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The Russian ARGUS Solution Reactor HEU-LEU Conversion: LEU Fuel Preparation, Loading and First Criticality

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ABSTRACT

ARGUS is a 20 kW solution reactor, which has operated at NRC "Kurchatov Institute" since 1981. Fuel was highly enriched (90%) uranyl (HEU) sulfate solution. The reactor has an inherent safety.

Feasibility of the reactor conversion to low enriched uranium (LEU) fuel was studied in 2010-2012. Positive results of neutron-physical and thermal-hydraulic calculations allowed taking a decision about the reactor conversion.

Preparation work was conducted at the reactor ARGUS in the period 2012-2014 and LEU-fuel was prepared and loaded, reaching first criticality in July 2014. LEU-fuel was produced by mixing current HEU-fuel and fuel with 1.8% enrichment. Mixing was conducted in the reactor ARGUS vessel. As a result the following fuel characteristics were obtained: enrichment 19.8%, uranium concentration 380 g/l, volume 25.7 l.

Work was conducted in cooperation with ANL of the USA and supported by USDOE under the Global Threat Reduction Initiative.

The next stage of work is to get a new license of Rostechnadzor and conduct experimental study of the LEU-fuel reactor ARGUS characteristics.

1. Introduction

The reactor ARGUS is intended for providing nuclear-physical methods of analysis and control and production of radionuclides. Scientific research in the field of neutron activation analysis and neutron radiography has been conducted at the reactor practically since its putting into operation. NRC "KI" is developing the methodical basis of radionuclide production in the solution reactor ARGUS. Such a type of the reactor gives a unique

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possibility of using the whole core for radionuclide production and selectively extracting them from the fuel solution [1, 2].

Feasibility of the reactor conversion to low enriched uranium (LEU) fuel was studied in 2010-2012 [3]. Positive results of neutron-physical and thermal-hydraulic calculations allowed taking a decision about the reactor conversion.

After conversion, the reactor ARGUS purpose will remain the same: to search for the best physical-technical solutions when developing nuclear-physical methods of analysis and control as well as to develop work on production of radionuclides. After conversion the reactor ARGUS can become a prototype of a facility for commercial production of Mo-99 at the solution reactor.

2. ARGUS Reactor

ARGUS is a 20kW homogeneous solution thermal-neutron reactor. The reactor core was the uranyl sulfate water solution enriched up to 90 % in ^{235}U located in a welded cylindrical vessel with a hemispherical bottom and a flat cover. Vertical "dry" channels are installed in the vessel: the central and two symmetric peripheral ones. Control and regulation rods are located in the peripheral channels. The core elements contacting with a fuel solution are made from stainless steel. The reactor vessel is surrounded with a side and bottom graphite reflectors (Fig. 1). There are five vertical channels and one horizontal channel for neutron beam extraction in the reflector.

Catalytic Regeneration System (CRS) is meant for regeneration of radiolytic gases hydrogen and oxygen produced in the fuel solution during the reactor operation at power and providing oxygen volume concentration in the free space of the vessel. It forms a closed loop with the core vessel and has the following units: a recombiner; a heat exchanger; a condensate collector (water accumulator); pipelines. The regeneration system is based on the principle of the natural circulation of a gas mixture along the circuit.

Reactor Cooling System is a system with two loops. The first loop is a coil in the reactor vessel. It is connected to the heat exchanger and the compensation tank. The water is pumped through the coil with help of a pump. The second cooling loop removes heat from water of the first loop in the heat exchanger by water.

The solution reactor has an inherent safety determined by the negative power reactivity effect, by the opportunity of cooling down due to the natural air convection, by the lack of local overheatings, low temperature of the solution (up to 90°C) and pressure in the reactor vessel (below the atmospheric one) [4].

