that the thermal and resonance neutron flux are dominant in the graphite column (blocks) with FC in the central reactor part; however, the thermal neutron flux is more intensive than the resonance neutron flux (though very insignificantly in a certain regions), see Figure 14 (Figure 14 a) shows the modelled neutron flux distribution in the absolute values, whereas Figure 14 b) shows the modelled neutron flux distribution in the relative units, i.e. normalized to the maximal value of the total modelled neutron flux).

The fast neutron flux is about 10 times lower than the resonance neutron flux and its distribution along the axial direction is almost the same as that of the resonance neutron flux (see Figure 14). There are two maximums in the distribution profiles of the fast and resonance neutron flux and one minimum in the area of the reactor core axial centre, which corresponds to the place of the fuel bundles connection in the fuel assembly. In the edges of graphite column (i.e. top and bottom reflector blocks) the fast neutron flux is ~150 (top reflector) and ~40 (bottom reflector) times lower than the maximal value of the fast neutron flux in the region of the active core (i.e. in the 700 cm height central part of the graphite blocks column). For the resonance neutron flux the differences are lower; the resonance neutron flux is ~100 (top reflector) and ~30 (bottom reflector) times lower than the maximal value of the resonance neutron flux in the region of the active core.

In the case of the thermal neutron flux, there is only one maximum in the neutron flux distribution profile and it is located in the central (lower) part of the reactor core (see Figure 14). Going further from that location the thermal flux decreases monotonically and in the edges of the graphite column i.e. top and bottom reflectors is ~24 and ~7 times lower than the maximal flux, respectively. The average (averaged over the 700 cm long central part of the graphite column) thermal neutron flux in the graphite column is ~1.2×10^{14} n/cm^{2}\cdot s while the maximal value of the thermal flux is nearly 2×10^{14} n/cm^{2}\cdot s.
Figure 14: Distribution of absolute a) and normalised b) neutron flux in graphite columns containing fuel channels in RBMK-1500 reactor central part at Ignalina NPP Unit 1
C-14 specific activity modelling results are presented in Figure 15 (Figure 15 a) shows the modelled C-14 specific activity distribution in the absolute values, whereas Figure 15 b) shows the modelled C-14 specific activity distribution in the relative units, i.e. normalized to the maximal value of the modelled C-14 specific activity). The modelling results show that C-14 specific activity distribution along the axial direction in the graphite columns containing fuel channels corresponds to the thermal neutron flux distribution (see Figure 14 and Figure 15). The average C-14 activity in the active core graphite blocks (i.e. average in the 700 cm height central part of the graphite blocks column) is ~5.4×10^5 Bq/g. The maximal C-14 activity (~8.7×10^5 Bq/g) position coincides with the maximal thermal neutron flux position, which is at mark “–125”, and (analogous to the thermal flux) the C-14 specific activity in the top and bottom reflector parts is respectively ~24 and ~7 times lower than the maximal activity. This confirms the fact that production of C-14 from carbon activation, as well as from impurities activation, is determined mainly by the thermal neutron flux.

The influence of the radioactive decay to the induced activity of C-14 during the analysed 300 y period after RFS in insignificant. As C-14 is a long-lived radionuclide having the half-life of ~5.7×10^3 y, during the 300 y period its activity decreases less than 4 percent.

Comparing C-14 specific activity modelling results of the current study with the ones obtained previously in EC 7th FP CARBOWASTE project [5, 6] (max. impurities case), it could be noted that the results are somewhat different. The average activity of the active core graphite blocks obtained in [5, 6] was ~3.6×10^5 Bq/g (~5.4×10^5 Bq/g here), the activity in the bottom reflector was just above 8×10^4 Bq/g (1.3×10^5 Bq/g here) and in the top reflector it was nearly 6×10^4 Bq/g (3.7×10^4 Bq/g here). This gives a clear indication that, on the whole, the results of the current study are higher, but in the upper region of graphite stack, the modelled activities are lower. This means that the distribution profiles of the modelled neutron flux and, consequently, C-14 activities have changed. The main reason for this is the full description of the RBMK-1500 reactor and its surrounding structures in this study (especially control rods in their withdrawn positions), thus representing their influence to the modelled neutron flux. Additionally, activation modelling (with the use of COUPLE module for cross-sections preparation and updated data libraries) had an influence.
Figure 15: Distribution of absolute a) and normalised b) C-14 specific activity in graphite columns containing fuel channels in RBMK-1500 reactor central part at Ignalina NPP Unit 1
3.2 **Graphite columns containing fuel channels in reactor 1st periphery**

