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# CARBOWASTE

***Treatment and Disposal of Irradiated Graphite and Other Carbonaceous Waste***

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# WP 1 Review Report - Russia

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| Integrated Waste Management Approach WP 1 Review Report - Russia |
| **Executive summary** |

In total 15 water-cooled graphite-moderated power reactors (11 RBMK-1000 and 4 EGP-6 reactors are being operated in Russia. Three first-generation water-cooled graphite-moderated reactors (AM reactor in Obninsk and 2 AMB reactors at Beloyarsk NPP) are permanently shut down. In 2006, Rosatom informed it was considering lifetime extensions and upgrading of its eleven operating RBMK reactors. Following significant design modifications made after the Chernobyl accident, as well as extensive refurbishment including replacement of fuel channels, a 45-year lifetime is seen as realistic for the 1000 MWe units. The Leningrad Nuclear Power Plant is the oldest one. The reactors were put in operation between 1974 and 1981. The oldest reactor received a 15 year prolongation its engineered life span in 2004. It is assumed that the best strategy for handling graphite during decommissioning of the power-generating units of a nuclear power plant with uranium-graphite reactors is localization and long-term storage of the graphite masonry together with the reactor in a concrete shaft at the plant site under surveillance. By the time when dismantling is required an ecologically acceptable and cost-effective technology for handling irradiated graphite reactors will be available. In 1998 the unique experiment on withdrawing of the graphite column from reactor RBMK-1000 of Leningrad NPP Unit 3 has been performed. The graphite column (all 14 graphite bricks) was withdrawn from the cell 12-36 after 18 years of reactor operation for long scale investigation of graphite brick (GB) behavior and estimation of methods of graphite core dismantling
13 high-capacity weapons plutonium production (industrial) reactors were in operation in Russia.
Decommissioning concept of these reactors was developed in the middle of 1990s, comprising the dismantling of low-activity structures, sealing of all reactor outlets, and filling of all reactor spaces with special compounds of concrete and bentonite. All these arrangements ensured multi-level protection with a number of safety barriers between the reactor and the environment. At that time a question about the possible reactor dismantling was not raised. The reactor shaft was intended to be transformed into the everlasting radwaste repository. In-shaft waste disposal concept came into conflict with new waste disposal regulatory documents (NP-055-04, NP-069-06, GOST R 50996-96). So, later-on instead of disposal of the reactor shaft the monitored safe storage period was defined. So, the general scheme of decommissioning of these reactors ("storage" on site) is:

- Preparatory period – reloading of the SNF;
- Localization of the stack in the shaft with additional protective engineering barriers;
- Period of the long-term storage (100 and more years) – monitoring and supervision of the reactor;
- Possibility for the dismantling of the reactor after long-term storage.

The D&D concept 'termination on site' for uranium-graphite reactors has been implemented in the Russian Federation for the I1, E2 and ADE3 reactors, with the capability to dismantle the rest of the equipment at the same time. A system of protective barriers (multi-level protection) is being developed in order to avoid environmental pollution from disposed radioactive waste and to protect the other reactor components from external natural or technical influences. A special monitoring system has been provided for the inspection of storage parameters, including measurements of temperature, humidity, gas composition, radioactivity, and location of support metal structures. A system of special environmental drill holes is provided on the site for inspection of the conditions. Unique technologies and equipment were developed for dismantling some of the structures and for the safe and successful performance of all works. As a result, the suggested D&D concept allowed qualification of the structure under the IAEA's Stage 2 and ensured its safety for over 300 years.
Results of rather detailed experimental investigations of the radioactive contamination of graphite masonry in the commercial reactors at the Siberian chemical combine, Mayak were obtained:

- About 500 samples have been taken from the graphite stack of the I-1, ADE-3 and EI-2 reactors and assayed. Contamination of these samples with radionuclides H-3, C-14, Cl-36, Co-60, Ni-63, Sr-90, Ba-133, Cs-134, 137, Eu-152, 154, 155, Pu-238, Pu-239, 240, Am-241, 243, Cm-244 and some others have been determined;
- It was discovered in this study that the dominant activity in the graphite is C-14, and its distribution in the graphite stack is a reflection of the thermal neutron flux. The concentration of C-14 in the graphite from this Tomsk reactor was about 6 times higher then in the graphite from a similar Hanford reactor. A significant fraction of the C-14 activity in reactor graphite was indeed due to the presence of nitrogen in graphite nitrogen cover gas);
- The tritium content in the reactor graphite has been measured, and the content appeared to be very small (about several hundred times smaller) than prediction. H-3 is distributed non-uniformly in the graphite stack;
- The dominant fraction of the Co-60 concentration in reactor graphite is due to the presence of original Co-59 impurity in graphite;

As it was indicated already the concept of the storage of the graphite masonry of the graphite reactors in situ isolated from the environment for about of 100 years is adopted in Russia. In relation with this, development of the technologies for the treatment of the irradiated graphite was not the primary task. So, today there is not industrially tested technology for the treatment of the irradiated graphite in Russia. But there is a problem of the management so called graphite sleeves that are under storage in different storage facilities at the reactor sites. Also during maintenance period's significant amounts of the graphite from graphite masonry were retrieved and replaced with new ones. There are three main directions in the development of the technologies for management of the irradiated graphite in Russia:

- Methods to establish engineering barriers for the long-term storage of graphite in the form of solid radwaste (storage of irradiated graphite in special containers);
- Methods of graphite chemical transformation for the purpose of the separation of the dose-generating isotopes and the reduction of the volumes for the further storage;
- Treatment of the irradiated graphite;
- Incineration of the irradiated graphite.

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# Chapter contents

# 1 Introduction

This chapter addresses the graphite associated with 15 water cooled graphite moderated power reactors and 13 reactors for plutonium production. Eleven of the former reactors are currently being considered for life-time extensions.

Decommissioning concepts for the plutonium production reactors was developed in the 1990s but has undergone revision to accommodate new waste disposal legislation.

The chapter discusses the graphite characteristics based on numerous analytical results encompassing many key parameters. The final sections describe i-graphite teatment/conditioning techniques to underpin interim storage and/or for final disposal.

# 2 The Legislative and Regulatory Framework

There are separate safety regulations for decommissioning of different types of nuclear installations in Russia [1]:

1. Safety regulations during decommissioning of industrial reactors (NP-007-98), Gosatomnadzor of Russia, 1998 (in Russian).
2. Safety regulations during decommissioning of the unit of nuclear power plant (NP-012-99), Gosatomnadzor of Russia, 1999 (in Russian).
3. Safety regulations during decommissioning of research nuclear installations (NP-028-01), Gosatomnadzor of Russia, 2001 (in Russian).
4. Safety regulations during decommissioning of nuclear power installations on ships (NP-037-02), Gosatomnadzor of Russia, 2002 (in Russian).
5. Safety regulations during decommissioning of nuclear fuel cycle facilities (NP-057-04), Rostechnadzor, 2004 (in Russian).
6. Requirements to the scope and content of the documents for demonstration of nuclear and radiation safety during operation and decommissioning of nuclear ships and other floating items with nuclear installations and radioactive sources (RD-06-14-97), Rostechnadzor, 2004 (in Russian).

Following the safety regulations for decommissioning of industrial reactors (NP-007-98), a comprehensive engineering and radiation examination of these reactors is now being conducted. These regulations require for, among other things, the extraction and analysis of graphite samples for determining the activity of long-lived fission products in the graphite masonry contaminated with fuel elements from damaged channels as a result of accidents during reactor operation.

# 3 Policy Objectives of Decommissioning

## 3.1 Policy of nuclear reactors decommissioning

### 3.1.1 Power reactors

A list of commercial NPP's with power reactors in Russia is presented in Table 1. Presently, there are 10 operating nuclear power plants (31 operating reactor) in Russia with most them located in the European part of the country. They produce 17% (total 21,743 MWe) of the electricity generated in the country. Concern Rosenergoatom is the only one operating organization in Russia now. It incorporates all Russian nuclear power plants [2, 3].

**Table 1. Power Reactors in Operation [4]**

| Reactor | Type V=PWR | MWe net, each | Commercial operation | Scheduled close |
|---|---|---|---|---|
| **Balakovo 1-2** | V-320 | 950 | 5/86, 1/88 | 2015, 2017 |
| **Balakovo 3-4** | V-320 | 950 | 4/89, 12/93 | 2018, 2023 |
| **Beloyarsk 3** | BN600 FBR | 560 | 11/81 | 2010 |
| **Bilibino 1-4** | LWGR EGP-6 | 11 | 4/74-1/77 | 2009, 09, 11, 12 |
| **Kalinin 1-2** | V-338 | 950 | 6/85, 3/87 | 2014, 2016 |
| **Kalinin 3** | V-320 | 950 | 12/04 | 2034 |
| **Kola 1-2** | V-230 | 411 | 12/73, 2/75 | 2018, 2019 |
| **Kola 3-4** | V-213 | 411 | 12/82, 12/84 | 2011, 2014 |
| **Kursk 1-2** | RBMK | 925 | 10/77, 8/79 | 2021, 2024 |
| **Kursk 3-4** | RBMK | 925 | 3/84, 2/86 | 2013, 2015 |
| **Leningrad 1-2** | RBMK | 925 | 11/74, 2/76 | 2018, 2020 |
| **Leningrad 3-4** | RBMK | 925 | 6/80, 8/81 | 2009, 2011, +20 yr |

| Reactor | Type V=PWR | MWe net, each | Commercial operation | Scheduled close |
|---|---|---|---|---|
| **Novovoronezh 3-4** | V-179 | 385 | 6/72, 3/73 | 2016, 2017 |
| **Novovoronezh 5** | V-187 | 950 | 2/81 | 2010 |
| **Smolensk 1-3** | RBMK | 925 | 9/83-1/90 | 2013, 2020 |
| **Volgodonsk 1 (Rostov 1)** | V-320 | 950 | 3/01 | 2030 |
| **Total: 31** | | **21,743 MWe** | | |

Generally, reactors are licensed for 30 years from first power. Late in 2000, plans were announced for lifetime extensions of twelve first-generation reactors (Leningrad 1&2, Kursk 1&2, Kola 1&2, Bilibino 1-4, Novovoronezh 3&4) totaling 5.7 GWe, and the extension period envisaged is now 15 years, necessitating major investment in refurbishing them by 2006. So far three 15-year extensions have been achieved for Novovoronezh-3&4, Kursk-1&2, Kola-1&2 and Leningrad-1&2. Bilibino 1&2 have been given 5-year license extensions. Replacement of all these twelve units after 2015-20 is planned (Table 1) [4].

In 2006, Rosatom informed it was considering lifetime extensions and upgrading of its eleven operating RBMK reactors. Following significant design modifications made after the Chernobyl accident, as well as extensive refurbishment including replacement of fuel channels, a 45-year lifetime is seen as realistic for the 1000 MWe units. In 2005 they provided 48% of Russia's nuclear-generated electricity. Upgrading of Leningrad 3 is under way with a view to 20-year life extension to 2029, and unit 4 will follow suit. Kursk 3&4 and Smolensk 1-3 will probably be next [4].

In total 15 water-cooled graphite-moderated reactors (11 RBMK-1000 and 4 EGP-6 reactors are being operated. Three first-generation water-cooled graphite-moderated reactors (AM reactor in Obninsk and 2 AMB reactors at Beloyarsk NPP) are permanently shut down, and two larger prototype VVER-440 units at Novovoronezh, a V-210 and V-365 type. The fuel removed from Novovoronezh units has been shipped to centralized storage in Zheleznogorsk and will be stored there for about ten years before reprocessing. The Beloyarsk fuel is still on site since reprocessing technology for it is not available [4].

It is assumed that the best strategy for handling graphite during decommissioning of the power-generating units of a nuclear power plant with uranium-graphite reactors is localization and long-term storage of the graphite masonry together with the reactor in a concrete shaft at the plant site under surveillance. By the time when dismantling is required an ecologically acceptable and cost-effective technology for handling irradiated graphite reactors will be available.

### 3.1.2 Plutonium production reactors

13 high-capacity weapons plutonium production (industrial) reactors were in operation in Russia [5].

Decommissioning concept of these reactors was developed in the middle of 1990s, comprising the dismantling of low-activity structures, sealing of all reactor outlets, and filling of all reactor spaces with special compounds of concrete and bentonite. All these arrangements ensured multi-level protection with a number of safety barriers between the reactor and the environment. At that time a question about the possible reactor dismantling was not raised. The reactor shaft was intended to be transformed into the everlasting radwaste repository. In-shaft waste disposal concept came into conflict with new waste disposal regulatory documents (NP-055-04, NP-069-06, GOST R 50996-96). So, later-on instead of disposal of the reactor shaft the monitored safe storage period was defined [5].

General scheme of decommissioning of these reactors ("storage" on site) is [6]:

- Preparatory period – reloading of the SNF;
- Localization of the stack in the shaft with additional protective engineering barriers;
- Period of the long-term storage (100 and more years) – monitoring and supervision of the reactor;
- Possibility for the dismantling of the reactor after long-term storage.

As a result, the suggested D&D concept allowed qualification of the structure under the IAEA's Stage 2 and ensured its safety for over 300 years. The concept of safe conditioning of reactor facilities, isolation from the environment and radioactive waste disposal is the first and main task in developing a D&D strategy and bringing an area to a partially usable condition.

After the reactors' shutdown the systems for humidity, temperature and radioactivity monitoring continued to operate.

## 3.2 Radwaste management policy

In Russia the law on radioactive waste management has been drafted and is planned to be introduced to the State Duma in 2008. At the present time financing of radioactive waste management program is within the Federal targeted program "Nuclear and radiation safety". 29.7 billion rubles will be spent on the issues of radioactive waste management. The radioactive waste management strategy includes construction and reconstruction of:

- Storage facilities for some 120 thousand cubic meters and of radioactive waste (RW) treatment complexes at nuclear fuel cycle enterprises;
- Storage facilities and complexes for RW treatment at nuclear power plants;
- Storage facilities at the "Radon" facilities for RW management coming from national economy for 140 thousand cubic meters.

Besides that decommissioning of 140 facilities, bringing 80 facilities to secure state, decontamination of territories, buildings and constructions with the total area of 1658 thousand square meters is planned by 2015. Execution of project and research work on construction of interregional intermediate level waste disposal facilities as well as feasibility study and construction of pilot high-level waste disposal facilities is planned. At realization of the RW management strategy the appearance of the national operator in Rosatom structure is planned by 2010, and by 2015 it is planned to design a surface disposal system, and to design a deep geological repository for final RW disposal by 2035. The government informed it plans to spend some $5 billion to 2015 on decommissioning and waste management [3].
Management of irradiated graphite [7]:

- Localization and storage of graphite masonry in situ in concrete shafts;
- Storage of irradiated graphite in special containers;
- Treatment of the irradiated graphite (when such technologies are available);
- Incineration of the graphite (when such technologies are available).