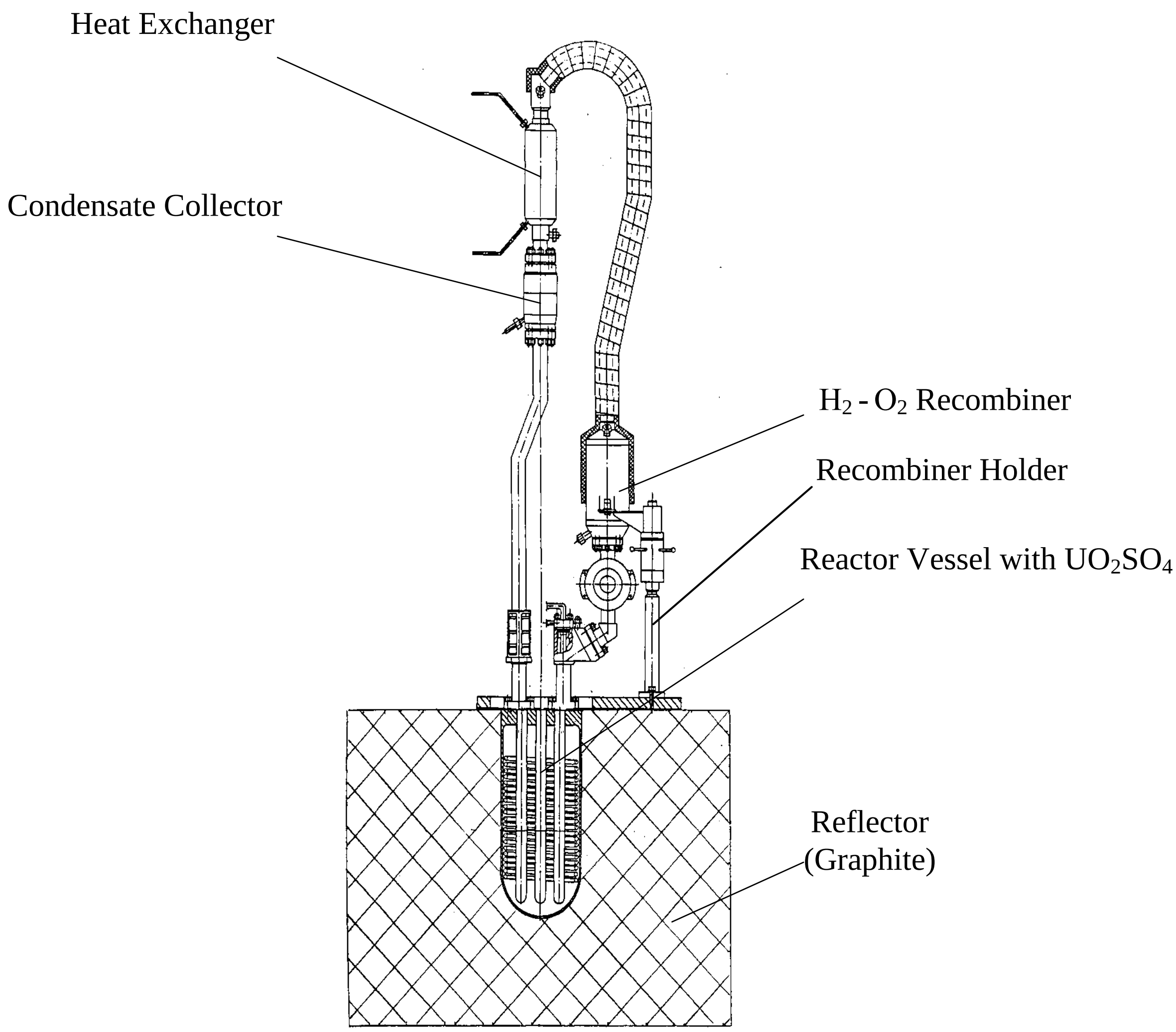

Figure 1 – Scheme of the Reactor ARGUS

3. Conversion Procedure

The reactor ARGUS conversion must result in the following parameters of LEU-fuel:

- composition is aqueous solution of uranyl sulphate,	UO_2SO_4
- enrichment, %	19.8
- uranium concentration, g/l	380
- uranium-235 operating charge, kg	1.880
- operating volume, l	~ 25
- pH	1.0

All standard HEU-fuel being inside the reactor vessel was used completely; also the reserve HEU-fuel (~450 g by the isotope uranium-235) stored in the storehouse was used during production of LEU-fuel with the required parameters. Uranyl sulphate solution with 1.8% enrichment in uranium-235 prepared from uranium dioxide was added to HEU-fuel to reduce enrichment. It was necessary to concentrate HEU-fuel preliminarily, to evaporate a part of water to ensure the final volume of LEU-fuel.

