DOWN-BLENDING OF HEU GRAPHITE FUEL IN KAZAKHSTAN

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Abstract

The National Nuclear Center of Kazakhstan (NNC) has an Impulse Graphite Reactor (IGR) with homogeneous uranium-graphite core. Graphite blocks of the reactor core are impregnated with water solution of uranyl nitrite with concentration of ~3.1 gr of uranium per one kilogram of graphite. Enrichment of uranium by isotope 235U is 90%. The reactor core consists of immovable and movable parts surrounded by graphite reflectors. The physical start-up of the IGR reactor took place in June 1960, while the energy start-up occurred in August 1961. In 1967 the IGR reactor was upgraded. The uranium mass in a core was increased from 7.46 kg to 9.0 kg and the diameter of the central experimental channel was enlarged. Since reactor modernization, about 2.5 kg of Highly Enriched Uranium (HEU) fuel from the first reactor core which was never used in a reactor has been stored at the reactor site. Last year the government of Kazakhstan made a decision to down-blend fresh HEU graphite fuel from the IGR reactor at the Ulba Metallurgical Plant (UMP) in Ust-Kamenogorsk, Kazakhstan.

The UMP developed a special technology to down-blend IGR graphite fuel which includes fuel crushing, graphite oxidation, dissolution of uranium oxides, 235U based correction using depleted or natural uranium, uranium extraction, precipitation, and calcination to oxides. The final product will have an enrichment that is less than 20%. The UMP facility preparation for graphite fuel down-blending was completed in August 2019 and the down-blending process, which may take up to one year, was started in October 2019. The down-blending will be conducted under the IAEA safeguards.

The irradiated first HEU core from IGR reactor was discharged in 1967 and is currently stored in a dry storage at the reactor site. The total core weight is about 2,600 kg, with the total mass of uranium (including uranium 235U and 238U) about 7.46 kg. The radionuclide composition is as follows: 137Cs, 90Sr, 151Sm, 99Tc, 155Eu, 93Zr, 135 Cs. Since this material was irradiated in a reactor, its down-blending at the UMP is not allowed under the current UMP license.

In 2018-2019 NNC conducted a feasibility study for down-blending and final disposition of the irradiated IGR graphite fuel at the NNC site. Based on preliminary results of the feasibility study, the dry mixing technology using natural uranium has been proposed as a prospective method for down-blending of the irradiated HEU graphite fuel with the following cementation of the down-blended material for permanent disposition.

1. IMPULSE GRAPHITE REACTOR BACKGROUND

The Impulse Graphite Reactor (IGR) was constructed in 1960 at the Semipalatinsk Test Site under the initiative of I.V. Kurchatov by specialists of the former USSR enterprises, mainly RRC “Kurchatov Institute” and NIKIET [1]. IGR refers to research nuclear reactors on thermal neutrons with a homogeneous uranium-graphite core. The IGR Reactor is one of the oldest research reactors in the world and is a unique source of neutron and gamma radiation characterized by a high dynamic of power change. It operates in the mode of programmable power impulses and is self-quenching according to the principle of impulse quenching. Physical start-up of IGR reactor was implemented in June 1960, while the energy start-up occurred in August 1961. In 1967, the reactor was upgraded. 235U mass was increased in the core (from 7.46 up to 9.0 kg) and the diameter of the central experimental channel was enlarged (from 180 mm to 290 mm).

Structurally, the reactor is a stacking of graphite blocks, assembled into columns, which are placed in sealed steel cylinder-shaped casing with a helium medium. The casing is placed in a tank with cooling water. Graphite blocks of the reactor core are impregnated with a water solution of uranyl nitrite (uranyl nitrate) to the concentration of ~3.1 grams of uranium per one kilogram of graphite. Enrichment of uranium by isotope 235U is 90% and the 235U charge in the core is 9 kg. The core consists of immovable and movable parts of stacking (IPS and MPS) surrounded by side and frontal graphite reflector.

The reactor has central and lateral experimental channels, which are equipped with loop water-cooled devices (ampoules). Control and protection operating elements of the reactor are 16 graphite control rods with absorbers made of gadolinium oxide.