# 4 Compliance with international guidelines and regulations

Although no papers/reports have been identified that indicate Russia is party to, recognises international guidelines and regulations there are several examples of Minatom of Russia cooperating with other countries in many fields of activities, for example:

- nuclear physics;
- fundamental research into matter properties;
- controlled thermonuclear fusion;
- physics of semiconductors and high-temperature superconductivity;
- isotopes;
- technologies of elementary particle accelerators and electrophysical equipment;
- atomic energy generation and nuclear fuel cycle;
- radioactive waste management;
- environment protection.

# 5 Decommissioning of Nuclear Reactors – Current Situation

Updated information on graphite moderated reactors and its shut down date (if relevant) based on the data provided in [8] is presented in **Table 2**.

**Table 2. List of Graphite Moderated Reactors [8]**

| Location | Reactor | Type | Thermal Power | Graphite in reactor tones | Graphite total tones | Commissioned | S/Down Date |
|---|---|---|---|---|---|---|---|
| Kurchatov | Kursk 1 | LWGR | 3200 | 1798 | 2000 | 1976 | |
| Kurchatov | Kursk 2 | LWGR | 3200 | 1798 | 2000 | 1979 | |
| Kurchatov | Kursk 3 | LWGR | 3200 | 1798 | 2000 | 1983 | |
| Kurchatov | Kursk 4 | LWGR | 3200 | 1798 | 2000 | 1985 | |
| Sosnovy Bor | Leningrad 1 | LWGR | 3200 | 1798 | 2638 | 1973 | |
| Sosnovy Bor | Leningrad 2 | LWGR | 3200 | 1798 | 1798 | 1975 | |
| Sosnovy Bor | Leningrad 3 | LWGR | 3200 | 1798 | 1798 | 1979 | |
| Sosnovy Bor | Leningrad 4 | LWGR | 3200 | 1798 | 1798 | 1981 | |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| Desnogorsk | Smolensk 1 | LWGR | 3200 | 1798 | 2158 | 1982 | |
| Desnogorsk | Smolensk 2 | LWGR | 3200 | 1798 | 1798 | 1985 | |
| Desnogorsk | Smolensk 3 | LWGR | 3200 | 1798 | 1798 | 1990 | |
| Beloyarsk (Zarechny) | AMB 1 | LWGR | 286 | 813 | 875 | 1964 | 1983 |
| Beloyarsk (Zarechny) | AMB2 | LWGR | 530 | 813 | 875 | 1967 | 1990 |
| Obninsk | AM-1 | LWGR | 10 | 41 | 41 | 1954 | 2004? |
| Chucotka (Bilibino) | Bilibino 1 | GBWR | 62 | 133 | 133 | 1974 | |
| Chucotka (Bilibino) | Bilibino 2 | GBWR | 62 | 133 | 133 | 1974 | |
| Chucotka (Bilibino) | Bilibino 3 | GBWR | 62 | 133 | 133 | 1975 | |
| Chucotka (Bilibino) | Bilibino 4 | GBWR | 62 | 133 | 133 | 1976 | |
| Cheliabinsk 65 (Chelyabinsk 40, Mayak) | A-Anotchka | LWGR | 500 | 1010 | 1010 | 1948 | 1987 |
| Cheliabinsk 65 (Chelyabinsk 40, Mayak) | IR-A1 | LWGR | 500 | 146 | 146 | 1951 | 1987 |
| Chelabinsk 65 (Chelyabinsk 40, Mayak) | AV-1 | LWGR | 2000 | 1473 | 2173 | 1950 | 1989 |
| Cheliabinsk 65 (Chelyabinsk 40, Mayak) | AV-2 | LWGR | 2090 | 1473 | 2173 | 1951 | 1990 |
| Chelabinsk 65 (Chelyabinsk 40, Mayak) | AV-3 | LWGR | 1500 | 1473 | 2173 | 1952 | 1991 |
| Zelenogorsk (Krasnoyarsk 26) | AD | LWGR | 2500 | 1960 | 3024 | 1958 | 1992 |
| Zelenogorsk (Krasnoyarsk 26) | ADE-1 | LWGR | 2500 | 1960 | 3024 | 1961 | 1992 |
| Zelenogorsk (Krasnoyarsk 26) | ADE-2 | LWGR | 2500 | 1960 | 3024 | 1964 | 2004?? |
| Seversk (Tomsk 7) | 1-1 Ivan-1 | LWGR | 2500 | 1366 | 2066 | 1955 | 1989 |
| Seversk (Tomsk 7) | 1-2 Ivan-2 | LWGR | 2500 | 1366 | 2066 | 1958 | 1990 |
| Seversk (Tomsk 7) | ADE-3 | LWGR | 2500 | 1960 | 3024 | 1961 | 1992 |
| Seversk (Tomsk 7) | ADE-4 | LWGR | 2500 | 1960 | 3024 | 1964 | 2008 |
| Seversk (Tomsk 7) | ADE-5 | LWGR | 2500 | 1960 | 3024 | 1965 | June 2008? |

## 5.1 Power reactors

### 5.1.1 NPP's with RBMK-1000 reactors

As it was already indicated above there are 11 RBMK-1000 reactors (Kursk NPP – 4, Leningrad NPP – 4, Smolensk NPP – 3) in Russia **Table 2**. The Leningrad Nuclear Power Plant is the oldest one. The reactors were put in operation between 1974 and 1981. The oldest reactor received a 15 year prolongation its engineered life span in 2004.

The "Big water channel reactor" RBMK was one of the two nuclear workhorses of the former Soviet Union, limited to Russia, Ukraine and Lithuania. Fuel bundles of low enriched uranium oxide are cooled by boiling water circulating in pressure tubes vertically inserted in a massive graphite pile (Figure 5-1).

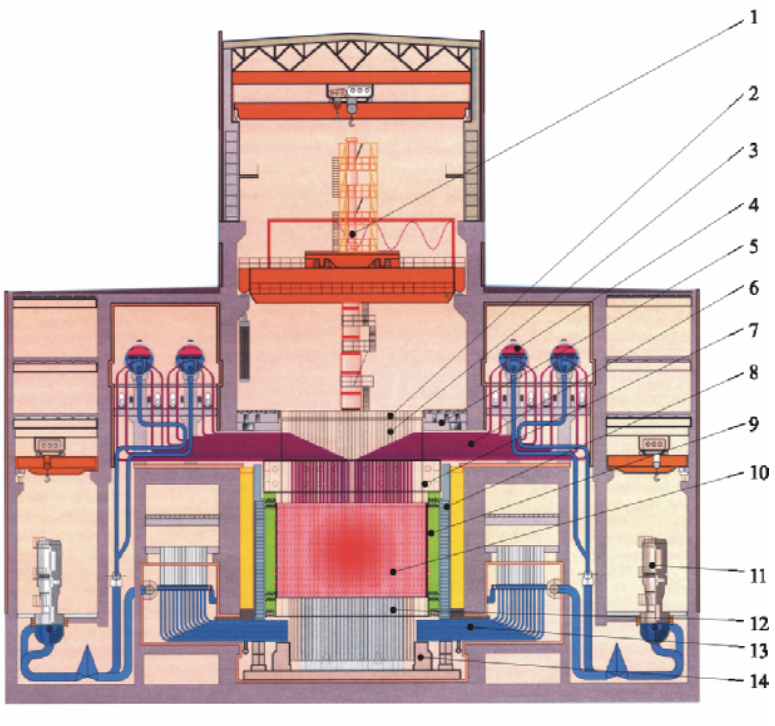


**Figure 5-1.** First Generation Reactor RBMK-1000 [6].

1 – refuelling machine; 2 – block segments; 3 – technological channels; 4 – separator-drum; 5 – top floor; 6– steam-water tubes; 7 – top biological shield; 8 – water shield; 9 – side-wall biological shield; 10 – reactor core; 11 – main circulation pump; 12 – bottom biological shield; 13 – bottom water tubes; 14 – supporting metal structure.

The coolant in the Russian RBMK reactors flows through a series of zirconium fuel-channel tubes. Thermal contact is maintained between the zirconium fuel channel tube and the graphite moderator bricks by a series of graphite rings. Fuel channel tubes of the RBMK reactors can be replaced during operation. This involves not only removal of the zirconium tubes but also removal of the graphite rings. This will create an additional amount of graphite waste. In addition, there is a small amount of graphite used as displacer elements associated with the control rods. Although this displacer graphite is small in quantity, it is possible that it may contain a significant amount of stored energy owing to the low temperature at which it was irradiated. There are also some graphite blocks from moderator repairs stored near some Russian reactors.

The great majority of the radioactive graphite arising from nuclear plant decommissioning is associated with the bulk moderator and reflector graphite in these reactors, together with shield-wall graphite (or other carbon-bearing material) in certain cases. The moderator and reflector components will mainly consist of large graphite blocks, e.g. 250 mm x 250 mm x (200, 300, 500 and 600 mm) for RBMK reactors [8].

### 5.1.2 Beloyarsk NPP

Beloyarsk NPP is located in one of the most picturesque parts of the Urals, on the banks of the river Pyshma, approximately 40 km from Ekaterinburg – the historical, industrial and scientific centre of the Urals. The construction of Beloyarsk led to the birth of the town of Zarechny, while the continental divide between Europe and Asia lays only 70km from the plant. The start of commercial operation at Unit 1 – a 100 MW water-cooled graphite moderated channel-type reactor – at Beloyarsk Nuclear Power Plant (NPP) in 1964 marked the birth of the use of nuclear power for commercial purposes in Russia. Before this time, the word 'nuclear' had only been associated with defense and weapons. The first (AMB-100) and second (AMB-200) reactors of the Beloyarsk nuclear power plant were uranium-graphite channel reactors with nominal power levels of 100 and 200 MW(e), respectively (Figure 5-2). Beloyarsk is the only nuclear power plant in Russia with different types of nuclear power units [9].

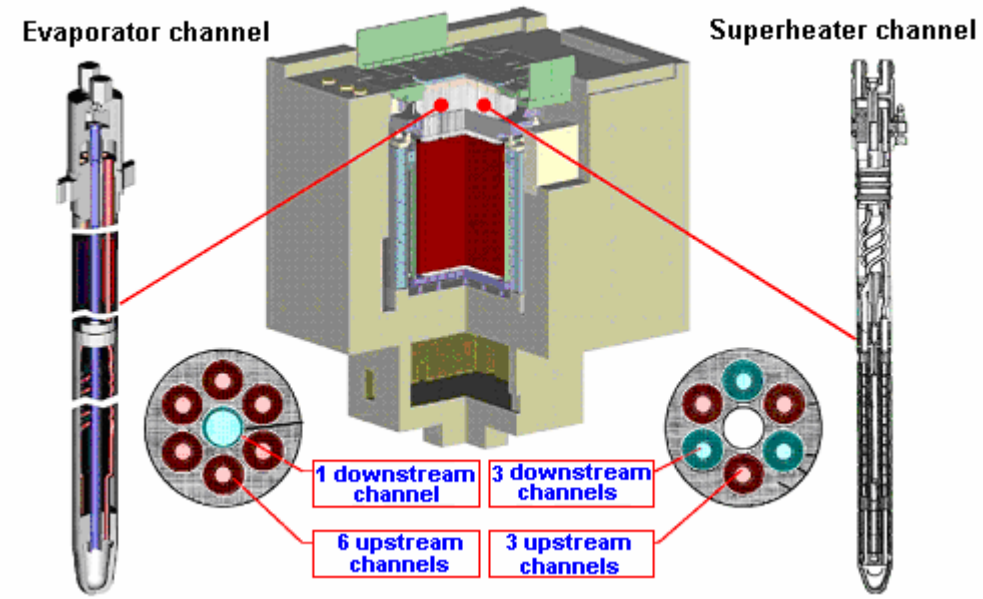

**Figure 5-2. AMB Reactor**

These reactors were shut down in 1981 and 1989, respectively. Nowadays, the condition of the units is in accordance with IAEA Stage 1 – storage under surveillance. The spent fuel has not been removed from the site and is stored in pools.

Radiation control includes monitoring in the production rooms and monitoring of the ventilation systems. This control is implemented with the same methods as are used during operation. Complete dismantling of the reactor equipment and use of the rooms for new nuclear activities has been planned. For such planning purposes, classification of the rooms into the following four categories has been proposed [10]:

- Radiation dose rate exceeding 0.5 mSv/h (50 mR/h); source term reduction by decontamination or decay is considered impractical. Dismantling may be executed only remotely (reactor support structures, graphite core).
- Radiation dose rate between 0.1 mSv/h (10 mR/h) and 0.5 mSv/h (50 mR/h). A combination of decontamination, storage and remote dismantling means is necessary (equipment of the coolant circuit, etc.).
- Radiation dose rate between 0.028 mSv/h (2.8 mR/h) and 0.1 mSv/h (10 mR/h). The working time of personnel must be limited during dismantling work.
- Radiation dose rate up to 0.028 mSv/h (2.8 mR/h). The working time of personnel is not limited during dismantling (equipment turbines, feed water pumps).

### 5.1.3 Bilibino NPP

Bilibino NPP reactors are combined heat and power reactors (Figure 5-3). They, as also AM and AMB power reactors at Obninsk and Beloyarsk, contain a unique fuel design where both the fuel elements and cooling tubes are located in the system of graphite sleeves [11].

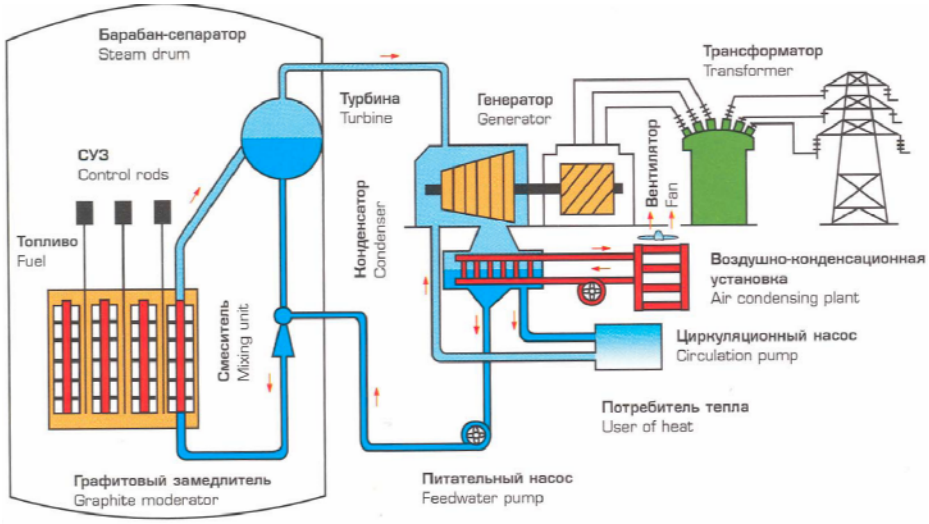


**Figure 5-3. Bilibino NPP with reactor GBWR (EGP-6)**

### 5.1.4 Obninsk NPP

In 1954 the world's first nuclear power reactor, the AM-1 in Obninsk (100 km from Moscow), began operation in the closed city of Obninsk at the Institute of Physics and Power Engineering (FEI). The AM-1 (Atom Mirny - peaceful atom) reactor is water-cooled and graphite-moderated, with a design capacity of 30 MWt or 5 MWe. It was similar in principle to the plutonium production reactors in the closed military cities and served as a prototype for other graphite channel reactor designs including the Chernobyl-type RBMK (reaktor bolshoi moshchnosty kanalny - high power channel reactor) reactors. Russians developed a reactor using LEU (Low enriched uranium) as fuel, graphite moderated and cooled with boiling ordinary water circulating in pressure tubes. This Obninsk prototype was also a dual-purpose plutonium production reactor. AM-1 produced electricity until 1959 and was used until 2000 as a research facility and for the production of isotopes [4].

