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MOLTEN-SALT REACTOR FOR NUCLEAR FUEL CYCLE CLOSURE ON ALL ACTINIDES

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It is proposed that a specialized molten-salt reactor facility for burning long-lived actinides be located in direct proximity to a pilot-demonstration center. A full-scale ZhSR-S with thermal power 2400 MW will make it possible to burn all transuranium elements released during the reprocessing of spent VVER-1000 nuclear fuel and part of the regenerated plutonium and in the process produce 1 GW of electricity and radioisotope products. The inclusion of a specialized reactor facility for burning transuranium elements from the spent nuclear fuel of VVER-1000 reactors into the nuclear energy system as an integral element will make it possible to solve the problem of closing the nuclear fuel cycle for actinides and reduce the volume and lower the cost of the long-term storage and subsequent disposal of high-level wastes produced in reprocessing.

Starting in 2020, the spent nuclear fuel of VVER-1000 reactors will be reprocessed in facilities of a pilot-demonstration center built on the Mining and Chemical Combine site that use innovative technology for regeneration of nuclear materials. After all technological stages are developed, this center will become the foundation base for creating the RT-2 plant which will support an environmentally and economically acceptable system for handling the spent nuclear fuel of VVER-1000 and -1200 reactors.

In accordance with the technological scheme, the formed flow of high-level wastes that contains long-lived actinides goes to conditioning in order to organize storage and disposal in the form of class-1 vitrified wastes. The extraction of long-lived actinide fractions and their secondary burning together with plutonium in a molten-salt burner reactor integrated with the pilot demonstration center will make it possible to reduce the volume of class-1 high-level wastes and make it possible to manage the remainder in the same manner as class-2 or, after decay, class-3 radwastes. The demonstration center incorporates a system of research chambers which can be used to organize fractionation of transuranium elements and their preparation for transfer into the ZhSR-S fuel cycle.

The operation with fuel loads containing transuranium elements requires reactor facilities that make it possible to vary the fuel composition, preserve the inherent safety properties of the reactor without changing the core design, and minimize the non-returnable losses of actinides upon recycling. The heterogeneous reactor systems with a fast neutron spectrum can be used for secondary burning of transuranium elements. However, the introduction of actinides into the fuel composition complicates the design of these reactors, leads to significant losses of fissile materials during recycling, and increases the cost of fuel fabrication. One of the most promising reactor facilities making it possible to solve the problem of burning transuranium elements is ZhSR-S with circulating fuel based on metal-fluoride melts. Its basic advantages are the possibility of widely varying the content of actinides in the fuel salt, without loss of the inherent safety property of the reactor, and the absence of

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operations associated with fuel-rod fabrication. Owing to their large separation factors between actinides and lanthanides metal fluoride melts make it possible to organize in the operation of ZhRS-S an effective system for purifying fuel salt by removing dissolved fission products and significantly reduce the fuel cycle time and irretrievable losses of actinides upon multiple recycling compared with heterogeneous reactors.

In summary, ZhSR-S, using the advantages associated with the liquid structure of the fuel, opens up prospects for significantly improving the nuclear energy technology in application to the closure of the nuclear fuel cycle on all actinides.

Molten-salt burner reactor. It is assumed that a nuclear fuel cycle with ZhSR-S will be organized as follows: the bulk of the regenerated uranium and plutonium is returned into thermal and fast reactors and the remaining transuranium elements are recycled in ZhSR-S (Fig. 1).

Since 2000, the Kurchatov Institute has been conducting comprehensive studies that have included neutronics calculations of the ZhSR-S core configurations with prescribed safety characteristics, obtaining data on the main physical and physical-chemical properties of fuel salt, study of the compatibility of fuel salt structural materials, and methods of monitoring and controlling the chemical state of molten fuel salt [1]. The experience gained thus far makes it possible to go from studying the computational and experimental possibilities of ZhSR-S to concrete technical and technological solutions.

