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Radiation Safety Issues of Using Regenerated Uranium in Nuclear Fuel Manufacturing at the Electrostal Plant

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Concerns about potential shortage of natural uranium and its increased cost, as well as the accumulation of stocks of regenerated uranium, created interest in using regenerated uranium in the process of manufacturing fuel assemblies for power reactors. However, using regenerated uranium in the fuel manufacturing process presents significant challenges from the radiation safety point of view, as this uranium contains radioactive isotopes associated with uranium-232. To explore the possibility of using regenerated uranium in the fuel manufacturing process, the Electrostal Machine-Building Plant explored the possibility of using uranium with uranium-232 concentrations of up to 5 ppb. The results of the pilot project suggest that large-scale production of reactor fuel from uranium with high concentrations of uranium-232 would require a substantial change of the technological processes and procedures for storage and handling and storage of the fuel produced with this uranium.

Concerns about potential shortage of natural uranium and its increased cost, as well as the accumulation of stocks of regenerated uranium created interest in using regenerated uranium in the process of manufacturing fuel assemblies for power reactors. In Russia, the share of natural uranium (grade “N”) in fuel manufacturing has been historically relatively small. Most of the uranium first passed through plutonium production reactors. This uranium, recovered during reprocessing and enriched, is known as uranium “RS” grade. It includes uranium isotopes uranium-236 and uranium-232 that are absent in natural

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uranium. Both isotopes affect the fuel manufacturing process. Uranium-236 affects reactivity of the fuel, requiring higher enrichments in uranium-235. The presence of uranium-232 significantly increases the radiation background of the material, which creates problems during the manufacturing of fresh fuel assemblies and the subsequent handling of these fresh fuel assemblies at nuclear power plants. Main contributors to the radiation background are decay products of uranium-232 (thorium-228, radon-220, lead-212, thallium-208, etc.) which include sources of hard γ-rays with energies of up to 2.6 MeV.

The handling of uranium hexafluoride (UHF) of the "RS" grade as well as fuel elements produced with it required taking measures that would limit the uranium-232 content in a feedstock of fuel manufacturing plants. The content of uranium-232 was limited at the level of about 1×10^{-7} percent (1 ppb) for those fuel manufacturing plants that performed manual operations of grinding and sorting of uranium dioxide pellets, of a control of its forms, of fuel elements fitting-out, and also operations on dioxide production from UHF. These operations account for a substantial part of the accumulated doses.[1]

The opening of the RT-1 reprocessing plant, which was reprocessing spent fuel of VVR-440 reactors, created regenerated uranium stocks that were obtained from fuel exposed to higher burn-ups with harder neutron spectra than that of production reactors. Uranium separated from irradiated fuel of power reactors was designated as "RT" grade. It should be noted that the RT-1 plant processes not only spent fuel of VVER-440 reactors but also fuel of research, transport and naval reactors, BN-600 reactor, and highly enriched fuel elements from production reactors. As a result, the "RT" grade uranium includes a fairly broad range of uranium materials.

There are two plants in Russia that produce fresh fuel for nuclear power reactors: the Electrostal' Machine-Building Plant (MSZ) and the Novosibirsk Chemical Concentrates Plant (NZKhK). Starting in 1996, the Electrostal plant started using some regenerated "RT" grade uranium in its manufacturing process.

In order to increase the amount of regenerated uranium used for fuel manufacturing, in 2002, JSC TVEL approved "The program of radiation and hygienic substantiation of the transition of JSC TVEL enterprises to regenerated uranium with uranium-232 content up to 2.2×10^{-7} mass percent." The program also called for an assessment of a possibility of using regenerated uranium with uranium-232 content up to 5×10^{-7} mass percent (5 ppb).

At the first stage of this work, the State Scientific Center, Institute of Biophysics (SSC-IBPh) developed a prognosis of the radiation environment at main technological areas of fuel manufacturing that assumed up to 2 ppb content of uranium-232. Results of this work allowed the experimental use of regenerated uranium with uranium-232 content of up to 2.2 ppb.[2]

In 2002 the Electrostal plant was issued a license to use regenerated uranium with uranium-232 content up to 2 ppb in the assembly manufacturing process.[3] As the work began, the effective annual dose for personnel directly involved in the production (category A personnel) did not show a significant increase. In 2004, the average annual dose was equal to 3 mSv, and largest part of category A personnel (about 85 percent) accumulated an effective annual dose of less than 5 mSv. For 15% of category A personnel effective dose was detected in the range of 5–20 mSv (the upper dose limit for category A personnel is 20 mSv per year).