As the gamma-irradiation dose rate from HEU-fuel was high, the decision was taken not to unload it from the reactor vessel, but to prepare the operating LEU-fuel directly in the reactor ARGUS vessel by adding uranyl sulphate solutions with necessary parameters.

The process of the operating LEU-fuel preparation includes the following stages:

- water evaporation (9.2 l) from HEU-fuel in the reactor vessel;
- loading the fuel component with enrichment 1.8% into the reactor vessel (8.6 l);
- loading the fuel component with enrichment 19.8% into the reactor vessel (5.5 l).

The following main work was conducted to enable conversion of the reactor ARGUS to LEU-fuel:

- LEU-fuel reactor ARGUS safety analysis was performed;

- necessary changes are made in the reactor design: a chiller was introduced in the cooling system; a heater was added to make it possible to evaporate fuel. A fuel loading system was mounted;

- physical start-up equipment was installed;

- fuel temporary storage device including alarm signaling system about occurrence of self-sustaining chain reaction was developed, mounted and passed examination;

- technology was developed, licenses were obtained, workplaces were prepared to prepare uranyl sulphate solutions;

- personnel were trained and prepared.

4. Fuel Preparation

Material UO_2 with enrichment 1.8% was chosen as raw material.

Uranyl sulphate was prepared from UO_2 in the following sequence [5-8]:

1. The raw material was dissolved in the concentrated nitric acid

$$UO_2 + 4HNO_3 \longrightarrow UO_2(NO_3)_2 + 2NO_2 + 2H_2O$$

The uranium dioxide was dissolved in the draft hood behind the shield of lead glass. Dissolution process was divided into 500 g portions; it was conducted in nitric acid in a vitreous glass with a backflow condenser.

2. Evaporation of the obtained solution to get anhydrous $UO_2(NO_3)_2$.

3. Anhydrous $UO_2(NO_3)_2$ incineration in the muffle furnace; UO_3 production.

The obtained brown precipitate was incinerated in the muffle furnace at temperature 250-400°C for 4-5 hours until it had constant weight. As a result of the operation, an orange-yellow powder was obtained: uranium trioxide (UO_3).

4. UO_3 dissolution in sulphuric acid.

$$UO_3 + H_2SO_4 \longrightarrow UO_2SO_4 + H_2O$$

400 g portions of uranium trioxide were dissolved in sulphuric acid solution at temperature 80-90°C and uranyl sulphate solution was obtained; raw material and prepared solution are shown in Figure 2.

Uranyl sulphate solutions with the following parameters was prepared:

Solution	Volume, l	U concentration, g/l	Enrichment, %
″Component 1.8%″	8.6	721.5	1.8
″Component 19.8%″	5.5	380	19.8

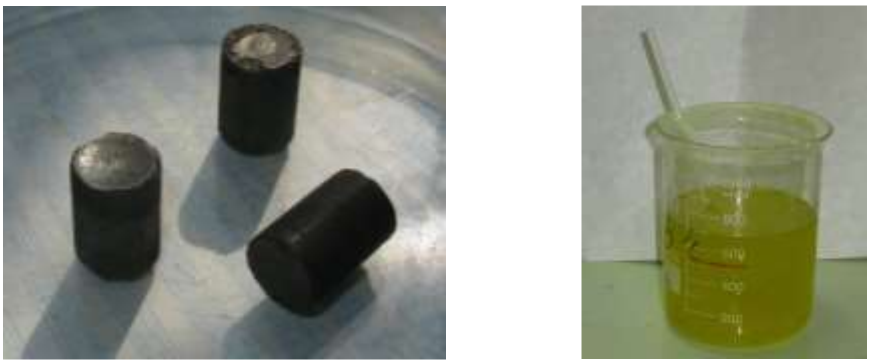

Figure 2 – Raw material – UO_2 with enrichment 1.8%
and prepared solution of uranyl sulphate

5. HEU-fuel Evaporation and LEU-fuel Loading

In April-May 2014 water was evaporated directly in the reactor vessel by the method of vapor content increase in its free volume and vapor-air mixture natural circulation creation in the catalytic regeneration system (CRS) loop. A thermal electrical heater with power 600 W was mounted on the outer surface of the reactor cooling system pipeline in order to increase the solution temperature (Fig. 3). The solution average temperature makes 50°C at the operating flow rate of the heat carrier 0.2 m^3/hour.