The reactor fuel is based on 4.4% and 10% enriched uranium. As of 1996, the core contained 122 fuel assemblies, 31 with an initial enrichment of 4.4% and 91 with an initial enrichment of 10%. The fissile material content in the assemblies was 120kg. The AM-1 is used to test fuel rods and reactor materials, to conduct nuclear physics and radiobiology research, and to produce isotopes for medicine and industry. As of July 1999 the reactor was being prepared for shutdown, scheduled for 2004. After closure, the reactor may become an exhibit at a planned museum of nuclear power [12].

## 5.2 Plutonium production (industrial) reactors

Many of Russia's fuel cycle facilities were originally developed for military use and hence are located in former closed cities in the country.

A brief review of the design of these reactors (Figure 5-4) presented in [13] shows, that they include a heavy graphite stack, aluminum tubes charged with fuel elements made of metallic natural enriched uranium and control system elements. The closed support and protective structures are made of carbon and corrosion resistant steel. The protective structures are filled with sand and iron ore mixture, and the side structures are filled with water. The overall mass of the metal structures is over 3000 tones. The water coolant used was: refined river water for the I1 reactor, and purified, chemically condensed water (95%, 99%) for the E2 and ADE3 reactors.

In accordance with the agreements between the Russian and US Governments on completion of weapons plutonium production, ten powerful industrial reactors were unloaded and decommissioned in the Russian Federation. Three such reactors, I1, E2 and ADE3 commissioned in 1955, 1957 and 1961, were decommissioned in 1989,1990 and 1992, respectively. In 1993, work started on the Siberian Chemical Plant for decommissioning of two, and later three, uranium-graphite reactors, which also included review of the project development and manufacture of equipment and accessories. The financial plan became the basis of the Branch Programme accepted by the Ministry, and the consolidated resources developed on the basis of this plan were used for the Branch tasks. During the research, project, manufacturing, and reactor maintenance stages the expenses exceeded 3 billion rubles.

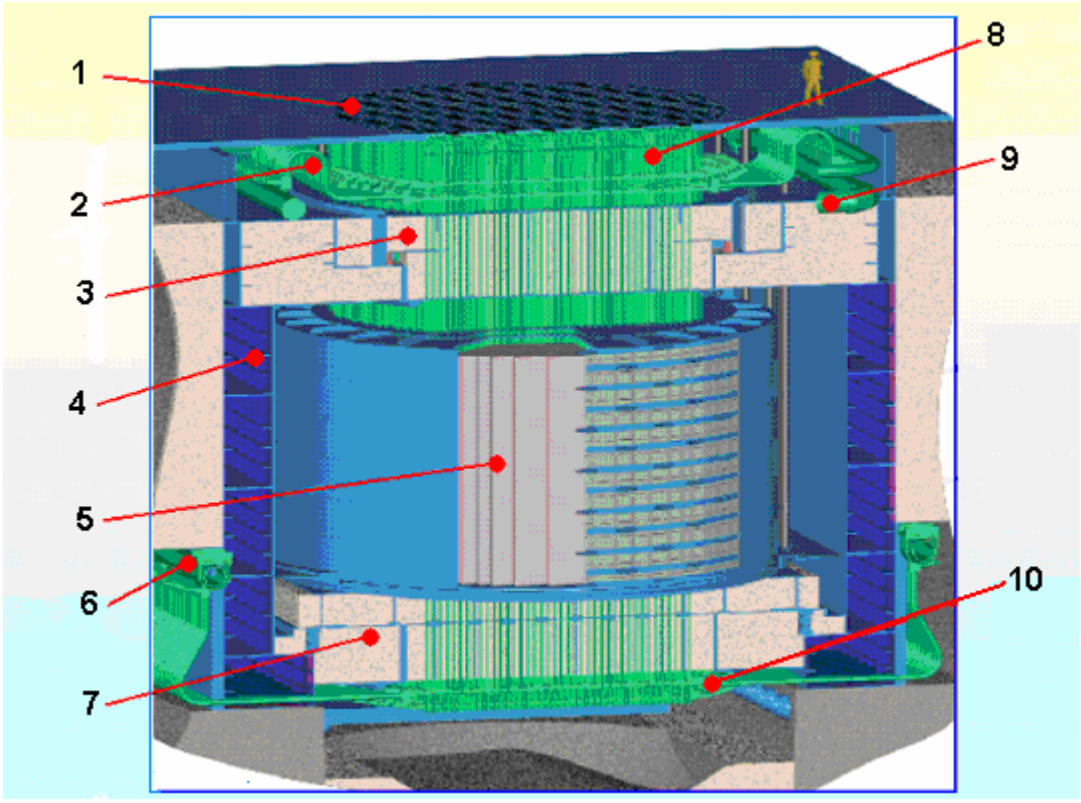


**Figure 5-4. Plutonium Production (Industrial) Reactor [7]. 1 – block segments, 2 – top distribution headers, 3 – top biological shield, 4 – side biological shield, 5 – graphite stack, 6 – bottom distribution headers, 7 – bottom biological shield, 8 – technological and control channels, 9 – top distribution header, 10 – bottom distribution headers.**

By 2010 over 8 billion rubles will be needed for all the work on three reactors and storage facilities on the plant's site, depending on the method of decommissioning, inflation rate, personnel salaries, etc [13].

As it is indicated in [7] there are 21000 t. of graphite with total activity of about 650 $10^3$ Cu (based on $C^{14}$) in closed reactors. Also, there are about 8000 t. of irradiated graphite rings in the storage facilities at reactors sites. Graphite stacks of the reactors are at some depth underground so ground waters can penetrate into the stack and release of the radionuclides could be possible.

### 5.2.1 Siberian Chemical Combine (Seversk, formerly Tomsk-7)

The closed city of Seversk, formerly Tomsk-7, is the location of the Siberian Chemical Combine (SKhK). Seversk and its related nuclear facilities are located about 15km north-west of the city of Tomsk. The Siberian Chemical Combine is made up of several large facilities: the Reactor Plant, the Isotope Separation Plant, the Radiochemical Plant, the Conversion Plant, the Chemical Metallurgical Plant, the Scientific Research and Design Institute, fissile material storage facilities, radioactive waste management facilities, and a number of auxiliary facilities.

SKhK was established in 1949, and began producing highly enriched uranium (HEU) for the Soviet nuclear weapons program in 1953. SKhK is one of the largest weapons-grade plutonium production enterprises in the world. The reactor plant houses five plutonium-production reactors. The Ivan-1 reactor began production on 20 November 1955; its sole purpose was the production of plutonium. Later reactors – Ivan-2 (September 1958), ADE-3 (14 July 1961), ADE-4 (1965), and ADE-5 (1968) – were dual use reactors that produced both plutonium and heat and electricity for local communities. The Ivan-1 and Ivan-2 reactors were shut down in 1990 and the ADE-3 reactor was shut down in 1992. ADE-4 was shut down in April 2008 and ADE-5 followed in June of that same year, effectively ending weapons-grade plutonium production at Seversk. The reactors were light-water cooled/graphite-moderated, with a capacity of 2,500 MWt, and were fueled by natural uranium [14].

#### *5.2.1.1 Methods of decommissioning*

For the D&D of these specified types of reactors indicated above a method of low activity reactor structure dismantling was chosen and implemented [13]. The remaining structures were left in the vault and all reactor outlets were sealed to the environment. The support structures were secured, and the biological shield tanks, the reactor space and some of its cavities were filled with a special compound of concrete in a betonite base. All these arrangements ensured multi-level protection, with a number of safety barriers between the reactor and the environment. As the first barrier all the cavities in the core material were filled with natural materials in a concrete base. The second barrier is metal and hermetic, using as a basis the metal structures that form the reactor's surroundings, and improved by sealing and filling the biological shield tanks with concrete. The third barrier was the vault itself based on the structures surrounding the shaft. Concrete walls were formed and the lower structures were

concreted, etc. The fourth barrier was the surrounding strengthened building structures. Additional safety barriers were also provided.

#### 5.2.1.2 Technology and equipment

A high radiation level and inaccessibility of the reactor components demanded that special technologies be developed for remote cutting of the channel pipe over the protective plate, as well as methods of remote cutting of holes in metal structures to fill them with special compounds, remote welding of seals to isolate the reactor from the environment, and technologies to cut specimens out of the material after neutron exposure in order to study its changed properties [13]. In order to develop these technologies, the following special equipment was developed: remote controlled equipment for plasma, electric-contact and machined cutting; remote controlled welding machines for sealing of the channels and other pipes; transport arrangements; cutters for pipelines of the humidity/temperature monitoring systems and for other structural parts. Special betonite based compounds were also developed whose fluidity guaranteed complete filling of any reactor cavity in the project. Special technologies and equipment were needed to apply about 2200 $m^3$ of the compound to each reactor thorough the above mentioned remotely cut holes. Great attention was paid to quality inspection of the filling and welding operations. For this purpose special optical and TV systems were used.

Work on two reactors of the Siberian Chemical Plant has just reached the final phase. The total exposure of the personnel is within permissible limits. A complex engineering-radiation study was performed on the A, AB1, AB2 and AB3 reactors in the PO MAYAK Enterprise, the results of which should help to define the scope of the work. The decommissioning principle will be the same. The following conclusions were drawn as a result of the data presented above:

- The D&D concept ‘termination on site’ for uranium-graphite reactors has been accepted and implemented in the Russian Federation for the I1, E2 and ADE3 reactors, with the capability to dismantle the rest of the equipment at the same time. A system of protective barriers (multi-level protection) is being developed in order to avoid environmental pollution from disposed radioactive waste and to protect the other reactor components from external natural or technical influences.

- A special monitoring system has been provided for the inspection of storage parameters, including measurements of temperature, humidity, gas composition, radioactivity, and location of support metal structures. A system of special environmental drill holes is provided on the site for inspection of the conditions.
- Unique technologies and equipment were developed for dismantling some of the structures and for the safe and successful performance of all works.
- The experience obtained by application of the technologies and equipment can be used, after relevant adjustment, for the decommissioning of all channel type uranium-graphite nuclear reactors.

### 5.2.2 Production Enterprise Mayak

The Federal State Unitary Enterprise 'Production Enterprise Mayak' (the FSUE 'PE Majak') was founded for the industrial production of plutonium for nuclear weapons. It is located in Cheljabinsk province in the South Urals, not far from the cities of Kishtym and Kasli. The Ozjersk city (or formerly Chelyabinsk-65 (previously known as Chelyabinsk-40)) in which the Majak staff and their families live is situated not far from the enterprise itself. Ozersk is located approximately 70km north of Chelyabinsk. Mayak facilities include plutonium and tritium production reactors; fuel reprocessing facilities; a plutonium processing, finishing, and component manufacturing plant (Plant 20); mixed-oxide (MOX) fuel fabrication plants; and nuclear waste treatment and storage facilities [14,15].

The first Soviet reactor built to produce plutonium for military purposes, called Annushka (A for short), began operating at Mayak in June 1948. The reactor was a single-purpose, water-cooled, graphite-moderated, single-pass reactor. Four additional graphite-moderated plutonium production reactors were built at Mayak between 1950 and 1952. In addition to the graphite-moderated reactors, one heavy water-moderated reactor was also built during this period. All five of the plant's uranium-graphite plutonium production reactors (A, IR, AV-1, AV-2, and AV-3) have been permanently shut down. Two tritium-producing reactors (Ruslan and Lyudmila) are still in operation [14].

### 5.2.3 Mining and Chemical Combine (Zheleznogorsk formerly Krasnoyarsk-26)

The city of Zheleznogorsk (formerly Krasnoyarsk-26), located about 70km northeast of the city of Krasnoyarsk, was established in 1950 to produce plutonium for nuclear weapons. The city's

population is approximately 100,000. Approximately 8,000 people continue to work at the Mining and Chemical Combine (GKhK) in Zheleznogorsk.

Three graphite-moderated ADE-type underground reactors were used to produce plutonium. Two were shut down in 1992, one in June and one in September, and the last remaining reactor was mainly used as a district heat and electricity producing plant. The ADE reactor uses natural uranium fuel and accumulates weapons grade plutonium at an estimated rate of about 0.5 MT per year. According to the 1994 Agreement Concerning the Shutdown of Plutonium Production Reactors and the Cessation of Use of Newly Produced Plutonium for Nuclear Weapons, the ADE reactor was also shut down [14].

## 5.3 Research Reactors

It is unclear exactly how many research reactors and critical/subcritical assemblies exist in Russia. It seams there are 113 research reactors and critical/subcritical assemblies in Russia. Of this total, approximately 40 are research reactors, with about half of them located in Moscow [14]. In the first half of 2000, Gosatomnadzor reported that it inspected 109 research reactors and critical/subcritical assemblies. Arriving at an exact number is complicated by the fact that the reactors and assemblies are owned by various Russian entities, including academic institutions and the ministries of Atomic Energy, Education, Health, and Defense.

At the very beginning, though, there were not so many possible choices. The only available fissile material was natural uranium, with its given 0.7% 235U. To sustain a chain reaction with natural uranium, one had to thermalize as quickly as possible the fast fission neutrons to avoid their capture in the resonances of 238U: the reactor could only be heterogeneous and moderated with light elements. Hydrogen being too absorbent, the choice of moderator was between heavy water, graphite or, possibly beryllium. At the time, the world stockpile of heavy water (painfully produced by electrolysis in Norway) was small, while pure graphite was industrially produced in the USA in quantities for electric furnace electrodes.

Consequently, CP1 had to be what it was: a regular and roughly spherical array of uranium pebbles (metal and oxide) within a pile of graphite bricks. Hence the name “atomic pile” which used to designate early nuclear reactors, while electrochemical piles retain the name of the

original Volta piles of wafers. For the same reasons, a similar design was adopted in 1946 for the first Soviet reactor, still visible today in the Kurchatov Institute [16].

Russia has had substantial R&D on nuclear power for six decades. The premier establishment for this is the Kurchatov Institute in Moscow, which has run twelve research reactors there, six of which are now shut down. The F-1 research reactor there was started up in December 1946 and recently marked its 60th anniversary in operation.