The results of the neutron flux and C-14 specific activity modelling in the graphite columns with FC located in the 1st peripheral part of the reactor (see Figure 16) are presented respectively in Figure 17 and Figure 18.

**Figure 16: Layout of graphite columns containing fuel channels in RBMK-1500 reactor first peripheral part at Ignalina NPP Unit 1**

Comparing Figure 17 with Figure 14 b) it can be seen that the distribution profiles of the neutron flux of all energy groups for the graphite columns with FC in the 1st peripheral part
are analogous to those of the central reactor part. In the active zone area, the thermal and resonance neutron flux is dominant, the second being slightly lower, while the fast neutron flux is approximately one order of magnitude lower than the thermal flux. In the regions of the top and bottom reflectors, the thermal neutron flux is dominant with the resonance flux being approximately one order of magnitude lower than the thermal flux and the fast flux being approximately one order of magnitude lower than the resonance flux. In absolute numbers, the neutron flux of each analysed energy group in the graphite columns containing fuel channels in the 1st peripheral part of the reactor are ~1.7 times lower compared to those located in the central reactor part, see Figure 17 and Figure 14 b.

As the induced activity of C-14 (Figure 18) directly corresponds to the thermal neutron flux (Figure 17), the changes in the thermal flux directly affects the changes in the induced C-14 activity. Thus it is obvious that, comparing Figure 18 with Figure 15 b), it can be seen that the distribution profiles of C-14 specific activity for the graphite columns with FC in the 1st peripheral part are analogous to those of the central part, but are lower by ~1.7 times.
3.3 Graphite columns containing fuel channels in reactor 2\textsuperscript{nd} periphery

The results of the neutron flux and C-14 specific activity modelling in the graphite columns with FC located in the 2\textsuperscript{nd} peripheral part of the reactor (see Figure 19) are presented respectively in Figure 20 and Figure 21.

Comparing Figure 20 with Figure 14 b) and Figure 17 it can be seen that the distribution profiles of the neutron flux of each analysed energy group for the graphite columns with FC in the 2\textsuperscript{nd} peripheral part are very similar to those of the central and 1\textsuperscript{st} peripheral parts. However, the distribution profile has some minor differences mainly due to the fact that graphite columns with FC in the 2\textsuperscript{nd} peripheral part are surrounded by the radial reflector zone, where no fuel, i.e. no neutron source, is present. This also affects the absolute values of neutron flux of all energy groups, resulting in the neutron flux decrease of \approx 3.5 times compared to those of the central reactor part (or decrease of \approx 2 times compared to those of 1\textsuperscript{st} peripheral reactor part).
Figure 19: Layout of graphite columns containing fuel channels in RBMK-1500 reactor second peripheral part at Ignalina NPP Unit 1

As the induced activity of C-14 (Figure 21) directly corresponds to the thermal neutron flux (Figure 20), the changes in the thermal flux directly affects the changes in the induced C-14 activity. Thus it is obvious that, comparing Figure 21 with Figure 15 b) and Figure 18, it can be seen that the distribution profiles of C-14 specific activity for the graphite columns with FC in the 1st peripheral part are very similar to those of the central reactor part, but are lower by ~3.5 times (or to those of 1st peripheral reactor part, but being lower by ~2 times).
Figure 20: Distribution of neutron flux in graphite columns containing fuel channels in RBMK-1500 reactor second peripheral part at Ignalina NPP Unit 1

Figure 21: Distribution of C-14 specific activity in graphite columns containing fuel channels in RBMK-1500 reactor second peripheral part at Ignalina NPP Unit 1
3.4 Graphite columns containing graphite rods

The results of the neutron flux and C-14 specific activity modelling in the graphite columns with graphite rods, i.e. RR columns (see Figure 22) are presented respectively in Figure 23 and Figure 24.