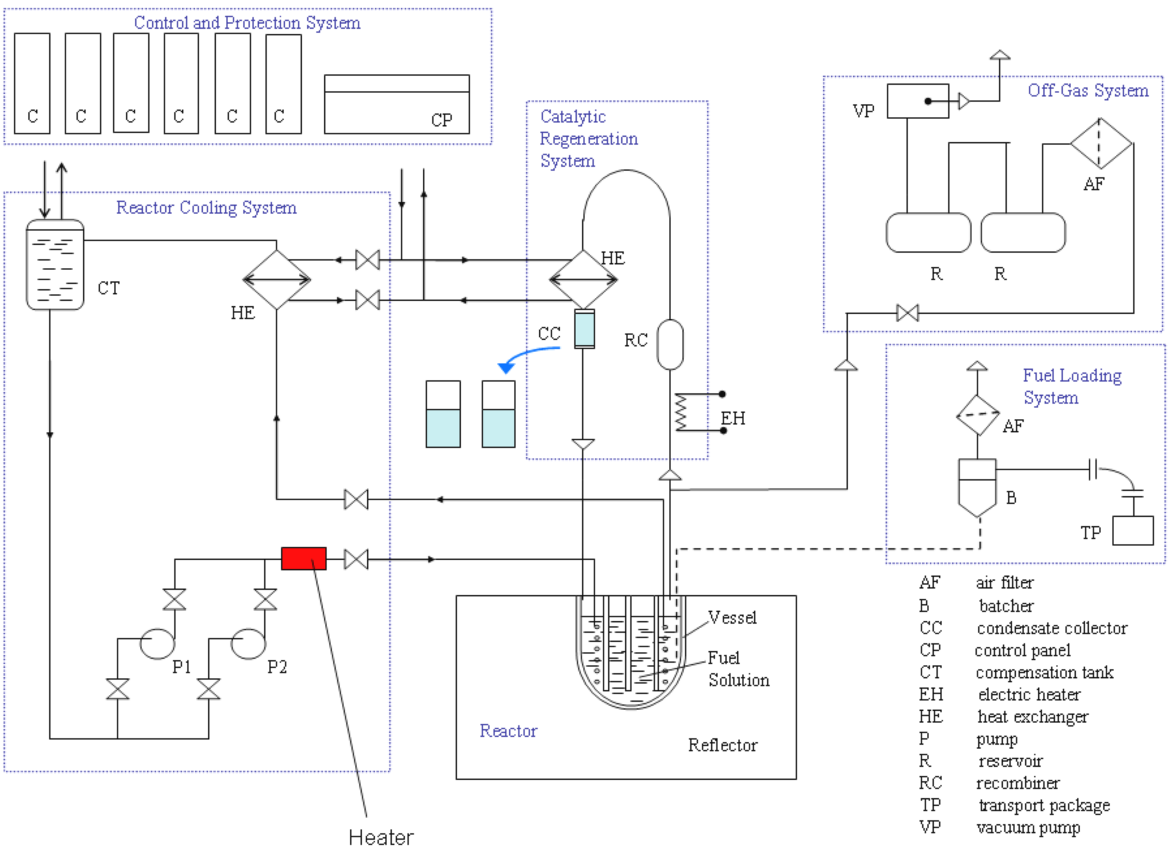

Figure 3 – Reactor Structural Chart

Water vapors were condensed in the CRS condensate collector with volume ~ 1.2 liters. The average velocity of the condensate collector filling made 0.5 liters of water per shift at the initial pressure in the free volume of the vessel minus 0.5 kg_f/cm^2. Water was discharged from the condensate collector with help of an installed valve with manual control. Discharged water amount was determined by measuring its weight in the reservoir. Radiation situation is under continuous control during the discharge process. Water portions as liquid radioactive waste were collected from the discharge reservoirs to the cans with capacity up to 5 liters

The total quantity of evaporated and discharged water being part of HEU fuel of the reactor was 8.5 liters.