# 6 Graphite Characteristics

## 6.1 RBMK reactors at Leningrad NPP

### 6.1.1 Experimental investigations

In 1998 the unique experiment of withdrawing the graphite column from reactor RBMK-1000 of Leningrad NPP Unit 3 has been performed. The graphite column (all 14 graphite bricks) was withdrawn from the cell 12-36 after 18 years of reactor operation for long scale investigation of graphite brick (GB) behaviour and estimation of methods of graphite core dismantling.
The total power production per this cell was 9060 MW day and maximal neutron fluence on the inner surface of graphite bricks was about $1 \cdot 10^{22}$ neutron/cm$^2$ ($E_n > 0.18$ MeV). The graphite temperature was estimated as 500-600$^o$C.

The research program includes the investigation of all physical-mechanical properties of graphite and internal irradiation induced stresses arising across the graphite brick wall during reactor operation. The information presented below is based on [17].

#### *6.1.1.1 Dimensional changes and physical-mechanical properties*

The results of dimensional changes investigation showed that graphite bricks have very complicated geometry due to irregular graphite shrinkage in different direction of GB. The shrinkage of graphite bricks was about 0.3-1.4% in radial (perpendicular to extrusion) direction and about 2-2.5% in axial (parallel) direction.

Preliminary results of investigation of graphite physical-mechanical properties showed that even for the lowest GB (GB No1 with the smallest achieved neutron fluence) properties of graphite changed essentially. So, the value of elastic modules increased up to 7.7 GPa (40 %)

for specimens of radial direction and up to 9 GPa for specimens' axial direction. The electrical resistivity of such specimens increased up to 29-30 · $10^{-6}$ Ohm m (40 %) and up to 18 · $10^{-6}$ Ohm m (30 %) correspondingly. (The initial properties of graphite are presented in Table 3).

**Table 3. Physics and Mechanical Properties of Graphite GR-280**

| Properties | Value |
|---|---|
| d, g/cm$^3$ | 1.71 |
| $R^{com}$, MPa | 34/24 |
| $R^{ten}$, MPa | 7.6/6.0 |
| $A_{com}$, % | 1.6/2.0 |
| $A_{ten}$, % | 0.2/0.2 |
| $E_{din}$, GPa | 6.5/5.0 |
| α, $10^{-6}$, 1/K | 4.0/5.4 |
| λ, W/m·K | 48/37 |
| ρ, $10^{-6}$, Ohm·m | 10/13 |
| $K_{1C}$, Mpa·m$^{1/2}$ | 6.1 |

Notes:

1. In the numerator-parallel direction, in the denominator-perpendicular direction.
2. CTE has been measured at 400°C.
3. λ has been measured at 650°C.
4. The sizes of GR-280 GB: 250x250x600 mm.

#### *6.1.1.2 Gas release under gamma-irradiation*

The results of investigation of gas release from graphite under gamma-irradiation showed that the rate of gas output from irradiated graphite increases about 10-11 times. Chromatographic analyses of gaseous products showed that it is mainly-hydrogen ($H_2$ – 19 %), nitrogen ($N_2$ – 36 %) and carbon dioxide ($CO_2$ – 40 %).

#### *6.1.1.3 Leach testing*

Leach testing showed that the total output of isotope Cs-137 in solution (150 cm$^3$) after 90 days of testing was estimated as 6.4·$10^3$ Bk and the rate of Cs-137 release was calculated as 4.7·$10^{-13}$ g/cm$^2$.day.

It should be noted that there was an incident with failure of fuel channel in 1992 year at this plant and spectral analyses showed that in graphite various radionuclides are present mainly isotopes Cs-134, Cs-137, Co-60, Mn-54 (Table 4).

**Table 4. Relative Contents of Radioactive Isotopes in Graphite Brick No 11 of Graphite Column Which Was Withdrawn from Leningrad NPP, Unit 3.**

| Outer surface | | Inner surface | |
|---|---|---|---|
| Isotope | Contents, Ku/probe | Isotope | Contents, Ku/probe |
| Co-60 | $3.1\ 10^{-7}$ | Co-60 | $3.6\ 10^{-7}$ |
| Cs -134 | $1.2\ 10^{-8}$ | Cs- 134 | $3.8\ 10^{-9}$ |
| Cs-137 | $2.0\ 10^{-8}$ | Mn-54 | $2.6\ 10^{-9}$ |
| Σγ | $3.4\ 10^{-7}$ | Σγ | $3.7\ 10^{-7}$ |

The specimens of such graphite (from graphite brick No 11- upper part of graphite column) were covered with a special compound and tested to estimate the rate of Cs-137 (and another radionuclides) release under basic/acid conditions in accordance with the method for accelerated leaching of solidified waste.

This compound was developed for conservation of radioactive graphite (and other materials) in process of NPP decommissioning. It was prepared on the base of epoxy furfural resins. Full-scale testing of it is being carried out in RRC KI and RDIPE.

Five cubic specimens (30×30×30 mm) were fabricated (Table 5).

**Table 5. Graphite Specimens for Leach Testing.**

| Specimens | Weight before imp., g | Surface, $cm^2$ | Weight after impr., g | Weight of compound , g | Thickness of compound coating,mm | Thermo treatment |
|---|---|---|---|---|---|---|
| 1 | 46.431 | 56.25 | 49.3368 | 2.9058 | 0.516 | 80-85 °C, 12 hr. |
| 2 | 43.8460 | 58.2 | 46.6568 | 2.8108 | 0.582 | |
| 3 | 49.6207 | 60.42 | 52.4695 | 4.7473 | 0.785 | 80-85 °C, 12 hr. |
| 4 | 52.1575 | 61.16 | 54.8000 | 4.7495 | 0.765 | |
| 5 | 44.4550 | | | | 0. | |

The total activity on the surface of specimens was $1.5{\cdot}10^2$, $3.3{\cdot}10^6$ and $6{\cdot}10^8$ Bk accordingly. All specimens were placed in vessels with distillate water (volume 150 $cm^3$) for testing. Gamma-spectral analyses of water were performed using gamma spectrometer with Ge-Li detector. Probes of water (0.65 $cm^3$) for investigation have been taken after 30, 60 and 90 days. The results of testing are presented in Table 6 and in Figure 6-1.

**Table 6. Activity of Distillate Water in Vessel Due to Leach Testing. Graphite Specimen Without Compound.**

| Time of testing | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| 30 days | | | 60 days | | | 90 days | | |
| α-activity, Bk | β-activity, Bk | γ-activity, Bk | α-activity, Bk | β-activity, Bk | γ-activity, Bk | α-activity, Bk | β-activity, Bk | γ-activity, Bk |
| $5.8 \cdot 10^2$ | $2.4 \cdot 10^4$ | $1.6 \cdot 10^3$ | $7.0 \cdot 10^2$ | $2.9 \cdot 10^4$ | $4.5 \cdot 10^3$ | $1.3 \cdot 10^3$ | $4.8 \cdot 10^4$ | $6.4 \cdot 10^3$ |

Note:

1. α-activity only for isotope Cs-137.
2. Volume of water – 150 $cm^3$.

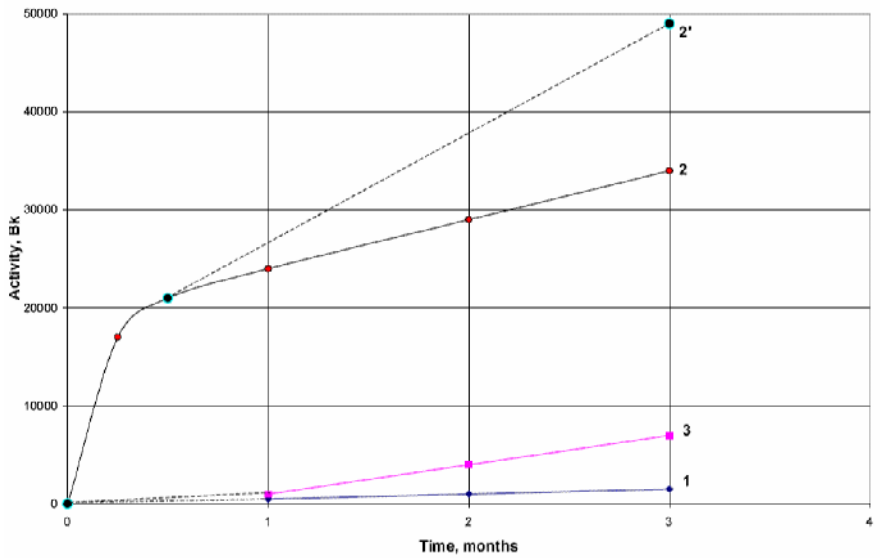


**Figure 6-1. Realease of α (1), β (2 and 2'), γ (3) – activities from irradiated graphite under leach testing (2' – after evaporation of solution).**

The results showed that after 90 days of testing the release of any radionuclides from specimens covered with compound was absent.

One specimen (No 5) was not covered with compound. The total release of isotope Cs-137 in solution (150 $cm^3$) after 90 days of testing was estimated as $6.4 \cdot 10^3$ Bk (Table 6) and the rate of Cs-137 release was calculated as $4.7 \cdot 10^{-13}$ $g/cm^2$.day.

#### 6.1.1.4 Radiation Stability Of Compound

The irradiation stability of compound specimens under gamma-irradiation was tested using RRC-KI γ-rig-GUT-200M (Co-60 source, dose rate 300 Rad/s) up to the dose 6000 MRad and investigation of rate of gaseous products release-using another γ-rig-RHM-γ200 (dose rate 80 Rad/s).

These results showed that γ- irradiation up to such dose gives rise to specimen's diameter shrinkage (about 0.6-0.9 %) and to specimens weight loss (about 1-1.7 %) (Table 7).
The investigation of radiation stability of such compound showed that the rate of gaseous products release from it under gamma-irradiation up to dose 6000 MRad was about 8.5 10-10 $cm^3$/g. rad that is 10 times smaller than such release for silicon-organic radiation resistant elastomer EKOR.

**Table 7. Dimensional Changes and Weightloss of Compound Specimens Under γ-Irradiation up to Dose 6000 MRad**

| Specimens | Diameter, mm. | | | Weight, g. | | |
|---|---|---|---|---|---|---|
| | Before irradiation | After irradiation | Diameter decreasing, % | Before irradiation | After irradiation | Weight loss, % |
| 1 | 10.16 | 10.10 | 0.590 | 1.4091 | 1.3865 | 1.6 |
| 2 | 10.19 | 10.09 | 0.981 | 1.4408 | 1.4256 | 1.05 |
| 3 | 10.15 | 10.08 | 0.689 | 1.3711 | 1.3548 | 1.18 |

### 6.1.2 Computational estimations

Computational estimates of the radiation characteristics of irradiated graphite after final shutdown of a nuclear power plant with an RBMK-1000 reactor were performed in [18]. The MCNP computer code is used to determine the spatial distribution of the neutron flux density in the interior volume of the graphite, the CHAIN code is used to determine the isotopic composition and the radiation characteristics of the irradiated graphite on the basis of the MCNP fluxes. The results of the calculation of the radiation characteristics of graphite from the reactors in the Nos. 2 and 3 units of the Leningrad nuclear power plant and the No. 1 unit of the Chernobyl nuclear power plant are presented and the contribution made by the accident to the flow of fuel mass into the masonry is estimated.

The calculations showed that the impurities present in the initial material are the main source of activity of irradiated graphite in RBMK masonry. The γ radiation and, correspondingly, the dose rate of graphite blocks are due primarily to Co-60. A calculation performed neglecting the possible escape of tritium from the masonry during operation and after the reactor is shut down shows that tritium dominates in the total activity of graphite, while the contribution of C-14 becomes appreciable only if the holding times are long.

The main source of error in calculating the radiation characteristics of irradiated graphite is the uncertainty in the initial content of the main impurities, such as cobalt and lithium, in the initial material. The radiation characteristics of graphite depend on the operating time of the power-generating unit, and if the unit operates for less than the nominal service life, the activity is higher because of under burning of impurity elements. Additional activity is added to the masonry during accidents with a rupture of a channel, which also can be taken into account by a computational method if sufficient information about the nature of the accident is available. At the present time, there is no accessible or adequate experimental database on the isotopic composition and radiation characteristics of irradiated RBMK graphite.

## 6.2 AM reactor at Obninsk NPP

Results on investigation of radioactive contamination of graphite samples from the AM reactor are presented in [19]. During operation, incidents occurred with loss of seal and sometimes loss of integrity of the fuel-element claddings in some cells and particles of the fuel and steam-water mixture entered the graphite masonry. This resulted in radiation contamination with a complex radionuclide composition.

An analysis of the radioactive contamination of the graphite masonry of the reactor in the world's first nuclear power plant (Obninsk) was undertaken to check and supplement the previously existing information [20]. As in other similar investigations [21], the data are needed for planning measures on decommissioning a reactor and management of radioactive wastes. We shall list certain features of the construction and operation of the AM reactor [22]:

- The graphite masonry consists of 151 vertical columns arranged in a triangular lattice

with spacing 120 mm and assembled from blocks with a hexahedral cross section with a 65 mm in diameter interior opening at the center, and 24 columns assembled from horizontal blocks at the periphery (in the reflector); the mass of the graphite in the masonry is 46.3 tons (10.3 tons at the center);

- The enrichment of the fuel loaded into the core during different periods ranged from 4.4 to 10% U-235;
- The reactor continued to operate for almost 48 yr with interruptions;
- The maximum thermal-neutron fluence was ~$2 \cdot 10^{22}$ $cm^{-2}$, the nominal thermal power was 30 MW, and the average power was 9.5 MW;
- The working temperature of the graphite reached 700°C.

It should also be noted that during reactor operation there were incidents in which the seal was broken and sometimes the integrity of the fuel-element cladding in certain cells was disturbed and particles of the fuel and steam-water mixture entered the graphite masonry. The neutron irradiation activated impurities present in the graphite, resulting in multicomponent contamination.

### 6.2.1 Gamma and x-ray spectrometric measurements

Twenty-eight samples were obtained to investigate the content of fission products, products of activation of impurities, and actinides in the graphite. These samples were prepared from cores taken from four cells in different parts of the masonry. The data obtained characterized the distribution of radionuclides over the height of the core and the volume of individual graphite blocks.

As established previously, the activity of Cm-244 and Am-241 on the surface of the blocks was 10–100 times higher than in the volume in the graphite of the reactors at the Siberian Integrated Chemical Plant (I-1, EI-2, ADE-3) [20]. In the graphite of the AM reactor, the activity of these radionuclides is close to the level of the volume contamination of the graphite in the masonry of the reactors at the Siberian Integrated Plant and much lower than the surface activity in these reactors. The ratio of the Cs-137 and Am-241 activity in the samples of the graphite from the AM reactor is approximately the same as in the sample from the reactors at the Siberian

Integrated Plant. The Am-241, Cm-244, and Cs-137 activity of the samples of all four channels from the center of the core was close (
Table 8).