Figure 22: Layout of RR graphite columns containing graphite rods in RBMK-1500 reactor at Ignalina NPP Unit 1

Results presented in Figure 23 show that the thermal neutron flux is dominant in the graphite columns of RR, being nearly 1 order of magnitude higher than the resonance flux.
The fast flux is the lowest and is slightly more than 2 orders of magnitude lower than the thermal flux. The distribution profiles of the fast and resonance flux are similar to those of the graphite columns with FC, e.g. from the central reactor part (see Figure 14 b). The thermal flux in the columns of RR is ~6.7 times lower than the thermal flux in graphite columns with FC from the central part (see Figure 23 and Figure 14 b) and the distribution profile has no peak in the area of the axial centre (zero mark).

The difference of the thermal flux is directly reflected in the induced activities of C-14, i.e. the distribution profile of the induced specific activities of C-14 in the RR graphite columns (see Figure 24) corresponds to the distribution of the thermal neutron flux (see Figure 23), and the specific activities of C-14 are on average ~6.7 times lower than those of the graphite columns with FC in the central reactor part (Figure 15 b).
3.5 Graphite columns containing RRC channels

The results of the neutron flux and C-14 specific activity modelling in the graphite columns containing RRC channels (see Figure 25) are presented respectively in Figure 26 and Figure 27.

The RRC graphite columns are the outer columns of the reactor (Figure 25) and are situated furthest away from the region with the fuel. This is directly reflected in the modelling results of the neutron flux, as presented in Figure 26. The thermal neutron flux is dominant, being ~2 orders of magnitude higher than the resonance and more than 3 orders of magnitude higher than the fast neutron flux.

The distribution profile of the neutron flux is similar to those of RR graphite columns, see Figure 23. In comparison with the results of the graphite columns containing FC in the central part (see Figure 14 b), the total neutron flux in the graphite columns containing RRC channels is ~2 orders of magnitude lower while the thermal neutron flux is ~50 times lower.
The analogy of differences in the thermal flux is retained and present in the modelled induced activities of C-14. The distribution profile of the induced specific activities of C-14 in the graphite columns containing RRC channels (see Figure 27) corresponds to the distribution of the thermal neutron flux (see Figure 25), and the specific activities of C-14 are on average ~50 times lower than those of the graphite columns with FC in the central reactor part (see Figure 15 b).
Figure 26: Distribution of neutron flux in graphite columns containing RRC channels in RBMK-1500 reactor at Ignalina NPP Unit 1

Figure 27: Distribution of C-14 specific activity in graphite columns containing RRC channels in RBMK-1500 reactor at Ignalina NPP Unit 1
3.6 Graphite columns containing CPS channels with MCR1

The results of the neutron flux and C-14 specific activity modelling in the graphite columns with CPS channels for MCR1 rods (see Figure 28) are presented respectively in Figure 29 and Figure 30.

**Figure 28: Layout of graphite columns containing CPS channels (MCR1) in RBMK-1500 reactor at Ignalina NPP Unit 1**

The graphite columns with CPS channels containing MCR1 type rods are distributed over the whole cross-section of the active core of the reactor, as presented in Figure 28. As these
CPS channels contain no fuel and their control rods are modelled in the full withdrawn position (i.e. neutron absorbing part is located directly above the active zone of the graphite stack, thus the graphite displacer part of the rod being positioned in the lower part of the active zone), this accordingly affects the neutron flux distribution, see Figure 29. The thermal neutron flux is dominant and it is ~2 times higher than the resonance flux in the active part of the core. In the top and bottom reflectors this difference is even higher – ~7 and ~10 times respectively. The resonance flux is the lowest and is more than 1 order of magnitude lower than the resonance.