The main operating modes of the reactor are unregulated impulse mode (neutron flash self-quenching mode) and regulated mode. To implement self-quenching mode, the reactor is provided some reactivity, which increases delayed neutron fraction that determines a shape, amplitude and semi-width of a neutron flash. Quenching of a flash occurs due to the negative temperature effect of reactivity (-0,03 βeff/degree). Regulated mode is implemented by displacement of the operating elements of the control and protection system (CPS), compensating negative temperature effect of reactivity by a specified law. The shape, amplitude (power level) and duration of regulated mode can vary and are determined by tests on the basis of a condition of maximum energy release in the reactor core, which makes 5.2 GJ and is the operation limit of the reactor corresponding to thermal neutron fluence 3.7×10^{16}.

The reactor includes a set of technological systems, which provide vacuuming and filling of casing with helium work medium, heat rejection from the reactor vessel and ampoules of experimental channels, control and emergency protection of the reactor, technology and radiation monitoring of operation limits, and conditions of the reactor safe operation.

Generally accepted design of reactor tests assumes use of experimental loop benches and installations to provide required thermohydraulic modes in test objects. In this regard, a pneumo-hydraulic bench was established at the IGR reactor. This presents a developed system of structures, equipment, shut-off and safety valves, pipelines, means for control and monitoring with areas of opened cooling circuits (opened loops), where coolant is pumped through test objects and drained from circuit into discharged (for gas) and waste (for liquid) hermetic systems.

2. DOWN-BLENDING FRESH IGR HEU FUEL

2.1. Fuel Feasibility Study for Down-blending Fresh IGR HEU

The purpose of the feasibility study was to determine the principal technical solutions and estimate capital costs for the facility upgrades of the existing HEU down-blending site to ensure possible processing and down-blending of the above material at the Ulba Metallurgical Plant (UMP) JSC. During the feasibility study the following nine works were performed:

(1) Examine possible options of the equipment placement at the existing areas of HEU down-blending site with consideration for the regulatory requirements of the Republic of Kazakhstan;
(2) Determine energy consumption and parameters to make technical solutions on the technical upgrade of the engineering systems and lines to support the site;
(3) Make the necessary evaluations of the water supply system, and the heat and vent systems, as well as make the preliminary evaluation schemes. Inspect the existing engineering systems to determine carrying capacity;
(4) Prepare initial data and request for the technical conditions;
(5) Prepare initial data to develop the feasibility study cost estimate documentation by sections;
(6) Develop the cost estimate documentation for commissioning;

(7) Make construction cost estimation;
(8) Draw up the text document and drawings related to the made solutions with specification of nuclear and radiation safety;
(9) Collect information, make estimation and draw up the text document titled "Environment Impact Preliminary Assessment. Statement of Environment Implications".

The nine areas of work were completed successfully. The review on environmental and on human health concluded that the HEU down-blending site operation would neither disrupt the existing ecological balance nor have unfavourable impact on human health. The results of the feasibility study concluded that IGR Fresh fuel could be successfully down-blended at Ulba.

2.2. UMP JSC Facility Upgrades for Down-blending Fresh IGR HEU Fuel

A HEU down-blending site existed at UMP JSC. The site has all the required operations to down-blend HEU enriched to 90% 235U to LEU by diluting with natural uranium. Nuclear fuel of the IGR reactor is in the form of graphite blocks and rods. The UMP JSC site is not designed for processing nuclear material in the form of blocks and rods of graphite, so it was necessary to perform a facility upgrade. This included upgrading the existing facility and installing new equipment. The upgrade of the site did not require any changes to the basic structure of the existing facility.

To accommodate the new process equipment, an existing metal wall partition was moved, and a heat exchanger fence was installed. Site planning was performed in accordance with sanitary rules. The site was isolated from the other production sites in the facility. The HEU site has rooms for both continuous and short time stays of the staff and personnel. Staff can come either in or out the short-stay room through an existing sanitary exit.

Nuclear material will be down-blended by crushing, milling, oxidizing to U3O8, dry mixing with U3O8 powder enriched to 0.71% 235U, and dissolving the produced powder in nitric acid. This will produce a Uranyl nitrate solution enriched to 19.8-19.92% 235U for further processing with existing equipment that is available at the facility processing site.