**Table 8. Am-241, Cs-137, and Cm-244 Activity in Different Cells at the Center of the Core of the AM Reactor, Bq/g.**

| Sample No. | Am-241 | Cs-137 | Cm-244 |
|---|---|---|---|
| 14 | $2.4 \cdot 10^2$ | $6.9 \cdot 10^3$ | $1.3 \cdot 10^4$ |
| 24 | $1.9 \cdot 10^2$ | $5.7 \cdot 10^3$ | $3.7 \cdot 10^2$ |
| 34 | $7.5 \cdot 10^2$ | $2.8 \cdot 10^4$ | $3.2 \cdot 10^3$ |
| 44 | $2.3 \cdot 10^2$ | $1.0 \cdot 10^4$ | - |

The distribution of actinides and fission products along the core is similar; their content decreases in the direction from the process channel into the bulk of a graphite block. Deep penetration of these nuclides into the interior volume of the graphite masonry was found; this was not observed in the graphite of the reactors at the Siberian Integrated Chemical Plant. This difference could be due to the different amount of graphite and the two times higher working temperature of the graphite in the AM reactor, the long operating period of this reactor, and the large number of incidents with disruption of the integrity of the fuel-element cladding and flooding of the masonry of AM, all of which occurred right up to the moment when the reactor was shut down. The distribution of the products of activation of impurities (Co-60, Zn-65) in the graphite does not exhibit a regular behavior. Nonetheless, the nonuniformity of the distribution of these radionuclides is much smaller than in the reactors at the Siberian Integrated Chemical Plant. The distribution of all radionuclides over the height of the graphite masonry exhibits a general feature: their content increases substantially at the bottom of the masonry (Table 9).

This is probably due to the spillage of particles of radioactive graphite during drilling and repair work done on the channels.

On the basis of the results obtained one can imagine the following mechanism leading to the formation of radioactive contamination in the graphite masonry. Temperature acting over a prolonged period of time resulted in cracking of the graphite blocks. The uranium particles,

which enter the coolant as a result of incidents, were disseminated by the steam–water mixture, penetrating deep into the cracks of the graphite moderator. Then, under the action of the

**Table 9. Average Am-241, Cs-137, and Cm-244 Activity along the Height of the Masonry of the AM Reactor, Bq/g (average over four channels).**

| Height | Am-241 | Cs-137 | Cm-244 |
|---|---|---|---|
| 1 (top) | 38 | $3.6 \cdot 10^3$ | $3.4 \cdot 10^2$ |
| 2 | 66 | $1.4 \cdot 10^4$ | $2.0 \cdot 10^3$ |
| 3 | 15 | $1.4 \cdot 10^4$ | $5.5 \cdot 10^2$ |
| 4 | $4.7 \cdot 10^2$ | $1.3 \cdot 10^4$ | $2.3 \cdot 10^3$ |
| 5 | $1.3 \cdot 10^2$ | $1.8 \cdot 10^4$ | $5.7 \cdot 10^3$ |
| 6 | 5 | $2.0 \cdot 10^4$ | $9.0 \cdot 10^2$ |
| 7 (bottom) | $3.7 \cdot 10^2$ | $3.8 \cdot 10^4$ | $2.5 \cdot 10^4$ |

neutron flux, the uranium present in the graphite was converted into fission products and heavy actinides. This is confirmed by the decrease in the content of fragment radionuclides in the direction away from the process channel into the interior space of a graphite block, similar to the distribution of fission products which have different migration properties, similar to the distribution of fission products and Am-241.

### 6.2.2 Measurement of the activity of C-14 and H-3 in graphite samples

Five samples were obtained from four cells at different heights in the masonry to determine the content of these radionuclides in the graphite. Following the procedure used in [23], these samples were burned at 700°C in a melt with a two-component oxidizer containing $V_2O_5$ and CuO. To perform measurements, approximately 50 mg pieces of graphite are pricked off the graphite cores and placed into a retort for burning. H-3 and C-14 were separated from the sample in a gaseous form (HTO and $^{14}CO_2$). The gaseous products $CO_2$ and $^{14}CO_2$ were passed through a trap, filled with NaOH, for complete absorption with formation of water-soluble compounds. After the graphite was completely burned, the contents of the trap were washed off with distilled water. The coefficient of transfer of C-14 from the sample into the liquid phase was 0.95 ± 0.05. The tritium-containing gas phase (vapor of tritium water), formed by oxidation of the graphite, and condensed in the nitrogen trap. The coefficient of transfer of tritium from the sample into the liquid phase was 0.75 ± 0.1. Some of the solution formed was introduced into a liquid scintillator for subsequent measurement of the activity of C-14 and tritium.

The results obtained show the following:

- The activity of most of the experimental samples does not differ by much, reaching maximum values at the bottom of the masonry at heights +500–+1500 mm;
- The average specific activity of C-14 in the experimental graphite samples taken from the AM reactor was about $10^5$ Bq/g, which is 10 times lower than in the graphite of the reactors at the Siberian Integrated Chemical Plant; however, it should be noted that the neutron fluence over the operating time of the reactors indicated is not the same and differs by more than a factor of 5;
- The average activity of the tritium in the graphite of the reactors being compared is almost identical (~$10^4$ Bq/g).

### 6.2.3 Determination of the uranium content and the ratio U-235/U-238 in graphite samples

The uranium content and the isotopic ratio of U-235/U-238 in samples taken from the graphite masonry were determined by neutron-activation analysis. The amount of uranium in the experimental samples with a mass of about 0.2 g was measured according to its content in a comparison sample which was made by evaporating a solution, containing a precisely known amount of natural uranium, onto a graphite substrate.

The experimental samples were irradiated in a vertical experimental channel of the IRT reactor at the Moscow Engineering Physics Institute. The thermal-neutron flux density in the irradiation zone was about $2 \cdot 10^{13}$ $sec^{-1} \cdot cm^{-2}$. The graphite samples placed in aluminum foil were irradiated for 2 h. The measurements performed with a germanium detector were started on the day after irradiation and continued for 3 days. A single measurement took 5 min to 1 h depending on the activity of the sample.

The uranium content was analyzed in 22 graphite samples from cells 13-12 and 11-06. The average content of uranium in the graphite of the cells studied differs by approximately a factor of 2. The isotopic composition of the uranium is almost the same (see Table 10). The content of uranium on different sides was compared for the two cores taken from cell 11-06: on the channel side and from the interior of a graphite block. In one core, the uranium content differed by a factor of 3 and in the second one by a factor of 10.

**Table 10. Average U-235 and U-238 Content per 1 g of Graphite.**

| Mass | Cell | |
|---|---|---|
| | 13-12 | 11-06 |
| U-235, ng | 47.3 | 87.3 |
| U-238, ng | 1280 | 2580 |
| U-235/U-238, % | 3.6 | 3.3 |

The investigation confirmed a previously drawn conclusion that the amount of C-14 in spent graphite masonry can be estimated from the results of a small number of analyses and computational data on the distribution of neutrons in the volume of the reactor. The uranium contamination of the graphite in the AM reactor is substantial and "younger," and the ratio U-235/U-238 is higher than in the commercial uranium–graphite reactors which were shut down at the Siberian Integrated Chemical Plant. Judging from the small sample obtained, the total mass of U-235 in the masonry of the AM reactor does not exceed 3 g.

The processes by which radionuclides penetrate into the masonry of the reactors being compared differed substantially: in the first case, surface contamination of the graphite blocks prevailed on the side of the process channel and at the end surface; in the second case, the dissemination of the fuel particles by the steam–water mixture along the cracks in the graphite led to volume contamination of the blocks by uranium with maximum contamination on the side of the process channels.

A correlation was established between the distribution of separate fission products and actinides. This shows that they are secured on sections where a fission event has occurred. In our opinion, this makes it possible to estimate the content of radionuclides which are difficult to measure, for example, Tc-99, I-129, and others, from the activity of radionuclides for which

measurements are easy to perform, for example, Cs-137, by using data on their yield and half-life.

## 6.3 Plutonium production (industrial reactors)

### 6.3.1 Siberian Chemical Combine (Seversk (Tomsk-7))

Results of rather detailed experimental investigations of the radioactive contamination of graphite masonry in the commercial reactors at the Siberian chemical combine are presented in[24,25,26,27,28,29]. The information presented below is based on the generalized information provided in [29]. The investigations were performed on I-1, EI-2, and ADE-3 reactors at the Siberian Chemical Combine in 1996-2001. Analysis of a large number of samples made it possible to construct a detailed picture of the contamination of the masonry, study the distribution of the radionuclides of different origin in the masonry, construct schemes for making estimates, and estimate the content of certain radionuclides, including C-14, H-3, Sr-90, Am-241, Cm-244, Pu 238-241, Cs-137,134, and Co-60.

The operating conditions of the reactors were as follows: the operation period was about 30 yr, the graphite was enclosed in a nitrogen atmosphere, and incidents with destruction of technological channels and repeated leaks of the water coolant ("wet" accidents) into the masonry occurred. As a result, radioactive contamination with a complex composition and volume distribution occurred. There were two sources of contamination: activation of impurities and inflow of radionuclides during the incidents. Quantitative estimates in both cases are difficult to make, since the data on impurities in graphite were incomplete and inaccurate and the inflow of radionuclides as a result of the incidents was random.

Masonry samples were obtained using an apparatus that could be moved from channel to channel. This apparatus made it possible to extract graphite cores at a given depth by horizontal drilling. Five samples at distances of 12 mm from one another were cut out, using a milling cutter, from the cores obtained. The samples were 8 mm in diameter and 2 mm thick graphite disks.

The vertical and radial distributions of radionuclides in the masonry and the local distribution of the radionuclides near the damaged cells had to be investigated to obtain the overall pattern

of contamination. At least 160-200 samples from each reactor needed to be analyzed to obtain such information.

#### 6.3.1.1 ß-emitting nuclides.

The β -spectrometry of graphite samples made it possible to draw the following conclusions:

- The C-14 activity in graphite is greater than the activity of all other radionuclides;
- Within the limits of measurement error, C-14 is distributed uniformly over the volume of an individual graphite block;
- The C-14 content in the graphite is linearly related with the neutron fluence.

The measured maximum specific activity of C-14 in samples of graphite taken from the reactor masonry is (MBq/g): 1.8 ± 0.4 for the I-1 reactor, 1.1 ± 0.2 for the EI-2 reactor, and 1.0 ± 0.2 for the ADE-3 reactor, and the computational value [30] is $4.74 \cdot 10^5$ Bq/g.

The measurements of the activity of C-14 at the center of the masonry and coefficients which take account of the variation of the neutron flux in the vertical and radial directions were used to estimate the C-14 content. The C-14 activity in I-1, EI-2, and ADE-3 masonries is as follows ($10^{15}$ Bq): 1.3 ± 0.3, 0.80 ± 0.17, and 1.2 ± 0.2, respectively.

The interaction of neutrons with Li-6 and N-14 makes the main contribution to the accumulation of tritium in the graphite. The triton formed remained, with a high probability, in the graphite matrix, since its average range is less than the grain size in graphite and greater than the pore size. But, some tritons, produced near grain boundaries, had insufficient energy to pass through the pores or the gap. Consequently, some of the tritium could remain in the gas filling the pores and gaps and escape with the gas from the masonry. The escape of tritium from graphite is probably due to oxidation of the graphite, as a result of which the tritium fixed in the graphite also was oxidized and escaped with the gas.

The average specific activity of H-3 in the reactor cladding was 3.4 ± 0.5 kBq/g for I-1, 5.4 ± 0.8 kBq/g for EI-2, and 4.9 ± 0.7 kBq/g for ADE-3. On the basis of the data obtained and the graphite mass in the cladding the H-3 content (in $10^{12}$ Bq) was estimated to be 4.6 ± 0.7 for I-1, 7.6 ± 1.1 for EI-2, and 9.6 ± 1.6 for ADE-3. The measurement results differ strongly from the numerical predictions [30]. The actual content of H-3 is approximately 200 times lower than

predicted; this can be explained by the escape of H-3 from the masonry during wet accidents. In addition, the difference found is due to the fact that in the calculation in [30] the value of the initial concentration of lithium in graphite was too high. According to the data obtained, the real concentration of lithium in the graphite of commercial reactors does not exceed $10^{-6}$ mass%, which is 10 times less than previously proposed.

In connection with the large effort required, only several samples of graphite were analyzed for the content of Cl-36 and Ni-63 in them. The measured activity was $2.5 \cdot 10^2$–$2.2 \cdot 10^3$ Bq/g for Ni-63 and 54–110 Bq/g for Cl-36. Comparing the specific activity, obtained experimentally and computationally, of Cl-36 in graphite shows that the calculation overestimates the content of this radionuclide by a factor of 100. This is due to two factors: the initial chlorine content used in the calculation was too high and the content in graphite during operation of the reactor was low. An estimate of the second factor was made in [31] and its value is ~10.

#### 6.3.1.2 *Actinides and fission products*

During the incidents, which occurred at the early stages of operation, an unknown quantity of uranium entered the reactor masonry. The uranium particles were spread by water vapor and settled on the graphite surface both on the side of the technological channels and in the gaps between the blocks. During prolonged neutron irradiation, the uranium adsorbed on the surface of the blocks was first converted into Pu-239, which largely burned up, partially converted into heavier actinides including americium and curium. The actinide content in the samples was determined by γ and x-ray spectrometric measurements.

To determine the isotopic composition, the plutonium contained in the graphite was separated from strongly contaminated samples and sources were prepared. Next, the spectrum in the range 10–200 keV was measured; Pu-238, Pu-240, Pu-239, and Pu-241 lines were observed here. The relative content was estimated.

The radiation of the graphite samples was also measured in the energy range 100–1500 keV using coaxial detectors consisting of ultra pure germanium. The spectra obtained contained lines of Cs-134, Cs-137, Ru-106, and Eu-154,155, and some other fission products.
Analysis led to the following conclusions:

- The samples taken from the surfaces of the blocks are most strongly contaminated by actinides and fission products, and it was established that a 2 mm thick graphite layer determines the surface contamination of the blocks;
- At the present time, the Cm-244 makes the greatest contribution to the activity of the actinide mixture;
- The distribution of actinides and fission products over the height of the cells is nonuniform, and the nonuniform (spotty) character of the contamination is probably explained by differences in the surface state of individual blocks (in the degree of destruction of the surface layer) and different gap widths between the blocks;
- The content of actinides and fission products in the samples which were taken from the cells next to the damaged cells is 10 times higher than in samples from cells far from the damaged cells;
- The specific activity ratios A (Am-243)/A (Am-241) and A (Cm-244)/A(Am-241) in samples taken at different heights are substantially different, which can be explained by the variation of the neutron flux density over the height of the cell (Table 11).

Analysis of the plutonium samples extracted from graphite 7–9 yr after the reactors were shut down showed that the isotopic composition is essentially the same for different reactors (Table 12). After the Pu-241 content was determined at the moment of the measurements, the amount of Am-241 produced in the period between reactor shutdown in the measurement was calculated. In addition, some Am-241 was already present in the graphite at the moment the reactor was shut down – the calculations show about 20% of the total content of Am-241 at the moment of measurement. The total value was used to determine the ratio Am-241/Pu-239.

Table 13 shows measurements of the content of individual fission products in the surface layer of graphite blocks from the reactor cores. The ratios Cs-134/Cs-137, Ru-106/Cs-137, and

**Sr-90/Cs-137 in different samples are essentially identical, indicating that the migration of fission products has very little influence. In Table 13 -**

**Table 15, and Table 17, the data are presented for a time 8 yr after reactor shutdown, and**

Table 18 presents data obtained 10 yr after shutdown.