The inherent properties of ZhSR-S are as follows:

– minimal amount of parasitic absorbers and, in consequence, smaller amounts of fissile materials in the core;

– deep burnup of fuel with minimum losses of actinides upon recycling;

– flexibility of the fuel cycle – possibility of operating with fuel having different nuclide make-up without special modifications of the core;

– on-site fuel reprocessing – temporary storage facilities for decay and transport of spent nuclear fuel are not required;

– high-efficiency owing to the high temperature of the fuel salt.

The technical feasibility of ZhSR-S is not in doubt. The configuration, materials, and characteristics of the ZhSR-S fuel loop were chosen first and foremost on the basis of considerations of technological soundness. The ZhSR-S core is of the cavity type with a fast neutron spectrum. The possibility of producing a high neutron flux density and the absence of structural materials in the core, which results in optimization of the neutron balance, as well as the possibility of adjusting the fuel composition without stopping the reactor creates favorable conditions for burning transuranium elements. The burn rate is directly proportional to the power density in the core.

In ZhSR-S, the experimentally studied melt with the molar composition $73LiF–27BeF_2$ was chosen as the solvent for the fluorides of transuranium elements. The solubility of the trifluorides of transuranium elements in it exceeds 2% with molar fraction of beryllium difluoride in the melt 0.27 and minimal temperature in the fuel loop 600°C. The calculations show that ZhSR-S with a solvent with this composition, starting on transuranium elements with the ratio Np + Am + Cm/Pu + Np + + Am + Cm = 0.1, can use as fuel makeup any composition with ratio to 0.35 without a change of the structural and temperature parameters of the fuel loop. This permits ZhSR-S with thermal power 2.4 GW to utilize up to 250 kg actinides/yr [2].

The compatibility with structural materials is comparatively easily resolved for metal-fluoride melts [1]. A nickel-based alloy of the type KhN80MTYu is compatible with fuel salt, and its corrosion depends weakly on the content of different trace impurities in the melt. Molten-salt fuel compositions based on fluorides have high radiation resistance. The saturated vapor pressure of these salts does not exceed atmospheric pressure to 1300°C. Molten-salt fluoride compositions do not enter into exothermal reaction with water, air, or other substances with which they can come into contact under the conditions of a nuclear reactor. The release of steam or gas which could lead to a rise of pressure and thereby destroy the structure of the reactor or containment is impossible in ZhSR-S. This facilitates the solution of radiation safety problems.

It is suggested that graphite or high nickel alloy KhN80MTYu be used to create the lateral and end-face reflectors of the core which are placed inside the reactor vessel. The radiation resistance of the materials of these structures determines the maximum admissible power density of the ZhSR-S core. If damage under the action of fast neutron fluence is decisive for graphite, plasticity reduction at temperatures above 500°C, associated with helium formation along grain boundaries, a process caused by fast as well as thermal neutrons, is significant for high nickel alloys. Analysis shows that in order to obtain acceptable service life of end-face and radial reflectors (at least 5 years) the specific power density must not exceed 150 W/cm^3. Otherwise, partial shutdown for reflector replacement will result in a reduction of the load factor of the reactor.

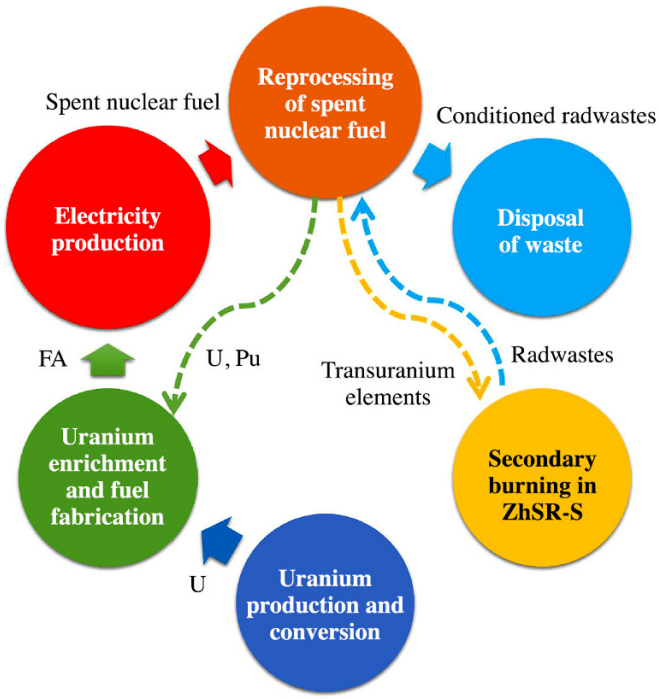

Fig. 1. Fuel cycle of a nuclear energy system with ZhSR-S.