The type and the amount of equipment were defined in accordance with the process requirements to assure the set production targets. The equipment was selected and chosen in accordance with the requirements of the industrial and radiation safety guidelines.

All operations with the nuclear material will be performed in HEU down-blending line of glovebox chambers. A separate chamber with glove handling is designed for each operation (see Fig 1).

Fig. 1. New Glovebox Processing Line

For mechanical fragmentizing of the nuclear material to no more than 40 mm, a Test-System pressing machine was installed to the mechanical fragmentizing chamber (see Fig 2).

Fig. 2. Newly Installed Test-System Press

A Retsch BB 50 Jaw crusher will be used for crushing the material into fines. The newly purchased equipment was installed into the upgraded crushing chamber (see Fig 3).

Fig. 3. Newly installed Retsch BB 50 Jaw Crusher

To burn the graphite in the crushed material and oxidize uranium to U3O8, the existing oxidizing chamber was equipped with four high-temp muffle furnaces (see Fig. 4) with natural air circulation and exhaust piping.

Fig. 4. Newly installed Nabertherm Furnace

Other HEU down-bending line chambers and equipment were unchanged. All HEU down-bending line chambers are connected to the existing ventilation system. Hot air is pumped out of the muffle furnaces through heat exchangers. The applicable inspection, measuring, and weighing equipment was registered in the state register.

New ventilation equipment (air cooling heat exchanger, plate-type water-water heat exchanger, filters, reserve tank) were installed. New water supply nets were arranged, designed for, cooling down of the installed equipment.

To distribute electric power to the HEU down-blending equipment, a power switchboard was installed together with automatic switches that protect the equipment and cable line from overload.

The upgrades to the HEU facility were completed on 23 August 2019. The first batch of fresh IGR HEU fresh fuel was delivered from NNC to UMP JSC on 09 September 2019. The down-blending process may take up to one year and was started on 14 October 2019. The down-blending will be conducted under the IAEA safeguards.

3. DOWN-BLENDING IRRADIATED IGR HEU FUEL

3.1. Feasibility Study for Down-blending Irradiated IGR HEU Fuel

A technical feasibility study was performed for irradiated IGR HEU fuel conversion to LEU material. To accomplish this, the following five tasks were performed:

(1) Collection of initial data on IGR fuel;
(2) Review of uranium- graphite fuel processing technologies;
(3) Study of filtration systems;
(4) Review waste disposal methods;
(5) Review of irradiated treatment technologies.

3.1.1. Irradiated IGR HEU Fuel Characteristics

Based on the collected data on IGR fuel, the irradiated IGR HEU fuel characteristics are:

— Uranium-graphite fuel with enrichment of 90% of 235U, total core mass of about 2.6 t in which total mass of uranium (including uranium 235U and 238U) is about 7.46 kg, radionuclide composition as of 2016 is: 137Cs, 90Sr, 151Sm, 99Tc, 155Eu, 93Zr, 135Cs.
— Fuel represents blocks and rods permeated by uranyl nitrate made of Graphite 11-03, having the following averaged characteristics in the initial state:

- Maximum hardness value in all blocks does not exceed 20 HV 10; micro hardness HV 0.1 of IGR fuel elements filling is ~26.4.
- B1 block and C1 rod material density comprises 1.71 g/cm3, open porosity – less than 12 % of volumetric, characteristics of C2 rod porosity in ~1.6 times more, sample density with porous – in 1.11 times less than in samples of B1 block and C1 rod.
- B1 block and C1 rod material has the greatest values of maximum strength and equal $\langle\sigma c.s.\rangle = 57.4 \pm 2.7$ MPa, $\langle\sigma f.s.\rangle = 24.5 \pm 1.3$ MPa and $\langle\sigma c.s.\rangle = 51.7 \pm 1.7$ MPa, $\langle\sigma f.s.\rangle = 20.3 \pm 3.6$ MPa respectively. C2 rod material has relatively low strength characteristics: $\langle\sigma c.s.\rangle = 30.1 \pm 0.4$ MPa and $\langle\sigma f.s.\rangle = 11.8$ MPa.
- Distribution of uranium on volume of the studied samples is relatively equal: average content of 235U comprised – 0.3 %.