**Table 11. Relative Content of Actinides Versus the Neutron Fluence.**

| Specific fluence | Reactor | A(Am-243)/A(Am-241) | A(Cm-244)/A(Am-241) |
|---|---|---|---|
| 0.997 | I-1 | 0.50 ± 0.12 | 430 ± 80 |
| | EI-2 | 0.49 ± 0.01 | 270 ± 20 |
| 0.71-0.78 | I-1 | 0.38 ± 0.06 | 230 ± 40 |
| | EI-2 | 0.31 ± 0.01 | 92 ± 3 |
| 0.5-0.55 | I-1 | 0.19 ± 0.04 | 74 ± 22 |
| | EI-2 | 0.22 ± 0.01 | 52 ± 3 |
| 0.4-0.47 | I-1 | 0.15 ± 0.02 | 42 ± 6 |
| | EI-2 | 0.08 ± 0.002 | 18 ± 1 |
| 0.19-0.23 | I-1 | 0.07 ± 0.04 | 14 ± 8 |

**Table 12. Average Ratio Between Plutonium Isotopes and Am-241.**

| Isotopic ratio | Experiment | | Calculation [30] |
|---|---|---|---|
| | I-1 | ADE-3 | |
| Pu-238/Pu-239 | 0.057 ± 0.004 | 0.055 ± 0.009 | 0.099 |
| Pu-240/Pu-239 | 0.65 ± 0.04 | 0.68 ± 0.08 | 1.16 |
| Pu-241/Pu-239 | 0.19 ± 0.01 | 0.28 ± 0.02 | 0.28 |
| Am-241/Pu-239 | 0.13 ± 0.01 | 0.14 ± 0.02 | 0.18 |

**Table 13. Specific Activity of Cs-137, Ru-106, and Sr-90 in Graphite Samples Taken from the Surface of Graphite Blocks.**

| Reactor | Cell No. | Conditional fluence | Cs-137, $10^5$ Bq/g | Ru-106, $10^3$ Bq/g | Sr-90, $10^4$ Bq/g | Ru-106/Cs-137, $10^{-3}$ | Sr-90/Cs-137 |
|---|---|---|---|---|---|---|---|
| EI-2 | 3425 | 0.46 | 1.38 | 0.65 | 7.4 | 4.8 | 0.54 |
| | | 0.96 | 1.23 | 0.6 | 4.7 | 4.9 | 0.38 |
| | | 0.61 | 32.3 | 14.6 | 160 | 4.5 | 0.48 |
| I-1 | 3644 | 0.55 | 5.62 | 1.9 | 29 | 3.5 | 0.52 |
| | | 0.82 | 3.00 | 1.2 | 17 | 3.8 | 0.57 |
| | | 0.80 | 1.00 | 0.30 | 7.5 | 3.0 | 0.75 |

* **Note.** The error does not exceed 20%.

**Table 14. Correlation Coefficients.**

| Reactor | Cs-137/Am-241, $10^2$ | Cs-137/Cm-244 |
|---|---|---|
| I-1 | 2.0 ± 0.3 | 4.2 ± 1.1 |
| EI-2 | 2.5 ± 0.3 | 4.2 ± 0.8 |
| ADE-3 | 1.8 ± 0.2 | 6.5 ± 0.7 |

**Table 15. Content of Certain Radionuclides in Reactor Masonry, Bq.**

| Radionuclide | I-1 | EI-2 | ADE-3 |
|---|---|---|---|
| Co-60 | $(2.6 \pm 0.6)\ 10^{12}$ | $(4.1 \pm 1.0)\ 10^{12}$ | $(8.2 \pm 2.0)\ 10^{12}$ |
| Cs-137 | $(6.9 \pm 1.5)\ 10^{12}$ | $(2.1 \pm 0.5)\ 10^{12}$ | $(8.8 \pm 1.9)\ 10^{12}$ |
| Sr-90 | $(4.3 \pm 1.0)\ 10^{12}$ | $(1.1 \pm 0.3)\ 10^{12}$ | $(3.1 \pm 0.7)\ 10^{12}$ |
| Am-241 | $(3.4 \pm 0.8)\ 10^{10}$ | $(8.4 \pm 2.2)\ 10^{9}$ | $(5.0 \pm 1.1)\ 10^{10}$ |
| Cm-244 | $(1.6 \pm 0.6)\ 10^{12}$ | $(5.1 \pm 1.6)\ 10^{11}$ | $(1.4 \pm 0.3)\ 10^{12}$ |
| Pu-238 | $(7.4 \pm 1.9)\ 10^{10}$ | $(1.8 \pm 0.5)\ 10^{10}$ | $(1.2 \pm 0.3)\ 10^{11}$ |
| Pu-239 | $(4.5 \pm 1.2)\ 10^{9}$ | $(1.1 \pm 0.3)\ 10^{9}$ | $(7.3 \pm 1.9)\ 10^{9}$ |
| Pu-240 | $(1.1 \pm 0.3)\ 10^{10}$ | $(2.8 \pm 0.8)\ 10^{9}$ | $(1.8 \pm 0.5)\ 10^{10}$ |
| Pu-241(β activity) | $(1.6 \pm 0.4)\ 10^{12}$ | $(3.9 \pm 1.1)\ 10^{11}$ | $(2.5 \pm 0.7)\ 10^{12}$ |

**Table 16. Ratios of the Computed and Measured Contents of Certain Radionuclides.**

| Ratio | I-1 | EI-2 | ADE-3 |
|---|---|---|---|
| $Cs_c$-137/$Cs_m$ 137 | 9.1 | 13.1 | 6.5 |
| $Cm_c$-244/$Cm_m$-244 | 12.7 | 16.5 | 15.0 |
| $Am_c$-241/$Am_m$ -241 | 10.9 | 19.7 | 6.8 |
| $Pu_c$-239/$Pu_m$-239 | 8.1 | 14.6 | 4.6 |

**Table 17. Radionuclide Content in I-1 Reactor Masonry, Bq.**

| Radionuclide | Experiment | Calculation [30,32] |
|---|---|---|
| C-14 | $(1.3 \pm 0.3)\ 10^{15}$ | $3.5 \cdot 10^{14}$ |
| Co-60 | $(2.6 \pm 0.6)\ 10^{12}$ | $4.4 \cdot 10^{14}$ |
| Cs-137 | $(6.9 \pm 1.5)\ 10^{12}$ | $6.4 \cdot 10^{13}$ |
| Pu-239 | $(4.5 \pm 1.2)\ 10^{9}$ | $3.6 \cdot 10^{10}$ |

Analysis of the data shows the following scheme of contamination of the masonry by actinides and fission products:

- As a result of the incidents uranium particles were spread throughout the masonry and became fixed on the surface of the blocks;

- Subsequent prolonged neutron irradiation produced the composition of the contamination; the observed relation between the isotopic ratio and the neutron fluence shows that the movement of radionuclides during irradiation was small.

**Table 18. Radionuclide Content in the Masonries of Domestic and Foreign Reactors, Bq.**

| Reactor | C-14 | H-3 | Co-60 |
|---|---|---|---|
| I-1 | $1.3 \cdot 10^{15}$ | $2.1 \cdot 10^{12}$ | $2.0 \cdot 10^{12}$ |
| EI-2 | $8.0 \cdot 10^{14}$ | $3.3 \cdot 10^{12}$ | $3.1 \cdot 10^{12}$ |
| DR [33] | $1.2 \cdot 10^{14}$ | $7.6 \cdot 10^{13}$ | $9.8 \cdot 10^{14}$ |
| AGR [34] | $1.9 \cdot 10^{14}$ | $1.0 \cdot 10^{14}$ | $1.6 \cdot 10^{14}$ |

Although the ratio Cs-137/Am-241 (Cs-137/Cm-244) varies along the height of the cell as a function of the neutron flux, because of the strong differences in the degrees of contamination of the surface of different blocks it is impossible to take account of this dependence when estimating the content. Consequently, a different approach was used: first the average ratio of Cs-137/Am-241 (Cs-137/Cm-244) was determined and then the value obtained was used. This value for different cells in different reactors is close (Table 14). Since direct measurements of actinides can be made only with a high degree of contamination, their content for a low degree of contamination was determined from Cs-137 measurements performed using correlation relations.

**The following scheme was used to estimate the content of actinides and fission products in the reactor masonries. It was assumed that these radionuclides are contained in a 2 mm thick surface layer on the graphite blocks. All cells in the masonry were divided into three categories according to the degree of contamination: close to the damaged and damaged cells, far from damaged cells in the central zone of the reactor masonry, far from the damaged cells in the peripheral zone and the reflector. For each group of cells, the average content of Cs-137 was calculated and the average content of other radionuclides was found using the correlation coefficients (**

Table 15). An attempt was made to estimate the content of actinides and fission products in the masonries of commercial reactors in [30, 37]. Some results of these works are presented, together with estimates made in the present work, in Table 16. Analyzing these results together led to the following conclusions:

- The absolute content obtained from measurements differs strongly from the calculations performed in [30, 37]; the reason for the discrepancies are most likely errors in the

calibration during neutron measurements, serving as the basis for the computational estimates;

- Experimentally obtained relations between the content of individual radionuclides are consistent with their computational predictions;
- The experimental relations between the content of these radionuclides in different reactors do not differ much from the computed relations.

Co-60 radionuclide was produced inside graphite blocks as a result of impurity activation and in a surface layer as a result of activation and transport with the steam–gas mixture during the incidents. Its content in the samples was measured by the γ -spectrometry method. Analysis led to the following conclusions:

- In certain cases the activity of the internal samples taken from different cores and sometimes from the same core differed several fold, which can be explained by the nonuniform distribution of cobalt in the graphite;
- The level of contamination of the surface of the graphite blocks is approximately the same for the entire masonry;
- Although the Co-60 activity in the samples taken from the surface of the graphite blocks is higher than the samples taken from the interior volume, the average ratio between the specific activity of the surface and internal samples is small: 4.7 for EI-2, 3.2 for I-1, and 3.1 for ADE-3; consequently, the Co-60 present in the interior volume of graphite blocks makes the main contribution to the content of this radionuclide in the masonry;
- The Co-60 content in the blocks from the center of the masonry and from the periphery of the core is the same to within the limits of error; the Co-60 content in the volume of the blocks of the reflector in the EI-2 reactor is approximately two times smaller than in the core blocks.

**On the basis of the data obtained, the Co-60 content in core graphite is estimated as the sum of the contribution of the surface and volume contamination (see**

Table 15).

The main results of these investigations are as follows:

- C-14 makes the determining contribution to the radioactive contamination of graphite;

- No regularities in the H-3 distribution could be established; diffusion removal with steam during flooding of the masonry decreased its content in the graphite;
- Of the two sources of contamination of the cladding by Co-60 – inflow during flooding and activation of impurities – the latter predominated; the difference in the initial content of the impurity Co-59 in individual blocks is so large that it masks the dependence of the Co-60 contamination on the neutron distribution in the masonry;
- Contamination of the masonry by fission products and actinides is of a surface and nonuniform character, and the content of the substances near the damaged cells is much higher than far from the latter;
- A correlation was found between Cs-137 and actinides; the correlation coefficients were determined and used to estimate from Cs-137 measurements the content of plutonium, americium, and curium in samples from slightly contaminated cells;
- It was confirmed that the content of important radionuclides (C-14, Co-60, Cs-134) in graphite could be estimated computationally provided that reliable data were available on the initial concentration of the corresponding impurity elements.

As a rule, the error of individual measurements was several percent, but in certain cases strong differences were observed between the contaminations of various samples. Consequently, the uncertainty of the average content of a radionuclide in a cell could reach 50–100% (Table 17). In reality the content of all radionuclides differs strongly from the predicted content. Fission products, actinides, tritium, and Co-60 are present in masonry in quantities tens and hundreds of times less than previously assumed, and this can substantially simplify the problem of disassembling masonry and decrease the dose load to participants. However, the C-14 content is 2–3 times greater than expected, which makes it more difficult to manage spent graphite. Comparing masonries from the three reactors, which were shut down, gave similar data on the radionuclide distribution and the degree of contamination of the graphite. Thus, the information obtained also characterizes the situation at other commercial uranium–graphite reactors, including those, which are not covered by the data obtained in this investigation. The results concerning the accumulation of radionuclides as a result of impurity activation is suitable for estimating the contamination of RBMK graphite.

**According to**

Table 18, the masonry of Russian reactors contains approximately seven times more C-14 than foreign reactors. This is explained by the fact that nitrogen was used as the protective gas preventing oxidation of the graphite, whereas in foreign reactors carbon dioxide was used. The differences in the content of other radionuclides could be due to differences in the composition of the impurities and the specific features of the operation of the reactors.
The radionuclide content in individual blocks varies manifold, so that sorting by degree of contamination can be used to make a decision concerning the storage of the blocks. The data, obtained in the present work, on correlations between individual radionuclides can be used to develop a fast sorting method.

The contamination of graphite bushings is now being studied. These are replaceable components, which are off-loaded from the three indicated reactors. The mass of the spent bushings is comparable to that of the graphite in the masonries. However, the contamination of most of these bushings should be much lower because of the relatively shorter irradiation time and the absence of incidents in which the fuel channels were destroyed in the subsequent 20 years of operation of the reactors. Consequently, different time periods and technology can be chosen for handling bushings. For example, the contamination of bushings from cell No. 1326, which was located far from the damaged cells, in the EI-2 reactor was investigated. The bushings were located in the reactor during the last two years of its operation. Compared with graphite from blocks of the same cell the content of C-14 was approximately 13 times lower, the content of Cs-137 was 320 times lower, and the content of Co-60 was 2.3 times lower; no actinides were found. It is known that some storage sites containing bushings together with other radioactive wastes are partially filled with water, which could result in a redistribution and escape of radionuclides into the environment. This question requires special investigation.
The variant currently being examined for handling spent graphite includes the following:

- A holding period prior to salvaging;
- Keeping the graphite masonry in the reactor building during the holding period;
- Continuation of operation of some storage sites for graphite bushings (if necessary, improving the storage sites);
- Reloading graphite bushings and other radioactive wastes from a different part of the storage sites, which do not meet the modern requirements.

The results of the investigations of graphite contamination will be used to determine the time required to disassembly masonry, make decisions concerning individual storage sites, develop a method for fast sorting of spent graphite components according to their degree of contamination, and plan similar measures for other reactors.