![Figure 29: Distribution of neutron flux in graphite columns containing CPS channels (MCR1) in RBMK-1500 reactor at Ignalina NPP Unit 1](image)

The distribution profile of the induced specific activities of C-14 in the graphite columns of CPS channels with MCR1 rods (see Figure 30) corresponds to the distribution of the thermal neutron flux (see Figure 29). In the active part of the core, the specific activities of C-14 are on average ~1.2 times lower than those of the graphite columns with FC in the central part (see Figure 15 b). In the top and bottom reflectors this difference is higher – ~2 and ~1.4 times respectively. These differences exactly correspond to the differences of
thermal neutron flux in the graphite columns of CPS channels with MCR1 rods (see Figure 29) and in the graphite columns with FC in the central reactor part (see Figure 14 b).

![Graph showing distribution of C-14 specific activity](image)

**Figure 30: Distribution of C-14 specific activity in graphite columns containing CPS channels (MCR1) in RBMK-1500 reactor at Ignalina NPP Unit 1**

### 3.7 Graphite columns containing CPS channels with MCR2

The results of the neutron flux and C-14 specific activity modelling in the graphite columns with CPS channels for MCR2 rods (see Figure 31) are presented respectively in Figure 32 and Figure 33.

The graphite columns with CPS channels containing MCR2 type rods are quite homogeneously distributed over the whole cross-section of the active core of the reactor, as given in Figure 31. These CPS channels contain no fuel and their control rods are modelled in the full withdrawn position (i.e. neutron absorbing part is located directly above the active zone of the graphite stack and the graphite displacer part of the rod extends over the whole height of the active zone), so this accordingly affects the neutron flux distribution, see Figure 32. As MCR1 and MCR2 control rods are of similar design, the neutron flux
modelling results for the graphite columns with CPS channels containing MCR1 and MCR2 type control rods are also very close, see Figure 29 and Figure 32.

The distribution profile of the induced specific activities of C-14 in the graphite columns of CPS channels with MCR2 rods (see Figure 33) corresponds to the distribution of thermal neutron flux there (see Figure 32), and is almost identical (also in absolute values) to the distribution of C-14 specific activity in the graphite columns of CPS channels with MCR1 rods (see Figure 30).

![Figure 31: Layout of graphite columns containing CPS channels (MCR2) in RBMK-1500 reactor at Ignalina NPP Unit 1](image-url)
Figure 32: Distribution of neutron flux in graphite columns containing CPS channels (MCR2) in RBMK-1500 reactor at Ignalina NPP Unit 1

Figure 33: Distribution of C-14 specific activity in graphite columns containing CPS channels (MCR2) in RBMK-1500 reactor at Ignalina NPP Unit 1
3.8 Graphite columns containing CPS channels with SAR

The results of the neutron flux and C-14 specific activity modelling in the graphite columns with CPS channels for SAR rods (see Figure 34) are presented respectively in Figure 35 and Figure 36.

Figure 34: Layout of graphite columns containing CPS channels (SAR) in RBMK-1500 reactor at Ignalina NPP Unit 1

The graphite columns with CPS channels containing SAR type rods are distributed on a quite regular grid over the whole cross-section of the active core of the reactor, as shown in
Figure 34. These CPS channels contain no fuel and their control rods are modelled in the full withdrawn position (i.e. neutron absorbing part is located directly below the active zone of the graphite stack (slightly entering it) and the graphite displacer part of the rod extends over the rest height of the active zone, as these rods enter the active core in the upward direction), so this accordingly affects the neutron flux distribution, see Figure 35.