— The measurements performed on eight uranium-graphite blocks showed that the average EDR value from the blocks is ~ 1.5 mSv/h closely and 0.3 mSv/h at a distance of 0.1 m. To ensure safe working conditions with uranium graphite fuel, it is necessary to use additional protective barriers.
— In the irradiated elements of the IGR reactor core, uranium is presumably in the form of carbides, which in the finely dispersed state have the ability to self-ignite in air in the absence of heating. They also actively interact with water at indoor temperatures with the formation of gaseous products - acetylene, methane, hydrogen, ethane, etc., that when handling fuel requires an inert environment (argon, nitrogen).

3.1.2. Conclusions on Well-Known Reactor Graphite Treatment Technologies

There are plenty of chemical technologies for HEU fuel conversion to LEU fuel, but all of them involve the possibility to separate 235U, so those should be eliminated. Therefore, only the technology of uranyl dinitrate UO2(NO3)2 dilution by natural uranium was considered.

Analysis of the existing technologies for reactor graphite thermal treatment (combustion) leads to the conclusion that most of them are at the development and testing stage. All the technologies have both advantages and disadvantages; consequences of technology implementation are poorly studied. Direct combustion is the simplest way to treat reactor graphite containing spent nuclear fuel spills.

3.1.3. Filtration Technologies

The current section reviews safety methods to be used during gaseous radioactive waste management such as 85Kr, 129I, T, aerosols, presenting salt and acid fogs containing liquid particles 10–2 – 10 g/m3. There is no industrial method for 14C radioisotopes trapping. This problem is solved only in laboratory studies. The possibility exists to use liquid adsorbents 14C based on hydroxides, nitrogen-carbon and aluminium silicates. Fluorocarbon is also used to catch 99.9% 14C at low temperatures (from –40 to +40C). Therefore, safety during carbon radioisotopes management lies in the preventing of its release into the environment in quantities exceeding allowable emissions, established according to HS SERRS. Methods for air cleaning from radioactive gases and aerosols are:

- filtration on fine-fibered polymers in form of fibres (for aerosols);
- filtration on packed filters (for aerosols);
- adsorption by solutions;
- gases adsorption on solid adsorbents;
- time-lag.

To design a gas purification system, it is necessary to know what gases may be separated during uranium-graphite fuel reprocessing at different temperatures and in what volumes. To do so it would be necessary to carry out thermogravimetric study of irradiated fuel samples to identify the output gaseous isotopes. The selection of ready gas purification systems used at nuclear enterprises (reactors, radiochemical plants) or creation of filter systems on the basis of collected material would be more suitable for laboratory study.

3.1.4. Radioactive Waste Disposal Methods

Based on the study into basic methods for radioactive waste disposal, a conclusion can be made that cementation is the most usable. Vitrification requires special equipment and knowledge of the process peculiarities. High temperature will cause radioactive waste part transfer to gases. Bitumization in comparison with cementation possesses less chemical, thermal and radiation strength. Large gas release also prevents using the application of bitumization. At least 25% of radioactive waste mass is subject to cementation. When calculating activity for pure uranium isotopes mixture (20% 235U, 80% 238U) value is ~3×104 kBq/kg, that is medium-active waste. Availability of other isotope, in particular, caesium and strontium, increase specific activity to high-level radioactive waste. Therefore, in order to meet the requirements of wastes disposal as low-level waste, dilution material shall be used as matrix modifier.

3.1.5. Proposed Irradiated Treatment Technologies

There is a lack of uranium-graphite fuel treatment information in the public press. Materials are available on treatment technologies for reactor graphite which was used as a moderator in reactor core and contained no fuel. The graphite activity is caused by fission products, falling on the graphite (spent nuclear fuel spill). Graphite of the IGR is fuel elements saturated by nuclear material with 235U enrichment of 90% and irradiated in the reactor. The treatment of this material requires: (1) At first stage reduce 235U content to the level ≤20% by diluting with natural 238U; (2) Treatment technology shall exclude stages enabling 235U separation.