In [35] discussing the Russian measurements performed on a number of graphite samples from the plutonium production reactors at Tomsk 7 such important observations relating to residual radionuclide concentrations and distributions from this comprehensive experimental study were made:

- About 500 samples have been taken from the graphite stack of the I-1, ADE-3 and EI-2 reactors and assayed. Contamination of these samples with radionuclides H-3, C-14, Cl-36, Co-60, Ni-63, Sr-90, Ba-133, Cs-134,137, Eu-152,154,155, Pu-238, Pu-239,240, Am-241,243, Cm-244 and some others have been determined;
- It was discovered in this study that the dominant activity in the graphite is C-14, and its distribution in the graphite stack is a reflection of the thermal neutron flux. The concentration of C-14 in the graphite from this Tomsk reactor was about 6 times higher then in the graphite from a similar Hanford reactor. A significant fraction of the C-14 activity in reactor graphite was indeed due to the presence of nitrogen in graphite nitrogen cover gas);
- The tritium content in the reactor graphite has been measured, and the content appeared to be very small (about several hundred times smaller) than prediction. H-3 is distributed non-uniformly in the graphite stack;
- The dominant fraction of the Co-60 concentration in reactor graphite is due to the presence of original Co-59 impurity in graphite;
- It was found that actinides and fission products are concentrated on the block surface (within a thickness of 2 mm). The penetration of radionuclides from block surface to its volume is very small. There is the correlation of specific activities between the fission products. Actinide distribution is a reflection of the neutron flux.

Some of these measurements are made by indirect methods and the quality of some of the results in not completely clear. For this reason there is a need for an international benchmark exercise so that standards of measurement technique can be set.

### 6.3.2 Production enterprise Mayak

The results of investigations of the radiation characteristics of graphite samples taken from the masonry in two commercial uranium-graphite reactors at the Mayak Industrial Association are presented in [36]. The specific activity of α, β and γ emitters present in the graphite in different parts of the masonry, including cells located next to the damaged cell, is determined. The specific activity of the main product of activation $^{60}Co$ is $10^2$-$10^4$ Bq/g. The concentration of Cs-137 in the graphite is higher than that of any other fission product (the specific activity in the cells located next to the damaged cell is $10^4$-$10^6$ Bq/g) Cm-244 makes the largest contribution to the total activity of transuranium elements; its activity in one sample was $2 \cdot 10^7$ Bq/g. More than 90% of the activity of the masonry graphite is due to C-14. The outer and inner surfaces of the graphite bricks are most highly contaminated with radionuclides. The results obtained are the basis for the high estimated content of radionuclides in the entire graphite masonry for each reactor.

All commercial uranium-graphite reactors, which operated at power in 1948-1952, were shut down in 1987-1990. In the AV-1 reactor, where accidents with fuel entering the graphite occurred in 1960 and 1975, there are nine damaged cells and in the AV-3 reactor there are four cells (the accidents occurred in 1955, 1957, and 1958). In 2002, specialists at the Siberian Integrated Chemical Plant and the Mayak Industrial Association obtained graphite samples from six cells in the AV-1 reactor and five cells in the AV-3 reactor, including one cell, which is located next to the damaged cell in each reactor. Measurements of the activity of the samples characterize the local contamination of the graphite and make it possible to establish relations for estimating the radionuclide contamination of the graphite cladding as a whole.

The samples (core samples) were taken from several points along the height in each cell (up to six). The thin walls of the graphite bricks were drilled through. Five samples were fashioned from each of the core samples to study the radionuclide distribution over the thickness. In all, 70 samples were obtained from the masonry bricks in the AV-3 reactor

and 120 samples from masonry bricks in the AV-1 reactor; the activity of γ-emitting nuclides from 180 samples, β-emitting Sr-90 + Y-90 from 130 samples, and α-emitters from 30 samples was measured.

The radionuclides which at present make the greatest contribution to the contamination of the graphite can be divided into the following groups: products of activation (Co-60 dominates), actinides, fission products, and long-lived β emitters, primarily, C-14, Sr-90, and H-3.
The β emitter C-14 makes the main contribution to the total activity of the masonry graphite in the reactors. Its activity was measured by liquid scintillation radiometry. The method is based on burning graphite samples in oxygen to convert the carbon into a gaseous form followed by absorption of carbon dioxide by a NaOH solution and measuring the activity of C-14 with a UGR-1 radiometer (Russia). The C-14 activity was measured for 15 samples of graphite from the masonry in each reactor. It was determined that the specific activity of the graphite is $3 \cdot 10^5$ -$1.8 \cdot 10^6$ Bq/g.

The results obtained show the following:

- The C-14 activity in the samples obtained from the same core sample is essentially identical to that of the other radionuclides, for which the difference in activity reaches factors of 10-100;
- The C-14 activity in core samples corresponds, to within the error limits, to the neutron fluence at points of the masonry from which these samples were extracted.

After the reactors were shut down, one of the main sources of γ radiation in the reactor graphite is Co-60. The main formation channel for this radionuclide is the reaction Co-59 (n, γ)Co-60; Co-59 is present as an impurity in unirradiated graphite and can appear when Fe-59 produced in the reaction Fe-59(n, γ) Fe-59 decays. The source of Co-60 in graphite is activation of impurities (over the entire body of the graphite bricks) and deposition on the surface of the bricks of products of corrosion, which enter the masonry with the coolant when the technological channels rupture.

The activity of γ emitters was measured using a SEG-01 PPD γ spectrometer (Russia) equipped with a Ge(Li) detector. Co-60 with the following activity was observed in all samples on which

measurements were performed: $1.5 \cdot 10^2$-$4.8 \cdot 10^4$ Bq/g for the AV-3 reactor and $6.9 \cdot 10^2$-$3.2 \cdot 10^4$ Bq/g for AV-1. The contamination was nonuniform over the thickness of the graphite bricks. In 70% of all samples, the highest Co-60 activity is observed near the inner surface of a brick; in all other samples, it is observed near the outer surface.

No large differences in Co-60 content in the graphite of cells located next to the damaged cell and cells located away from the accident site were observed. In the AV-1 reactor, the contamination of graphite at the center of the masonry and the peripheral cells by Co-60 is almost identical. At the periphery of the AV-3 reactor, the content of this radionuclide is several-fold lower than at the center. No clear relation between the Co-60 content and the sampling marker could be found.

In summary, the influence of thermal-neutron fluence at the sampling points is modified by the different concentration of Co-59 impurity and by the deposition of corrosion products containing cobalt on the surface of the graphite bricks. Nonetheless, the measurement results permit constructing computational models and estimating the Co-60 quantity in the entire graphite masonry, dividing the masonry conditionally into a more highly contaminated layer near the surface of the bricks and the less contaminated interior volume of the graphite.
The radionuclide Cs-137 is present in almost all graphite samples. Its specific activity varies from $10^4$-$10^6$Bq/g in cells located next to the damaged cells to $10^2$-$10^3$ Bq/g in cells located far from the damaged cells. The Cs-137 content in graphite is nonuniform over the thickness of a brick. In 70% of samples from the AV-1 reactor and 90% of samples from the AV-3 reactor, the highest activity is recorded on the outer surface of the bricks and in all other samples on the interior surface.

The difference in location of maximum activity of Co-60 and Cs-137 is due to the fact that migration mechanisms of these radionuclides are different. Cesium is the more mobile element, especially at high graphite temperature (700-800°C), and propagates along gaps in the masonry together with nitrogen purge flow, subsequently settling on the surface of the bricks. Cs-134 is also present in appreciable quantities in samples with a high Cs-137 content; the Cs-134 activity is approximately 100 times lower than that of Cs-137. In peripheral cells,

Cs-134 is absent or its activity is low – 10-100 Bq/g. The highest contamination with Cs-134 is found on the outer surface of the graphite bricks.

The presence of the products of fission of Eu-154 and Eu-155 with specific activity reaching $10^4$ Bq/g was recorded in individual samples.

The activity of the β emitters Sr-90 + Y-90 was measured with a SEB-02STs scintillation spectrometer (Russia). It ranged from $2.8 \cdot 10^4$ to $7.5 \cdot 10^6$ Bq/g.

Transuranium elements were found in the most contaminated samples from cells located next to the damaged cell. Ordinarily, when measurements are made on such samples the γ spectrometer is recording Am-241. The measurements of the α-emitting radionuclides in the samples were conducted with a SEA-01 PPD semiconductor spectrometer. The targets for such measurements were prepared by depositing on a substrate solution, which were obtained by burning samples of graphite placed in a crucible or quartz boats, and then dissolving the ash residue in nitric acid. It was determined that Cm-244 makes the largest contribution (from 83 to 98%) to the total a activity of transuranium elements; Pu-238, Pu-239, Am-241 were also found, but their fraction is ten times lower and is largely masked by the background when measurements are performed with an α spectrometer. The highest actinide content is observed on the outer surface of the bricks. The contamination with these actinides on the interior surface is also 100 times higher than in the central layers of the bricks.

The maximum specific activity of Cm-244 observed in AV-1 and AV-3 reactor graphite is $6.5 \cdot 10^5$ and $2 \cdot 10^7$ Bq/g, respectively. This is because the accident at the location of the sampling in the AV-1 reactor occurred 20 years later than in the AV-3 reactor, and the Cm-244 content is proportional to the neutron fluence raised to the power 4.3.

It should be noted that some of the results of studies of the radionuclide content in graphite in the reactors at the Mayak Industrial Association and the Siberian Integrated Chemical Plant agree with one another. These include the following:

- The surface layers of the graphite bricks are most highly contaminated by radionuclides;

- The contribution of Cm-244 to the total activity of a emitters predominates;
- The activity of transuranium elements can be determined only in the most contaminated samples obtained near the damaged cells.

This means that the content of radionuclides in all of the masonry can largely be estimated using the approaches employed for the reactors at the Siberian Integrated Chemical Plant. At the same time, a smaller diversity of radionuclides was observed in the graphite in the AV-1 and AV-3 reactors than in the I-1, EI-2, and ADE-3 reactors. This is because a substantial amount of time elapsed (12-13 yr) between the time when the reactors were shut down and studies of the graphite began at the Mayak Industrial Association, and during this period of time the radionuclides with relatively short half-lives decayed.

# 7 Treatment/Conditioning Techniques/Process Used on Retrieved I-Graphite

To prevent activity leaching from graphite waste, many options have been proposed worldwide. These include graphite coating, encapsulation in various matrix materials, sealing graphite into containers, etc.

As it is indicated in [37] the concept of the storage of the graphite masonry of the graphite reactors in situ isolated from the environment for about of 100 years is adopted in Russia. In relation with this, development of the technologies for the treatment of the irradiated graphite was not the primary task. So, today there is not industrially tested technology for the treatment of the irradiated graphite in Russia. But there is a problem of the management so called graphite sleeves that are under storage in different storage facilities at the reactor sites. Also during maintenance period's significant amounts of the graphite from graphite masonry were retrieved and replaced with new ones. There are three main directions in the management development of the technologies for management of the irradiated graphite in Russia:

- Methods to establish engineering barriers for the long-term storage of graphite in the form of solid radwaste.
- Methods of graphite chemical transformation for the purpose of the separation of the dose-generating isotopes and the reduction of the volumes for the further storage;
- Incineration of the irradiated graphite.

## 7.1 Immobilization of the radionuclides in the irradiated blocks [38]

In the paper [38] the results of investigations on the immobilization of radionuclides by the cold-hardening fixative "atomik" in samples of GR-280 graphite from the graphite column extracted from the No. 3 unit of the Leningrad nuclear power plant after 17 years of operation are presented. The graphite blocks were contaminated (primarily, from the surface) by fission products and fuel as a result of rupturing of the technological channel and part of the fuel from the unsealed fuel assemblies entering the masonry.

The proposed fixative is an experimental development (TU 2257-18826195-01). It contains an epoxy oligomer compacted with a furane heterocyclic alydehyde, a fill material, and the target additive. The composition solidifies in the presence of special aromatic amines.

This is the first investigation performed in Russia of the possibility of immobilizing radionuclides using the fixative "atomik" by permeating real samples of reactor graphite extracted from RBMK masonry. The samples were permeated with the fixative under pressure 20-30 mm of a water column followed by molding under pressure ~0.1 MPa. The leaching tests were performed in distilled water after a holding period of 24 months at temperature 20 ± 2°C and 12 months at 50°C. No radionuclides were found to escape from the cured samples into the water samples.

In order to use a fixative to fix radionuclides, the properties of the fixative must remain unchanged for a long period of time under irradiation. To validate the use of "atomik" as a fixative for radionuclides, the radiation resistance of "atomik" was investigated. This yielded data on the influence of the γ irradiation dose on the change in the physical-mechanical properties and radiation gas release. A fixative with (20% soot) and without fill was used for the tests.

### 7.1.1 Physical-mechanical tests

The strength under compression, the fracture deformation, and the elastic modulus were investigated in order to assess the radiation resistance of the fixative. In addition, the fixative was examined visually for the presence of cracks, cleavages, and so on. The maximum γ -ray exposure dose was 3000 Mrad. At this irradiation dose, the samples of the fixative maintained their integrity and had no cracks.

Analysis shows that the strength of the samples of fixative decreases only with irradiation dose about 15000 Mrad. According to preliminary estimates, this converts to an average specific activity of RBMK masonry equal to 300-500 yr holding period.

### 7.1.2 Radiation gas release

The radiation-chemical yield of gaseous products of radiolysis in vacuum from the samples of the fixative is $12 \cdot 10^{-10}$ $cm^3/(g \cdot rad)$; chromatographic analysis of the composition of the gases released showed that mainly hydrogen is released.

The experiments established that α, ß, and γ-emitting radionuclides are leached from samples of graphite extracted from the reactor, and this process proceeds continually. When the irradiated graphite samples were permeated with the fixative "atomik," no radionuclides were released at 20°C for 2 yr; at 50°C there was no release for 1 yr. Testing is continuing.

## 7.2 The technology of removing INF spills from damaged irradiated reactor graphite of the uranium-graphite reactors subject for decommissioning [39]

During operation of graphite reactors there were incidents with spilling of irradiated nuclear fuel (INF) to the graphite stack. Graphite waste contaminated with INF spills can be classified as high-level waste with fissile materials (HLW FM). In accordance with the regulatory requirements applicable to the nuclear power, such waste shall be immobilized, i.e. converted to the stable physical and chemical form. However, the specific feature of such waste is the substantially high ratio of graphite volume (mass) to INF spill volume (mass) – in excess of $10^3 \ldots 10^4$. In order to reduce volume of waste, it is expedient to separate INF spills from the other main portion of graphite.

To resolve this problem, NIKIET under the contract with «Rosenergoatom» concern has been developing the technology of INF spills removing from irradiated reactor graphite.

### 7.2.1 The radiation characteristic of damaged graphite containing INF spills

Long lived activation products: Be-10, C-14, Cl-36, Ca-41, Ni-59, Ni-63, Ag-108m, etc.

Long lived fission products: Sr-90, Y-90, Ru106, Cs-134, Cs-137, Pm-147, etc.

Actinides: U-235, U-236, U-238, Np-239, Pu-238, Pu-239, Pu-240, Pu-241, Pu-242, Am-241, Am-242m, Am-243, Cm-242, Cm-243, Cm-244, etc.

The physical and chemical form of INF spills contained in graphite varies from metals to oxides, hydroxides and other more complicated compounds.
Therefore the technology proposed for conditioning of damaged graphite with INF spills should provide the comprehensive solution, namely: retaining of as much radionuclides as possible, particularly actinides which are found in such graphite in the form of various compounds.

Table 3 presents the information on the radiation risk estimation of some radionuclides.