![Graph showing neutron flux distribution](image)

**Figure 35: Distribution of neutron flux in graphite columns containing CPS channels (SAR) in RBMK-1500 reactor at Ignalina NPP Unit 1**

The thermal neutron flux is dominant and it is ~2 times higher than the resonance flux in the active part of the core. In the top and bottom reflectors this difference is even higher – ~9 and ~7 times respectively. The resonance flux is the lowest and is more than 1 order of magnitude lower than the resonance flux.

The distribution profile of the induced specific activities of C-14 in the graphite columns of CPS channels with SAR rods (see Figure 36) corresponds to the distribution of thermal neutron flux (see Figure 35). In the active part of the core and top reflector, the specific activities of C-14 are on average ~1.2 times lower than those of the graphite columns with FC in the central reactor part (see Figure 15 b). In the bottom reflector this difference is
higher – it reaches ~2.4 times. These differences exactly correspond to the differences of the thermal neutron flux in the graphite columns of CPS channels with SAR rods (see Figure 35) and in the graphite columns with FC in the central reactor part (see Figure 14 b).

![Graphite columns with CPS channels containing FASR rods](image)

Figure 36: Distribution of C-14 specific activity in graphite columns containing CPS channels (SAR) in RBMK-1500 reactor at Ignalina NPP Unit 1

### 3.9 Graphite columns containing CPS channels with FASR

The results of the neutron flux and C-14 specific activity modelling in the graphite columns with CPS channels for FASR rods (see Figure 37) are presented respectively in Figure 38 and Figure 39.

The graphite columns with CPS channels containing FASR type rods are distributed on a regular grid over the whole cross-section of the active core of the reactor, as shown in Figure 37. These CPS channels contain no fuel and their control rods are modelled in the full withdrawn position (i.e. neutron absorbing rod is located directly above the active zone of the graphite stack, providing empty space inside the active zone), and this accordingly affects the neutron flux distribution, see Figure 37.
The thermal neutron flux is dominant in the graphite columns of CPS channels with FASR rods and it is nearly 2 times higher than the resonance flux in the active part of the core (see Figure 38). In the top and bottom reflectors this difference is even higher – ~5 and more than 7 times respectively. The resonance flux is the lowest and is more than 1 order of magnitude lower than the resonance flux.
Figure 38: Distribution of neutron flux in graphite columns containing CPS channels (FASR) in RBMK-1500 reactor at Ignalina NPP Unit 1

Figure 39: Distribution of C-14 specific activity in graphite columns containing CPS channels (FASR) in RBMK-1500 reactor at Ignalina NPP Unit 1
The distribution profile of the induced specific activities of C-14 in the graphite columns of CPS channels with FASR rods (see Figure 39) corresponds to the distribution of thermal neutron flux (see Figure 38). In the active part of the core and bottom reflector, the specific activities of C-14 are on average ~1.2 times lower than those of the graphite columns with FC in the central reactor part (see Figure 15 b). In the top reflector this difference is higher – it reaches ~2 times. These differences equal the differences of the thermal neutron flux in the graphite columns of CPS channels with FASR rods (see Figure 38) and in the graphite columns with FC in the central reactor part (see Figure 14 b).

3.10 Graphite columns containing CPS channels with PDMS-A

The results of the neutron flux and C-14 specific activity modelling in the graphite columns with CPS channels for PDMS-A sensors (see Figure 40) are presented respectively in Figure 41 and Figure 42.

The graphite columns with CPS channels dedicated for PDMS-A sensors are distributed on a regular grid over the whole cross-section of the active core of the reactor (12 pcs.), and in the first row or the radial reflector region (8 pcs), as shown in Figure 40. These CPS channels contain no fuel and no control rod, however the suspension bracket for in-core power density sensor of axial monitoring (PDMS-A) is located inside. This steel bracket is described in the MCNP model for the neutron flux estimation and accordingly affects the modelled neutron flux distribution, see Figure 41.