Four methods to treat irradiated IGR HEU fuel were selected for further evaluation: Chemical Technologies, Dry Mixing, Combustion, and Combined Combustion and Chemical. Each method has advantages and disadvantages. Dry mixing is the simplest process that requires no complex equipment and eliminates the possibility to separate HEU fuel at any stage. On this account, the dry mixing method was selected as the most preferable. Research and development shall evaluate the reasonability of the dry mixing technology application for HEU fuel depletion.

The technology of HEU fuel dry mixing with natural uranium is proposed as a prospective method for irradiated IGR fuel down-blending due to its simple implementation. It is supposed that the same fine particles of natural uranium and HEU, mixed to necessary concentrations, will be difficult to separate, and further cementation will make the process nearly impossible.

The existing technologies for nuclear graphite processing are mostly based on isolating radioactive graphite from the environment. Graphite reactors' RW contain various radionuclides such as T and 14C, as well as corrosion/activation products (57Co, 60Co, 54Mn, 59Ni, 63Ni, 22Na, etc), fission products (134Cs, 137Cs, 90Sr, 152Eu, 144Ce, etc.) and transformation elements (238Pu, 239Pu, 241Am, 243Am, etc.). According to the researches of TPU [2], 14C content is 95% of all irradiated graphite activity.

The preliminary scheme of the HEU fuel dry mixing with natural uranium is provided in five steps:

(1) Crushing of uranium-graphite fuel blocks and rods;
(2) Grinding of uranium-graphite fuel blocks and rods;
(3) Mixing highly enriched uranium with natural uranium
(4) Cementation
(5) Storage and Disposal

First, graphite blocks are crushed and milled. For grinding uranium-graphite fuel blocks and rods, a grinding jaw crusher can be used on pieces sized from 25 to 5 mm. In order to reduce 235U enrichment, grinding shall be accompanied by HEU fuel mixing with natural uranium until the concentration of 235U is below 20%. Simultaneous grinding and mixing may be performed using ball or rotary mill for fine milling. Cement will be added, and the mixture will be allowed to cure. The advantage of this method is that the technology excludes application of liquid agents and avoids problems related to liquid radioactive waste disposal. The method is attractive due to a relatively small number of processing stages, as well as requiring no expensive agents.

3.2. Regulatory Requirements for Storage and Disposal of Irradiated IGR HEU Fuel

Requirements for storage and disposal of radioactive waste are determined by the following legislative and regulatory acts of the Republic of Kazakhstan:

3.2.1. Law of the Republic of Kazakhstan "On the Use of Nuclear Energy"

Article 17. Management of radioactive wastes and spent nuclear fuel

The Law of the Republic of Kazakhstan "On the Use of Nuclear Energy" stipulates that radioactive waste generated in the territory of the Republic of Kazakhstan must be disposed so as to provide radiation protection for the population and the environment for the entire period of time during which they can be a potential hazard.

3.2.2. Environmental Code of the Republic of Kazakhstan

Article 272. The environmental requirements for storage and disposal of radioactive materials and waste

The Environmental Code of the Republic of Kazakhstan defines the requirements for storage and disposal of radioactive materials and waste. When storing and disposing radioactive materials and waste, nature users must ensure following:

- The impossibility of spontaneous nuclear chain reactions, and protection from excessive heat;
- The effective protection of the population and the environment through the use of appropriate methods of protection in accordance with the rules and regulations of radiation safety;
- The risk assessment of biological, chemical or other risks that may be associated with the storage and disposal of radioactive materials;
- The preservation of records relevant to the location, design and contents of the disposal;
- Control and limit of unauthorized access to radioactive materia1s, as well as the prevention of unplanned release of radioactive substances into the environment.

3.2.3. Rules for selection of the location of nuclear facilities and disposal sites (NFDS)

Article 2. The procedure for selection a location for NFDS

According to the rules, the selection of the location of a nuclear facility and of a disposal site must include a preliminary study of the designated area, as well as the identification of at least three other competitive sites.