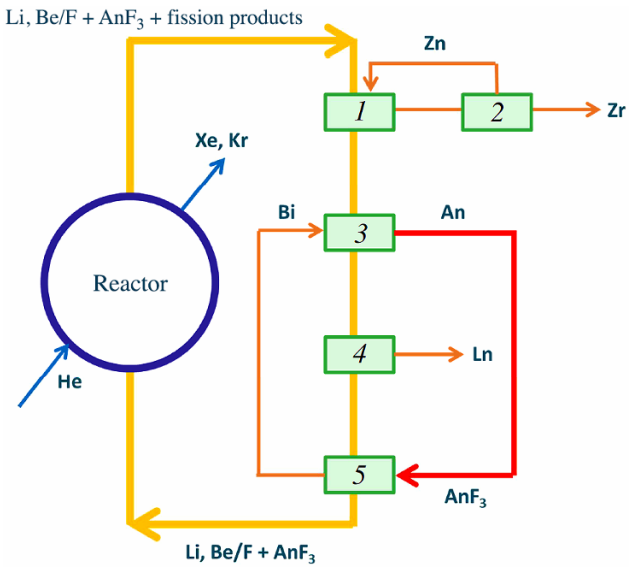

Fig. 2. Diagram of the recycling of actinides and removal of soluble fission products from the fuel salt of ZhSR-S: *1*) extraction of zirconium into liquid metal (cadmium, zinc, or bismuth); *2*) distillation of liquid metal; *3*) reductive extraction of actinides into liquid metal; *4*) purification by removal of lanthanides; *5*) recycling of actinides.

Moreover, with the given restriction on the power density the problem of heat removal from the core does not arise, and there is no need to increase the heat-resistance and high-temperature strength. The service life of the reactor vessel fabricated from the alloy KhN80MTYu is at least 50 years.

Fuel cycle. The ZhSR-S reactor comprises a complex including the nuclear reactor itself as well as nuclear fuel reprocessing systems. The volume of the fuel salts and ZhSR-S with thermal power 2.4 GW is equal to 50 m^3, and half of this

volume is located in the core. Aside from the loop with liquid fuel, there is a bypass loop in which the molten fuel salt is continuously adjusted by removal of impurities and fission products and by replenishment with fresh fuel.

The preliminary procedures of purifying fuel by removing contaminants (boron, cadmium, or lanthanides) do not pose any problems, since ordinarily they are not present in the raw material. The compounds LiF and BeF_2, as a rule, contain small quantities of sulfur in the form of the sulfate ion, fluorides of transuranium elements – oxides and oxyfluorides, and all components – water, are easily hydrolyzed to oxides and oxyfluorides at high temperatures. The methods of purification are similar to those used for preparing materials in well-known engineering solutions – treatment with a mixture of gas H_2 + HF at high temperature ~600°C and then by pure hydrogen and equipment made of nickel or copper in order to reduce sulfate to sulfide and remove it in the form H_2S, removal of Cl^- in the form HCl, and conversion of oxides and oxyfluorides into fluorides. The final treatment with hydrogen serves to reduce FeF_3 and FeF_2 to insoluble iron and remove NiF_2, which can form upon hydrofluorination. Purification can provide pure and homogeneous fuel material for the initial operation of the reactor and for fuel replenishment with fresh fuel.