The thermal neutron flux is dominant in the graphite columns of CPS channels for PDMS-A sensors and it is nearly 2 times higher than the resonance flux in the active part of the core (see Figure 41). In the top and bottom reflectors this difference is even higher – ~9 times. The resonance flux is the lowest and is more than 1 order of magnitude lower than the resonance. These differences of the thermal, resonance and fast flux are similar to those of the graphite columns containing CPS channels with FASR and MCR2 control rods, see Figure 38 and Figure 32 respectively.

The distribution profile of the induced specific activities of C-14 in the graphite columns of CPS channels for PDMS-A sensors (see Figure 42) corresponds to the distribution of
thermal neutron flux (see Figure 41). In the active part of the core, the specific activities of C-14 are on average nearly 2 times lower than those of the graphite columns with FC in the central reactor part (see Figure 15 b). In the top and bottom reflectors this difference is slightly higher by a factor just above 2 times. These differences exactly correspond to the differences of the thermal neutron flux in the graphite columns of CPS channels with PDMS-A sensors (see Figure 41) and in the graphite columns with FC in the central reactor part (see Figure 14 b).

Figure 40: Layout of graphite columns containing CPS channels (PDMS-A) in RBMK-1500 reactor at Ignalina NPP Unit 1
Figure 41: Distribution of neutron flux in graphite columns containing CPS channels (PDMS-A) in RBMK-1500 reactor at Ignalina NPP Unit 1

Figure 42: Distribution of C-14 specific activity in graphite columns containing CPS channels (PDMS-A) in RBMK-1500 reactor at Ignalina NPP Unit 1
3.11 Graphite columns containing CPS channels with fission chambers

The results of the neutron flux and C-14 specific activity modelling in the graphite columns with CPS channels for fission chambers (see Figure 43) are presented respectively in Figure 44 and Figure 45.

![Figure 43: Layout of graphite columns containing CPS channels (fission chambers) in RBMK-1500 reactor at Ignalina NPP Unit 1](image)

The graphite columns with CPS channels for fission chambers are distributed in the first row of the radial reflector region, as shown in Figure 43. These CPS channels contain no
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fuel and no control rod, however the suspension bracket for fission chamber is located inside. This steel bracket is described in the MCNP model for the neutron flux estimation and accordingly affects the modelled neutron flux distribution, see Figure 44.

![Graph showing neutron flux distribution](image)

**Figure 44**: Distribution of neutron flux in graphite columns containing CPS channels (fission chambers) in RBMK-1500 reactor at Ignalina NPP Unit 1

The thermal neutron flux is dominant in the graphite columns of CPS channels for fission chambers and it is ~3 times higher than the resonance flux in the active part of the core (see Figure 44). In the top and bottom reflectors this difference is higher – by more than 1 order of magnitude. The resonance flux is the lowest and is by more than 1 order of magnitude lower than the resonance flux.

The distribution profile of the induced specific activities of C-14 in the graphite columns of CPS channels for fission chambers (see Figure 45) corresponds to the distribution of the thermal neutron flux (see Figure 44). In the active part of the core and top reflector, the specific activities of C-14 are on average ~5 times lower than those of the graphite columns with FC in the central reactor part (see Figure 15 b). In the bottom reflector this difference is slightly higher by just above 6 times. These differences correspond to the differences of the
thermal neutron flux in the graphite columns of CPS channels with fission chambers (see Figure 45) and in the graphite columns with FC in the central part (see Figure 14 b).

![Graph showing distribution of C-14 specific activity](image)

**Figure 45:** Distribution of C-14 specific activity in graphite columns containing CPS channels (fission chambers) in RBMK-1500 reactor at Ignalina NPP Unit 1

### 4 Estimation of C-14 inventory

As it was already mentioned, the aim of this work was to estimate C-14 inventory in the graphite stack of the Ignalina NPP Unit 1 reactor combining C-14 activity modelling and measurement results.

