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High-flux lead reactors with average neutron energies up to 1 MeV

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Abstract. The paper is devoted to the physics and technology of special nuclear reactor operating on hard neutrons. As is known, a fast neutron spectrum is realized in sodium fast reactors (SFR) at neutron flux densities up to \(8 \times 10^{15}\) neutrons/cm\(^2\)·s. In SFRs the coolant and fuel limit the average neutron energy by a value of not more than 0.5 MeV, with a relatively small fraction, 15%, of hard neutrons, \(E_n > 0.8\) MeV. Future high-flux reactors on hard neutrons, with an average neutron energy up to 1 MeV and neutron flux densities on the order of \((3-5) \times 10^{15}\) neutrons/cm\(^2\)·s, may be constructed and claimed in the medium term as multipurpose reactors that combine the functions of reactors - transmutators of minor actinides (MA), as well as isotope and research reactors. The requirements for obtaining a hard neutron spectrum in the core of the reactor lead to the need to use nuclear fuel and a coolant that slow moderate neutrons. Metallic fuel should be used as a fuel, and as a coolant - natural lead, whose isotopic composition accounts for the fraction of the slow-moderating \(^{208}\)Pb isotope a value of 52.3%. An even more harder neutron spectrum can be obtained in the case of using a coolant based on enriched at gas centrifuges \(^{208}\)Pb or radiogenic \(^{208}\)Pb extracted from lead-rich thorium ores. The next condition leading to a hard neutron spectrum is the small dimensions of core, \(D \times H \approx 0.5 \times 0.5\) meters, in which the initial neutron fission spectra of \(^{235}\)U, \(^{239}\)Pu, \(^{241}\)Pu are not strongly changed. This requirement means that reactors on hard neutrons should be classified as small power or research reactors, within which their thermal capacity does not exceed a value of the order of 200-300 MW.

1. Introduction
At present, many publications are devoted to the transmutation of minor actinides (MA) into fission products of these nuclei [1-4]. The content of \(^{241}\)Am, for example, in the composition of MOX fuel of thermal reactors (TRs) should be minimized, both for the safe handling of fuel during its fabrication, and for the safe management of the reactor. The presence of significant amounts of \(^{241}\)Am in buried high-active waste (HAW) is also undesirable due to its high heat generation and high volatility.

As is known, in one of the scenarios of the two-component (VVER + BN) system [4] of nuclear power engineering (NPE) of Russia, sodium fast reactors (BN in Russian) are assigned the role of low-level radioactive plutonium producer for MOX fuel for TRs. At the same time, BN will be supplied with power-grade plutonium obtained by regenerating fuel discharged after irradiation in VVER reactors. It is assumed that in this case, the slightly fissile MAs that are part of spent nuclear fuel (SNF) of TRs will be transferred to fission products. However, for the effective transmutation of the MA, the neutron spectrum of the core of large-scale fast reactors, sodium and lead, does not appear to be sufficiently hard: the average neutron energy in the core does not exceed 0.5 MeV [5], which limits \(^{241}\)Am fission probability by about 15%.
As a result, some of the MA does not burn out, or is converted to long-lived other isotopes, and the equilibrium content of MA in the fuel of sodium or lead FRs can be of the order of 0.4-0.7% [3, 6]. Perhaps, for several reasons, including technological ones, MA will have to be separated from the regenerated fuel, instead of leaving it in fuel for subsequent loading as is customary now in the known schemes. These MAs, taken from the SNF of large scale BNs (for example, BN-1200), together with nuclear fission products, should either be buried or burnt in a reactor with a harder neutron spectrum, in which MA fission probability exceeds 15%. According to calculations of the OKBM named after I.I. Afrikantov [6], the projected BN-1200 commercial reactor will generate up to 25 kg of MA per year. If we consider their number, about 10 BNs, which are supposed to be built in the future two-component system of NPE of Russia, and the lifetime of each BN of the order of 60 years, the mass of MA ravaged from SNF of BNs for their entire lifetime, can amount to about 15 tons. Perhaps the incineration of this amount of MA will require the creation of special MA transmutation reactors.

The paper is devoted to the possibility of creating one of such special transmutator of 25 MW thermal capacity, a high-flux reactor with a large fraction of hard neutrons in the neutron spectrum, created using innovative fuel composition and an innovative heavy liquid-metal coolant, Pb-nat.

As an innovative reactor, a reactor BRUTS-25 with a metallic fuel consisted from uranium free plutonium doped with zirconium, 58wt%Pu-power-grade - 42wt%Zr [7], is considered.