Based on this experience, the Electrostal plant and its parent company TVEL undertook studies of the possibility of increasing the uranium-232 concentration in raw materials up to 5 ppb.

The transition to higher concentration of uranium-232 required the regulators to consider a number of additional factors before issuing a license. Key among those was an increase in the level of γ-radiation in production areas and higher air aerosol contamination levels. Other factors, that were negligible at lower uranium-232 concentrations, had to be taken into account as well—irradiation of skin (hands and crystalline lenses in particular) and radiation doses related to activity of radon-220. As a condition of the licensing process, the Electrostal plant had to install instrumentation and develop procedures for accurate measurements of effective radiation doses for personnel and general population.[4] The regulatory agency, Rostekhnadzor, required the plant to conduct measurements of equivalent doses for crystalline lens, hands, feet, and skin of the personnel. Also, it required monitoring of α-activity for each radioactive nuclide in liquid radioactive waste, radionuclide composition in a soil, building materials, in air at industrial area, buffer zone, and the residential area surrounding the plant.[5]

In 2004, the Electrostal plant conducted an evaluation of the radiation environment in main technological areas involved in processing of uranium of "RS" and "RT" grades.

As part of this evaluation, the plant and the regulators compared external irradiation doses during production of uranium dioxide powder by two technologies, Saturn and dry conversion. The Saturn technology, which is based on gas-flame reduction, had been the primary conversion process used at the Electrostal plant before it introduced a new technology, dry conversion, in 2002. This technology is patented by Siemens and used at the plant in Lingen, Germany. The dry conversion process is highly automated, allowing the use of regenerated uranium with higher concentration of uranium-232 without increasing radioactive burden on personnel.

Radiation measurements, conducted in various points of the body: head, chest, lower belly, feet, and hands showed that with the exception of hands, the body is irradiated uniformly. Average dose rate levels at Saturn facilities were equal to 2.4 ± 0.4 μSv/hour, while at the workplaces of the dry conversion process, average dose rates were equal to 0.5 ± 0.08 μSv/hour.

Relatively uniform irradiation of personnel was also registered at workplaces where fuel tablets were handled and materials were returned for reprocessing. Averaged by all operations, γ-radiation dose rate was equal to 3.3 ± 2.6 μSv/hour. Dose rates in the vicinity of UHF containers were two to three times higher. Hands had dose rates about 1.5 times higher.

At the fuel pellet production line, dose rate values were measured in characteristic points on the head, chest, lower belly, feet, and hands. The highest levels of γ-radiation dose rates were registered at the raw materials, intermediate products, and final product accumulation areas: at containers with uranium dioxide powder and fuel pellets and in the interim storage warehouse for final product. Particularly high levels were detected in the storage for rejected products in the vicinity of a container with rejected pellets. γ-radiation, dose rates could be as high as 200 μSv/hour. Among technological operations, the highest radiation burden was observed at the point of assembling the fuel assemblies. Irradiation of personnel at these workplaces was not uniform, with head, chest and hands areas receiving higher radiation doses.

It should be noted that irradiation during technological operations is not the only source of radioactive dose burden on personnel. Another major source is the volumetric activity of radionuclides in the air. Earlier studies, conducted in 1996 with uranium of RT grade, showed presence of thorium-228 as well as plutonium in the air. In the 2004 study with RS grade uranium, all samples taken at the workplaces involved in conversion and pellet production showed consistent presence of thorium-228. Concentrations of plutonium were below the detection threshold.

Measurements of radiation doses performed in the course of the work with uranium with 2 ppb uranium-232 content were used to estimate radiation doses for uranium with 5 ppb uranium-232 content. The estimate assumed that the uranium is enriched to 2.6% uranium-235, which corresponds to the enrichment used in fuel of RBMK reactors. Depending on the grade of uranium chosen as a base case (from N and RS grades to RT grade), a transition to the uranium with 5 ppb uranium-232 content would lead to:

- An increase in γ-radiation dose rate by a factor of 1.2–2.1;
- An increase in activity of radioactive aerosols by a factor of 1.9–3.8;
- An increase of activity emissions outside of production areas by a factor of 1.9–3.8.