Sampling of the Ignalina NPP Unit 1 RBMK-1500 reactor graphite stack has been already done by the Ignalina NPP staff and it was expected that radiological characterisation of the taken samples (activity of C-14 is of most importance for CAST project) will be made in the course of the CAST project. However, to the date of issue of this report (D5.17), no results of C-14 activity measurements in GR-280 grade graphite were made publically available, except in a paper [10]. This paper presented C-14 activity measurements in one sample (see Figure 46 for location) of the graphite bushing from the temperature channel of the reactor.
Consequently, the exhaustive combination of the modelled and measured C-14 activities for the more detailed and more confident estimation of C-14 inventory in the whole RBMK-1500 reactor graphite stack has not been achieved to date, as the only C-14 activity measurement data that could be used for the inventory estimation was that from a paper [10]. This paper presented preliminary studies of the experimental C-14 activity estimation in several graphite sub-samples, attributed to the same sample of the GR-280 grade graphite bushing from the temperature channel of the Ignalina NPP Unit 1 RBMK-1500 reactor. Based on the statistical analysis of the sub-samples activity, the average value of the C-14 specific activity in this graphite sample was $1.67 \times 10^5$ Bq/g [10]. According to the information on the location of the analysed sample [10], the sample was taken from the area of graphite columns with fuel channels in the RBMK-1500 reactor first peripheral part (see Figure 46: Position of graphite sample taken from the area of graphite columns in RBMK-1500 reactor first peripheral part at Ignalina NPP Unit 1).
Figure 46) and the measured specific activity of $1.67 \times 10^5$ Bq/g corresponds to the point at +175 cm elevation of the modelled specific activity distribution in the first peripheral part of the reactor (see Figure 18), as marked in Figure 47. This gives that the modelled specific activity of C-14 (at +175 cm elevation in the first peripheral part of the reactor) is 0.258 relative units (see Figure 47) and corresponds to the measured specific activity of $1.67 \times 10^5$ Bq/g in that location.

![Figure 47: Indication of measured C-14 specific activity in the distribution of modelled C-14 specific activities](image)

By having the modelled distributions of the specific C-14 activity in each group of the graphite columns in relative units (see Figure 15, Figure 18, Figure 21, Figure 24, Figure 27, Figure 30, Figure 33, Figure 36, Figure 39, Figure 42, Figure 45), by knowing the correspondence of the measured specific C-14 activity to the exact point (location) of the modelled specific activity distribution (see Figure 47), and by applying quantities and masses of the graphite columns in the respective group (see subsection 1.2), the integral C-14 activity in the graphite stack was calculated.
As a final emphasis of this report it could be stated, that combining C-14 activity modelling and measurement results, the following numbers regarding C-14 inventory in the irradiated graphite stack of the Ignalina NPP Unit 1 RBMK-1500 reactor at the time of RFS were obtained:

- Total C-14 activity in the graphite stack: \(3.222 \times 10^{14}\) Bq;
- Total mass of the graphite stack: \(1.700 \times 10^9\) g (1700 t);
- Average C-14 activity in the graphite stack: \(1.895 \times 10^5\) Bq/g.

However, it should be also noted that the above numbers are only for the reactor RBMK-1500 graphite stack consisting of GR-280 grade graphite blocks. The remaining graphite components in the reactor (graphite rings, sleeves, rods and other graphite parts in minor quantities) are not taken into account. For these parts, as a first attempt, the same average specific activity could be applied, which together with the mass of ~200 t results in \(3.790 \times 10^{13}\) Bq of C-14. Finally, this sums to \(3.601 \times 10^{14}\) Bq of C-14 in ~1900 t of the irradiated graphite of all types in one RBMK-1500 reactor.

Additional caution should be paid to the above presented estimation of the total and the average specific activity of C-14 in Ignalina NPP Unit 1 reactor RBMK-1500 graphite, as the estimation performed relays only on the preliminary measurements of only one graphite sample. It could not be totally excluded that the measurements of the sample are somehow misleading, sample is not representative, etc. Anyhow, these are the only officially published experimental measurement results of C-14 activity in the GR-280 grade graphite of Ignalina NPP Unit 1 reactor and they were used in the study presented in this report.

References

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