2. Description of reactors main parameters

In the study, uranium was completely excluded from the fuel composition as a chemical element with a higher inelastic neutron scattering cross section, compared to other chemical elements in the fuel and coolant: Zr, Pb and Pu. Table 1 gives the inelastic scattering cross sections of hard neutrons, with energies from 0.8 to 10.5 MeV for the chemical elements: Zr-nat, Pb-nat, \(^{235}\)U, \(^{238}\)U, \(^{239}\)Pu, \(^{240}\)Pu. These cross sections are borrowed from the ENDF B-VII.1 library and are presented in five energy groups, \( g = 1-5 \), of the Russian ABBN-93 system. Further, from the values of the cross sections in these five energy groups, the average values of the inelastic neutron scattering cross sections by the isotopes under consideration were calculated.

| \( g \) | Lower margins MeV | Upper margins MeV | Inelastic scattering of neutrons cross section, barn |
|-------|------------------|-----------------|---------------------------------------------------|
| 1     | 6.500            | 10.500          | Zr-nat 2.38149 1.01154 1.01826 0.71642 0.83171 1.03054 |
| 2     | 4.000            | 6.500           | Pb-nat 1.38157 2.22165 2.66934 1.40124 1.67949 2.06246 |
| 3     | 2.500            | 4.000           | U-235 0.62579 2.40206 3.09087 1.59455 1.82475 2.07960 |
| 4     | 1.400            | 2.500           | U-238 0.2446 2.31038 3.16768 1.53693 1.82806 2.00325 |
| 5     | 0.800            | 1.400           | Pu-239 0.20541 1.93283 2.52269 1.13524 1.55875 1.66886 |
| Average value: | | | 1.13862 0.93174 1.97569 2.49374 1.27688 1.54455 1.76894 |

It follows from Table 1 that \(^{238}\)U moderates hard neutrons more strongly than zirconium, lead and plutonium, and therefore it should be excluded from the reactor fuel on hard neutrons. The diagram of fuel composition chosen as 58wt%Pu - 4 2wt%Zr is given in figure 1 taken from [7].
From the diagram follows that plutonium fuel doped with zirconium to a level of 42% by weight, remains in the solid phase at temperatures of the order of 1400 °C and therefore can be considered as a potential fuel for small power or research reactors on hard neutrons. The structural and thermal and physical parameters of the reactor BRUTS-25 are given in table 2.

**Table 2.** Structural and thermal and physical parameters of the reactor BRUTS-25

| Parameter                                      | Value  |
|-----------------------------------------------|--------|
| Thermal capacity [MW]                         | 25     |
| Equivalent diameter of the core [mm]          | 400    |
| Height of the core, [mm]                      | 420    |
| Number of FAs in the core                     | 7      |
| Number of pins in the FA                      | 120    |
| Heat stress in the core [kW/l]                | 397    |
| Average linear load on the pin [kW/m]         | 68     |
| Mass of power-grade plutonium loaded in fuel [kg] | 92    |
| Coolant                                       | Pb-nat |
| Coolant temperature, inlet/outlet [°C]        | 450 / 530 |
| Cladding temperature [°C]                     | 610    |
3. Method of calculations
Densities of neutron fluxes at the core center of the reactor BRUTS-25, with 28 group partitioning of the neutron spectrum, were calculated using the MCNP/4B program [8] with a cross sections library based on ENDF/B-VII.1 estimated nuclear data files. Based on the obtained neutron spectra and using the same nuclear constants, the neutron and physical parameters of the reactor and the one-group nuclear cross sections of the actinide were calculated.

4. Results
Table 3 shows the results of the calculation of the neutron characteristics of the core of the reactor BRUTS-25 and the one-group nuclear cross sections of actinides in the calculated neutron spectrum of the core center of the reactor.

Table 3. One-group fission and radiation neutron capture cross sections and fission probabilities for actinides in the neutron spectra of the reactors BRUTS-25

| Parameter                                           | Value         |
|-----------------------------------------------------|---------------|
| Neutron multiplication effective coefficient, $K_{eff}$ | 1.00496       |
| Density of neutron flux in the core center [1/s/m$^2$] | $3.4 \times 10^{15}$ |
| Average neutron energy in the core center [MeV]     | 0.869         |
| Fraction of hard neutrons, $E_n > 0.8$ MeV, in the neutron spectrum [%] | 34.61         |
| $^{240}$Pu one-group fission cross section [barn]    | 0.759         |
| $^{242}$Pu one-group fission cross section [barn]    | 0.599         |
| $^{241}$Am one-group fission cross section [barn]    | 0.640         |
| $^{241}$Am one-group neutron capture cross section [barn] | 0.784         |
| $^{241}$Am fission probability [%]                  | 44.92         |

Thus, the use of an innovative metallic fuel, 58Pu - 42Zr and a heavy liquid metal coolant, Pb-nat (containing 52.3% $^{208}$Pb), in a relatively small dimensions core, $D \times H = 0.40 \times 0.42$ m$^2$, leads to the following relatively high core parameters and attractive results from the point of view of burning plutonium and americium:
• The average neutron energy in the core center - 0.869 MeV;
• Fraction of hard neutrons, $E_n > 0.8$ MeV, in the neutron spectrum of the core center - 35%;
• $^{240}$Pu one-group fission cross section - 0.759 barn;
• $^{242}$Pu one-group fission cross section - 0.599 barn;
• $^{241}$Am one-group fission cross section - 0.640 barn (0.3 barn in BNs),
• $^{241}$Am fission probability - 45% (15% in BNs).

5. Conclusions
The increased average energy of neutrons in the core, and, the most importantly, a high fraction, 35%, of the hard neutrons is provided due to the following three factors: small dimensions of the core, uranium free plutonium fuel and natural lead coolant.
To burn large quantities of MA it will be required a line of several, up to 7-8, reactors of the BRUTS. In addition to burning actinides, such reactors may be in demand for obtaining radioisotopes and studies of the durability of nuclear materials in hard neutron spectra.

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