Based on these estimates, the regulators determined that in order to proceed with the licensing process it must obtain experimental data about radiation loads by processing a batch of regenerated 5 ppb uranium of no less than 150 tons. Also, it was concluded that processing of 5 ppb uranium would require a number of measures:

1. The time between production of UHF and receipt of the fuel at the reactor should not exceed 1.5 years to limit accumulation of uranium-232 decay products.
2. The fuel production process has to include thorium-228 monitoring of UHF, uranium dioxide powder and fuel pellets, as well as the air in and outside of work areas.

The decision to begin reprocessing an experimental batch of 150 tonnes of uranium with 5 ppb content of uranium-232 was made in December 2004.[6] The UHF would be supplied by the Siberian Chemical Combine (SCC) in Seversk. A decision was made to use both “Saturn” and “dry conversion” technologies at the stage of uranium conversion. The production process would include manufacturing of uranium dioxide pellets, fuel elements, and fuel assemblies for RBMK-1000 reactors as well as LWR fuel assemblies for Framatome.[7]

In 2005, the Ministry of Health formally approved the pilot project to reprocess a 150-tonne batch of “Fm” grade UHF with uranium-232 content of up to 5 ppb to produce fuel assemblies for RBMK-1000 reactors.[8] The “Fm” grade UHF was produced at the SCC by processing regenerated uranium supplied by Framatome. This uranium was recovered from irradiated fuel of light water reactors with a burn-up of up to 40 MW days/kg. The average concentration of individual radionuclides and their total activities in “Fm” grade UHF received in 2005 are presented in Table 1. The material, supplied as triuranium octoxide (U_3O_8), was purified, converted to hexafluoride, and slightly enriched to bring enrichment to 3% uranium-235. The enrichment was higher than the 2.8% normally used in RBMK fuel instead to compensate for neutron absorption by uranium-236.

Table 1: Average concentrations of radionuclides and their activities in "Fm" grade UHF received by MSZ in 2005. Total activity of γ-radiating nuclides is equal to 1.78×105 becquerel/g uranium.

Radionuclide	Average content of radionuclide in mass percent	Percent part of specific activity	Specific activity in becquerel/g uranium
Uranium-232	$(4.82 \pm 0.05) \times 10^{-7}$	2.2	3987
Uranium-234	0.061 ± 0.001	79.1	140780
Uranium-235	2.96 ± 0.01	1.4	2136
Uranium-236	0.79 ± 0.02	10.6	18868
Uranium-238	96.19	6.7	11944
Thorium-228	—	0.08–0.11	140
Neptunium-237	—	1.7×10^{-5}	0.03
Plutonium-238	—	0.8×10^{-5}	0.015
Plutonium-239	—	0.8×10^{-5}	0.015

The first question addressed during the pilot project with “Fm” uranium was the dose rate from UHF containers. Analysis of the data on γ- and neutron radiation from the containers concluded that:

- γ-radiation dose rates from full containers with “Fm” grade UHF is directly proportional to the time elapsed from the time the UHF was produced.
- γ-radiation dose rates from emptied containers is higher than that from containers with UHF, as radiation from nonvolatile isotopes left in the container is no longer shielded by uranium.
- Neutron radiation dose rates from full containers with “Fm” grade UHF is equal to 15.5 μSv/hour. This is on average three times higher than the dose rate from emptied containers.
- Neutron radiation during work with containers with enriched uranium hexafluoride generates through (α,n)-reaction on fluorine nuclei and it may increase with large accumulation of containers in storage and their treatment.

To decrease the dose load on personnel during work with the 5 ppb uranium-232 the Electrostal plant undertook extensive modernization of its production lines. The measures that have been implemented include:

- Minimizing the storage time of UHF containers;
- Decreasing the fuel processing time;
- Decreasing the hold time of material in interim storage in plant shops;
- Excluding some quality control operations and optimizing others;
- Modifying the equipment to reduce the concentration of radioactive isotopes in the air;
- Modification of work areas in order to avoid hand contact with fuel pellets;
- Allocation of separate lines and areas for work with uranium with 5 ppb uranium-232 content;
- Personnel and work areas were equipped with new radiation monitors that allowed continuous measurements of radiation dose rates.