The operations of adjusting the composition of the fuel salt include the continuous removal of krypton and xenon by bubbling, addition of transuranium elements in order to replace the burnt up fissile material, recycling of all actinides, removal of soluble fission products (predominantly rare-earth elements, oxides, and insoluble noble and semi-noble products of fission), and maintaining the oxidation-reduction potential of the fuel at the required level. The full cycle for reprocessing of the fuel salt is two years, which corresponds to fuel-salt flow ~70 liters/day through the reprocessing loop.

The technological scheme for reprocessing ZhSR-S fuel slat consists of three basic parts: removal of gaseous products of fission, removal of insoluble noble and semi-noble metals, and purification by removal of the fission products which are soluble in the fuel salt – lanthanides and corrosion impurities (Fig. 2).

The system removing the gaseous fission products xenon, krypton, and tritium is based on their lowest solubility in the fuel salt. Gaseous fission products rapidly migrate toward the salt–gas interface and go into the gas phase. To increase the volume of the gas phase, a bubbler saturating the fuel column with helium is installed in the bypass loop. A separator, in which the gas and liquid phases are separated, is also installed in the bypass loop. Helium goes from the separator into the reactor's gas purification system. The main problems of system removing gaseous fission products are reliable cooling of the settling tanks, periodic replacement of filters, and seal tightness of the circulating radioactive gas loop.

A nickel-based sectional filtering system is provided for collecting the hardly soluble fission products (noble and semi-noble metals) circulating in the fuel salt. The system is placed in the bypass of the reactor loop, since these fission products can form deposits on the interior surfaces of the pipelines. The main requirement of the system is to maintain low pressure losses, which is accomplished by parallelizing the main flow of the fuel salt.

Purification of the fuel solt by removing the soluble fission products and lanthanides can be accomplished by extraction into a liquid metal (cadmium, zinc, or bismuth). At the starting stage, extraction is done into liquid metal zirconium, followed by extraction of actinides and purification of the fuel salt by removing lanthanides. Reductive extraction of actinides into the fuel salt is performed at the final stage of reprocessing. The distribution ratio of the actinides and lanthanides in the system Li, Be/F–liquid bismuth with respect to plutonium at 600°C is equal to 6 for curium, 3000 for neodymium, and $25 \cdot 10^3$ for lanthanum.

Corrosion products are removed from the fuel salt by passing it through filtering systems.

The radioactive wastes formed upon reprocessing of ZhSR-S fuel can be sent to a facility of the pilot-demonstration center for conditioning as class-2 or class-3 (after decay) wastes and storage before disposal.

Conclusion. A full-scale 2.4 GW(t) ZhSR-S will make it possible to burn the yearly volume of the actinides produced at the pilot demonstration center and ~500 kg of regenerated plutonium, at the same time producing 1 GW of electricity. The technological features of ZhSR-S which are associated with continuous circulation of the fuel salt also make it possible to implement continuous distillation of highly volatile short-lived radionuclides, such as ^{99}Mo and ^{131}I, organizing production of radioisotope products. Estimates show that a 1 MW molten-salt reactor can produce ^{99}Mo with activity ~1900 TBq [3].

The development of full-scale ZhSR-S is to be preceded by the construction of an experimental reactor facility with power to 50 MW in order to demonstrate operation with different compositions of transuranium elements in the fuel salt and to perfect processes and equipment.

The technical and technological possibilities of the Mining and Chemical Combine site make it possible to place ZhSR-S in direct proximity to the spent nuclear fuel reprocessing plants, tying its infrastructure to the pilot demonstration center. The industrial site of the combine possesses the requisite infrastructure, automobile and railroad access roads, areas for storage facilities, heat, electricity, and water supply systems, and electrical networks.

The extraction of long-lived actinides from high-level wastes from reprocessing of spent nuclear fuel and their subsequent recycling in ZhSR-S will make it possible to close the nuclear fuel cycle on actinides, reduce the volume of high-level waste from reprocessing of spent nuclear fuel from VVER-1000, -1200, and reduce the cost of long-term storage and subsequent disposal and thereby increase the commercial attractiveness and public acceptability of nuclear energy.

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