In July 2014 HEU-fuel of the reactor ARGUS was transformed into LEU-fuel. Fuel solutions with enrichment 1.8% and 19.8% were poured by portions into the reactor vessel when the emergency protection (EP) working parts were withdrawn and compensating working parts (CWP) were introduced into the core.

The reactor subcriticality was estimated with help of physical start-up equipment during the solution pouring operations.
It was possible to supply air from atmosphere through the fuel solution core to the preliminarily evacuated free volume of the reactor vessel in order to improve the solutions mixing process.
Subcriticality was controlled before pouring each portion of solution and mixing with air.
The reactor ARGUS fuel parameters change at different stages of conversion is shown in Fig. 4.

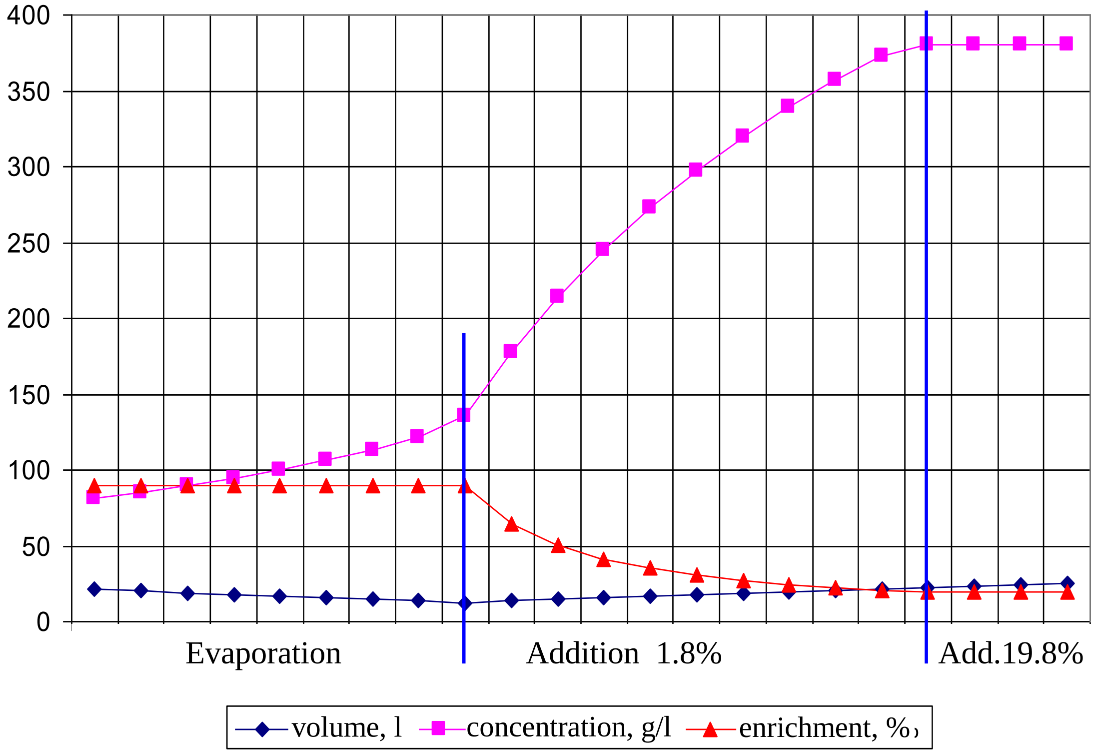

Figure 4 – Change of fuel parameters in the reactor vessel

6. Conclusion

The following main parameters of the reactor ARGUS LEU-fuel were obtained as a result of conversion: enrichment 19.8%, uranium concentration 380 g/l, volume 25.7 l, pH ~ 1. Reactivity margin made ~ 4 β_{ef}. The core subcriticality by EP rods withdrawn from the core and CWP and MR (AR) introduced into the core was not less than 0.98 ($K_{ef} < 0.98$) what meets research reactors nuclear safety requirements.

The next stage of work is to get a new license of Rostechnadzor and conduct experimental study of the LEU-fuel reactor ARGUS characteristics. Application for this license was submitted on April/2014.

Acknowledgement

Work was conducted in cooperation with ANL of the USA and supported by USDOE under the Global Threat Reduction Initiative.

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