Among the significant findings of the pilot project was the relatively low concentration of thorium-228 in the uranium that entered the fuel production line. The observed value was an order of magnitude lower than the estimate made during preliminary studies, which suggested that the thorium-228 contents would correspond to 140 becquerel/g uranium. The difference is most likely explained by the absence of thorium carry over from cylinders during

UHF evaporation in a modern technological process. Also, the measurements did not detect accumulation of uranium-232, thorium-228 and their daughter decay products in the equipment at the fuel production line. Actual values of effective doses for the category A personnel was equal to 1.7, 1.68 and 1.66 mSv/year in 2008, 2009 and 2010 respectively.

Substantial modifications of the processes that were introduced at the MSZ plant and the results of the pilot project allowed the regulator, Rostekhnadzor, to amend the MSZ operating license to permit processing of uranium with uranium-232 concentrations of up to 5 ppb.[9]

At the same time, the pilot project demonstrated that an increase of the throughput of reprocessed uranium with high uranium-232 concentrations would require a significant modification of the technological process, as the relatively simple and straightforward measures implemented for the pilot project such as protective screens, hermetic sealing of the equipment, and reducing the of personnel residence time in work areas have largely reached their limit.

Handling of the unirradiated fuel produced with regenerated uranium also presents a potential problem. The experimental batch of fuel assemblies produced during the pilot project was delivered to the Leningrad nuclear power plant that operates RBMK reactors. The uranium-erbium fuel with three percent enrichment was approved for use in the reactor and for storage at the reactor site. However, the use of 5 ppb uranium will significantly increase the radiation load on the personnel who handle fuel at the reactor site. The dose associated with the regenerated uranium fuel could be responsible for about half of the maximum allowed dose. This share would increase if the fuel is stored at the reactor site. For example, for fuel that was in storage for four years, the personnel of the storage site would reach the limit of allowed dose during fuel acceptance and delivery operations that normally take only a very small fraction of working time.

Overall, while the Electrostal MSZ plant demonstrated the possibility of a limited use of regenerated uranium with uranium-232 concentrations of up to 5 ppb, its experience also suggests that the measures that were undertaken during the pilot project appear to have reached the limit of what can be accomplished without significant modification of the plant. A transfer to large-scale production of reactor fuel from uranium with high concentrations of uranium-232 would require a substantial change in the process and procedures for storage and handling of the fuel produced with this uranium.

NOTES AND REFERENCES

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2. This work was carried out according to provisions of the Gosatomnadzor license, sanitary-and-epidemiologic opinion letters of the Center for Sanitary and

Epidemiological Supervision No. 21 of the Russian Federal Medical and Biological Agency (TsGSEN) with the support of SSC-IBPh.

3. Gosatomnadzor licenses No. GN-03-115-0885, GN-03-115-0799.

4. Letter from Deputy of Roctekhnadsor Chairman No. 7–38/451 from 8 July 2004 to Vice-president of JSC "TVEL".

5. "Standards of Radiation Safety," NRB-99 SP 2.6.1.758–99 (State chief sanitary inspector of Russian Federation, 1999); "Collection, Processing, Storage and Conditioning of Liquid Radioactive Wastes, Safety Requirements," NP-019–2000 (Gosatomnadzor, 2000); "Collection, Processing, Storage and Conditioning of Solid Radioactive Wastes, Safety Requirements," NP-020–2000 (Gosatomnadzor, 2000).

6. Technical meeting with representatives of JSC "TVEL", FGUP "NIKIET", FGUP "VNIINM", JSC "MSZ", RNC "KI", FGUP "KPI", SSC-IBPh, FGUP "Rosenergoatom", "Rostekhnadzor", FU "Medbioextrem" took place on 14 December 2004.

7. International Atomic Energy Agency, "Management of Reprocessed Uranium Current Status and Future Prospects", IAEA-TECDOC-*1529*, (February 2007), 50.

8. Authorization No. 32–028 dated 21 December 2005.

9. Amendment of JSC "TVEL" license to the work of JSC "MSZ" provisions with including of No.3 modification was executed 19 October 2009.