3.2.4. Technical Regulation "Nuclear and Radiation Safety"

Article 7. Requirements for the design of storage facilities for nuclear materials, nuclear fuel, radioactive materials and waste

The technical regulation "Nuclear and Radiation Safety" defines the requirements for the design of storage facilities for nuclear materials, nuclear fuel, radioactive materials and waste.

According to the Regulation, and due to the possibility of achieving criticality in the storage facilities for nuclear and radioactive materials, fresh and spent fuel during their placement and movement is physically excluded by ensuring the corresponding characteristics of the storage facilities. In the spent fuel and radioactive waste storage facilities, the project must provide for heat sink systems and corresponding chemical composition of the heat sink medium to prevent interaction, as a result of which radioactive materials could enter the facility premises or the environment.

3.2.5. Rules for organizing the collection, storage and disposal of radioactive waste and spent nuclear fuel

Article 1. General requirements for the collection, storage and disposal of radioactive waste

The Rules define the general requirements for storage and disposal of radioactive waste. Storage and disposal of radioactive waste is documented. During storage and disposal, the radioactive waste register is maintained and a certificate for a radioactive waste batch transferred for conditioning, storage, and disposal is filled.

Radioactive waste storage and disposal is subject to the sanitary rules "Sanitary and Epidemiological Requirements for Radiation Safety", approved by the Order of the Acting Minister of National Economy of the Republic of Kazakhstan dated March 27, 2015 No. 261.

3.2.6. Sanitary Rules "Sanitary and Epidemiological Requirements for Radiation Hazardous Facilities"

Article 43. Requirements for reprocessing and disposal of radioactive waste

According to the Sanitary Rules "Sanitary and Epidemiological Requirements for Radiation Hazardous Facilities", radioactive waste is disposed of in the "dirty" zone of the territory of the radioactive waste disposal site in special containers. Disposal, as well as temporary storage of radioactive waste outside these containers, is not allowed.

3.2.7. Sanitary Rules "Sanitary and Epidemiological Requirements for Radiation Safety"

Article 10. Radioactive Waste Management Requirements

According to the Rules, selection of radioactive waste disposal sites should be made taking into account hydrogeological, geomorphological, tectonic and seismic conditions. At the same time, radiation safety of the population and the environment should be ensured over the entire period of waste isolation, considering long-term forecast. The effective dose of the population due to radioactive waste at all stages of their management should not exceed 10 µSv/year.

3.2.8. Regulatory Requirements Summary

According to the legislation of the Republic of Kazakhstan, both storage and disposal of radioactive waste for the purpose to ensure radiation protection of the population and the environment for the entire period of time during which they can be a potential hazard are allowed. In Kazakhstan, there is also a procedure for the construction of disposal facilities, including the stages of approval and decision by the Government of the Republic of Kazakhstan on the construction of a disposal facility, site selection, design, surveys, examinations, and public hearings. Storage facilities provide a safe place for radioactive waste. These facilities will be designed and constructed to prevent the release of radionuclides into the environment in quantities exceeding the limits established by hygiene standards and to ensure the storage life of at least the life of the collection processing, conditioning and storage system.

4. CONCLUSION

The National Nuclear Center of Kazakhstan (NNC) has an Impulse Graphite Reactor (IGR) with homogeneous uranium-graphite core. Graphite blocks of the reactor core are impregnated with water solution of uranyl nitrite with concentration of ~3.1 gr of uranium per one kilogram of graphite. Enrichment of uranium by isotope 235U is 90%.

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The irradiated first HEU core from the IGR reactor was discharged in 1967 and is currently stored in a dry storage at the reactor site. The total core weight is about 2,600 kg, total mass of uranium (including uranium 235U and 238U) is about 7.46 kg, radionuclide composition is as following: 137Cs, 90Sr, 151Sm, 99Tc, 155Eu, 93Zr, 135 Cs. Since this material was irradiated in a reactor, its down-blending at the UMP is not allowed under the current UMP license.

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In November 2019, the fresh IGR HEU fuel was shipped from NNC to Ulba. The down-blending of the last fresh HEU in Kazakhstan should be completed in 2020.

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