**Table 19. Radiation Risk Estimation of Some Radionuclides in Contaminated Graphite**

| Radionuclide | Specific activity a, Bq/kg | Type of inhalation compound | Dose coefficient (NRB-99) D, Sv/Bq | Radiation risk D x a, Sv/kg |
|---|---|---|---|---|
| C-14 | $8 \cdot 10^{8}$ | C<br>$CO_2$<br>CO | $5.8 \cdot 10^{-10}$<br>$6.2 \cdot 10^{-12}$<br>$8.0 \cdot 10^{-13}$ | 0.46<br>$4.96 \cdot 10^{-3}$<br>$6.4 \cdot 10^{-4}$ |
| Cl-36 | $2.3 \cdot 10^{7}$ | Compound with H, Li, Na, K, Rb, Cs, Fr<br>Other compounds | $3.4 \cdot 10^{-10}$<br>$6.9 \cdot 10^{-9}$ | $7.8 \cdot 10^{-3}$<br>0.16 |
| Sr-90 | $5.8 \cdot 10^{7}$ | All compounds (except $SrTiO_2$) | $2.4 \cdot 10^{-8}$ | 1.39 |
| Cs-137 | $1.1 \cdot 10^{8}$ | All compounds | $4.8 \cdot 10^{-9}$ | 0.53 |
| Pu-239 | $1.0 \cdot 10^{6}$ | Oxides, hydroxides<br>Other compounds | $4.7 \cdot 10^{-5}$<br>$1.5 \cdot 10^{-5}$ | 47<br>15 |
| Pu-240 | $1.3 \cdot 10^{6}$ | Oxides, hydroxides<br>Other compounds | $4.7 \cdot 10^{-5}$<br>$1.5 \cdot 10^{-5}$ | 61.1<br>19.5 |
| Am-241 | $1.1 \cdot 10^{7}$ | All compounds | $3.9 \cdot 10^{-5}$ | 429 |

### 7.2.2 The solution proposed for conditioning of damaged graphite with INF spills

Separation of INF spills from the main portion of graphite using the technology of molten salt oxidation (MSO). It is important to note that conversion (oxidation) of actinides ($UO_2$, $PuO_2$),

and subsequent vitrification of spent molten salt containing INF spills could be potentially carried out at the same MSO plant.

### 7.2.3 Advantages of MSO Technology

- Maximum capability for retaining radionuclides and heavy metals (up to 99.9% and higher)
- Lower operating temperature (750-900ºC) as compared to combustion of graphite in flame furnace
- Reprocessing of graphite without its preliminary crushing
- Satisfactory dry cleaning of gases without scrubbers
- Neutralization of acid gases
- Lower flow rates of flue gases as compared to combustion in flame furnace

The flow diagram is presented in Figure 6-1.

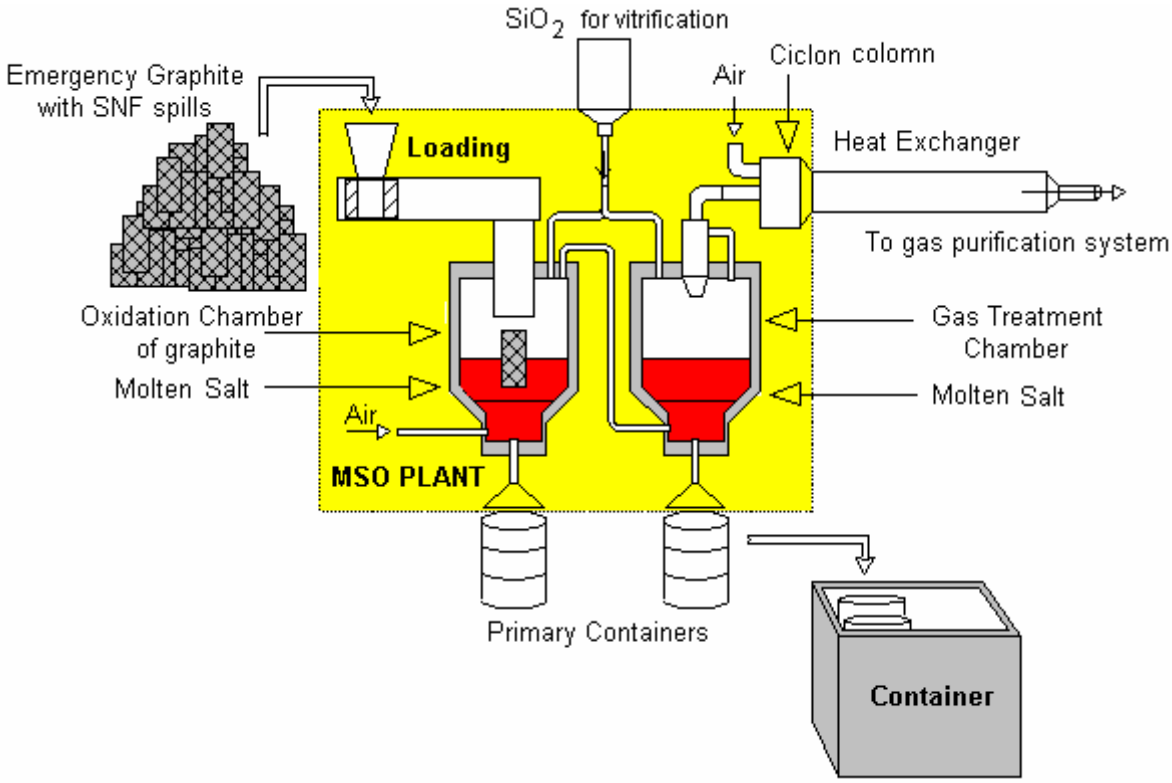


**Figure 7-1. Flow Design of Contaminated Graphite Treatment.**

Main parameters and economic efficiency of the technology is presented in Table 20.

**Table 20. Technical and Economic Efficiency of Proposed Technology**

| Parameter | | Molten salt oxidation and vitrification of HLW, disposal of LLW in metallic containers | Disposal of HLW in sic containers, Disposal of LLW in metallic containers | Disposal all graphite in ferroconcrete containers |
|---|---|---|---|---|
| 1. Total amount of graphite: | $T/m^3$ | 1637/992 | | |
| 2. HLW | $T/m^3$ | 27.29/16.55 | | - |
| 3. LLW | $T/m^3$ | 1609.71/975.6 | | - |
| 4. Packing radwaste: Sic containers V=44 $dm^3$<br>Metallic containers $v=lm^3$<br>Ferro-concrete containers<br>Containers for salt (after vitrification) | | -<br>1973<br>-<br>1 | 376<br>1973<br>-<br>- | -<br>-<br>1263<br>- |
| 5. Radwaste volume for disposal: | | | | |
| -Near surface disposal | $m^3$ | 1973 | 1973 | - |
| -Deep geological disposal | $m^3$ | 5 | 40 | 4585 |
| 6. Production costs:<br>-Total | Mil. Rub. | 289.5 | 363.0 | 1251.8 |
| -Graphite conditioning | Mil. Rub. | 97.9 | 162.8 | 55.4 |
| -Storage during packing | Mil. Rub. | 6.8 | 6.8 | 6.4 |
| -Temporary storage | Mil. Rub. | 62.1 | 62.1 | 62.1 |
| -Near surface disposal | Mil. Rub. | 121.5 | 121.5 | - |
| -Deep geological disposal | Mil. Rub. | 1.2 | 9.8 | 1127.9 |
| 7. Summary cost | Mil. Rub. | 339.1 | 405.0 | 1287.7 |
| 8. Cost ratio | - | 1.0 | 1.2 | 3.8 |

## 7.3 Other immobilization technologies [40]

Russian technologists propose that graphite falling into ILW or LLW categories should be impregnated with a sealant (known only as “F preservative”) or an inorganic phosphate.

An interesting simple immobilization method for graphite contaminated with uranium and actinides has been proposed in the Russian Federation also. After milling the graphite, powders of Al and oxides of Y, Ce, Ti are added; then, after some initial heat, a self-propagating high temperature synthesis is produced in hermetic steel containers. This process is similar to that of the thermite process. The resulting product (of density 2-4 $g/cm^3$) has a structure $TiCa_{0.9}N_{0.1}+Al_2O_3+Y_3Al_5O_{12}$. During high temperature synthesis, atoms of Y can be replaced with

uranium and actinide atoms. The product is a stable carbide-oxide composite material, ready for disposal. C-14 has also been successfully locked into this structure. This technology is claimed to be fully ecologically safe.

Russian technologists are developing a special procedure for dealing with graphite, which is classified as high-level waste as a result of contamination with fuel and fission products following fuel failures in its production reactors and also at the Beloyarskaya NPP reactors. This involves the so-called Self-Propagating High Temperature Synthesis process similar to the classical "Thermit" process, in which graphite is intimately mixed in stoichiometric proportions with aluminium and titanium oxide according to the equation

$$3C + 4Al + 3TiO_2 = 2Al_2O_3 + 3TiC \quad (1)$$

The reaction is initiated electrically and is thereafter self-propagating. It has been demonstrated on the laboratory scale and it is planned to build a plant, presumably at NIKIET Sverdlovsk (adjacent to Beloyarskaya NPP). It has the advantage of immobilising all significant isotopes present in the oxide and carbide matrices (including C-14 in the latter) and results in a highly unreative and insoluble product with very good leaching characteristics.

Another interesting but simple immobilization method was evaluated by the Russian Federation for immobilization of graphite contaminated with uranium and actinides. After milling the graphite, powders of Al and oxides of Y, Ce and Ti are added. Next, after some initial heat, a self-propagating high temperature synthesis is produced in hermetic steel containers. This process is similar to that of the thermite process. The resulting product is a stable carbide-oxide composite material, ready for disposal. C-14 has also been successfully locked into this structure. This technology is claimed to be fully ecologically safe.

## 7.4 Carbon isotope separation methods [41]

Currently the most effective industrial method of stable isotope separation is the cryogenic rectification of liquefied carbon monoxide (CO). It is required 200t of liquefied nitrogen to process of 1 ton of carbon (1 liter of nitrogen = 1 Euro).

$$R_2N^{12}CO + {}^{13}CO_2 \Leftrightarrow R_2N^{13}CO + {}^{12}CO_2 \quad (2)$$

$\alpha = 1.01$

Reaction between carbon-dioxide and amine carbomats (as example: dibutyl amine dissolved in benzol). The process is at normal conditions (atmospheric pressure and room temperature).

## 7.5 Burning of graphite [42]

The possibility of liquidating irradiated graphite by burning is examined by analyzing the computed and experimentally obtained information about the activity of C-14 produced over the period of operation of uranium-graphite reactors and appearing naturally in the Earth's biosphere. In nature, C-14 activity fluctuates in the range ±1%. This work employs the criterion that the technogenic addition of C-14 activity as a result of burning irradiated graphite should not exceed the fluctuations of the natural activity. It is shown that under this criterion an amount ~$3.5 \cdot 10^4$ tons/yr can be burned with $^{14}C$ activity in irradiated graphite ~$1.8 \cdot 10^{11}$ Bq/ton. Burning the irradiated graphite has already been tested in France on a low-capacity experimental facility (hundreds of t/yr). However, the following factors prevent wide adoption of the method:

- Atmospheric emissions of $CO_2$ with isotopic ratio C-14/C-12 which is $10^6$ times greater than that found in nature;
- The amount of irradiated graphite in the world is large (> 100,000 tons);
- The requirements mandating the elimination of risk due to all radionuclides are stringent. The first factor is the main reason.

The C-14 activity in the biosphere is $3.4 \cdot 10^{17}$ Bq. Taking account of the exchange reservoirs close to the biosphere the activity is ~$6.8 \cdot 10^{17}$ Bq. Let us assume that burning irradiated graphite will not increase this value by more than 1% (the level of natural fluctuations in $^{14}C$ activity). Then, assuming the activity of C-14 in irradiated graphite is $1.85 \cdot 10^{11}$ Bq/ton, an amount $6.8 \cdot 10^{15} / 1.85 \cdot 10^{11} = 3.7 \cdot 10^4$ tons of such graphite can be burned.

Reactor masonry can be divided into three constituent parts in order of decreasing C-14 activity: core blocks ($1.85 \cdot 10^{11}$ Bq/ton), reflector blocks ($\leq 7.4 \cdot 10^{10}$ Bq/ton), and periodically extracted bushings ($< 3.7 \cdot 10^{10}$ Bq/ton). These estimates show that it is possible to burn large

mass of irradiated graphite (~$5 \cdot 10^4$ tons) without any global ecological consequences accompanying $CO_2$ gas emissions.

The average lifetime of C-14 is $8.3 \cdot 10^3$ yr, so that for any period of time ranging from 10 to 100 or more years during which irradiated graphite is burned the atmospheric emissions are small. The activity of long-lived radionuclides present in the ash impurities of graphite, activation and fission products, and actinides and not C-14 will limit the burn rate.

The main materials in which C-14 is produced are fuel, moderator (graphite, light or heavy water), coolant (water), and nitrogen in the gas loop. We shall present estimates of C-14 production in RBMK-1000 reactors assuming the following nitrogen impurity fractions (%): $7 \cdot 10^{-3}$ in fuel, $5 \cdot 10^{-4}$ in water, $3 \cdot 10^{-3}$ in graphite, 10 in the gas used for purging the reactor space, Bq/yr·$10^{-10}$:

| | |
|---|---|
| $UO_2$ fuel, 192 tons, enrichment 2%: | |
| N-14(*n*, *p)*C-14 reaction | 110 |
| O-17*(n,* α)C-14 reaction | 7.4 |
| ternary fission | 3.7 |
| Graphite moderator, 2000 tons: | |
| C-13*(n,* γ)C-14 reaction | 370 |
| N-14(n, *p)*C-14 reaction | 370 |
| Coolant: | |
| O-17(n, α)C-14 reaction | 37 |
| N-16*(n, p)*C-14 reaction | 18.5 |
| Purging gas (He + $N_2$): | |
| N-14(n, *p)*C-14 reaction | 370 |

The results of these approximate estimates are close to those published in [43], where it was also noted that in the air layer at the ground near nuclear power plants with RBMK-1000 reactors the activity of technogenic C-14 is negligibly low compared with the natural background activity, and it is not dose-forming and requires no observations.

The mass of the ash residue which must be immobilized in a radiation-resistant matrix for burial is ~1 % of the mass of the liquidated irradiated graphite.

### 7.6 Burning of graphite close to thermal power plant [40]

In order to mitigate the impact of the $CO_2$ release containing C-14 on the environment, in the Russia and in France, an idea is in consideration on the possibility to localize a small fluidized bed incinerator for burning low contaminated graphite waste in the vicinity of a large thermal power plant burning oil or coal, which creates a depleted zone of C-14 due to rejection of large quantities of $CO_2$ depleted by C-14. For instance a 10 kg/h fluidized bed burning of low contaminated graphite should have no measurable impact on the C-14 concentration if the off-gas is ejected (mixed) with the off-gas of a 600 MWe plant burning coal. In this case, the average concentration of C-14 in the environment will not alter background levels.

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