Analysis of the attractiveness of materials as applied to the fuel cycle of high-power fast reactor of Bn-type

E.M. Lvova*, A.N. Chebeskov

JSC «SSC RF-IPPE» n.a. A.I. Lypunsky, 1 Bondarenko sq., Obninsk, Kaluga region 249033, Russia
Available online 24 August 2016

Abstract

Nuclear fuel cycle of fast reactors contains materials which potentially can be used for fabricating nuclear explosives or nuclear weapons. It is customary to apply to such materials in addressing the problem of non-proliferation of nuclear weapons and nuclear terrorism the concept of attractiveness allowing evaluating potential possibility of their use in clandestine activities. Attractiveness of nuclear materials is evaluated, first of all, according to their neutronics properties. Results are presented of analysis of attractiveness of different types of fuel compositions as applicable to fuel cycle of fast sodium-cooled high-power nuclear reactor of the BN-1200 type for different options of its starting fuel loads and utilized regimes for reaching steady-state fuel composition. The object of the present study were the simplest systems containing fuel compositions of fast reactor of BN-1200 type in the form of bare spherical assemblies without neutron reflector and assemblies surrounded with simplest neutron reflectors. Criticality conditions were determined for each system and main neutronics properties of the fuel compositions under examination were determined for these criticality conditions.

Copyright © 2016, National Research Nuclear University MEPhI (Moscow Engineering Physics Institute). Production and hosting by Elsevier B.V. This is an open access article under the CC BY-NC-ND license (http://creativecommons.org/licenses/by-nc-nd/4.0/).

Keywords: Non-proliferation of nuclear weapons; Attractiveness of nuclear materials; Fast sodium-cooled high-power reactor; Uranium oxide fuel; Uranium nitride fuel; Mixed oxide uranium–plutonium fuel; Mixed nitride uranium–plutonium fuel; Critical assembly; Neutron reflector; Critical mass; Beryllium; Tungsten.

Introduction

Power units equipped with fast reactors are capable to significantly expand the fuel resources for nuclear power generation and to reduce the volumes of accumulated radioactive wastes due to the organization of the closed nuclear fuel cycle. Only very few countries possess fast reactor technologies with Russia being the world leader in this field. It is sufficient to note the successful operation by the Beloyarskaya NPP during 35 years of BN-600 - the only working fast reactor in the world, which is, indisputably, the world class achievement. Construction of fast sodium-cooled BN-800 reactor on the Beloyarskaya NPP site, its connection to the power grids and operations for reaching 100% power level conducted as of the present moment demonstrate new progress achieved by Russia in the implementation of fast reactor technologies [1-5]. BN-800 reactor in contrast to BN-600 will be operated with MOX-fuel load and elements of the closed fuel cycle will be developed [2,6].

Nuclear fuel cycle (NFC) of fast reactors contains materials which can potentially be used for fabricating primitive nuclear explosives (NE) and, with certain additional processing, for fabricating nuclear weapons (NW) as well [3,7]. It has to be taken into consideration here that potential proliferators at the level of state will attempt to develop functional and powerful enough nuclear weapon with comparatively moderate mass and dimensions in order to be able to deliver the weapon beyond the limits of the country. [3,8–10]. Such state will, evidently, be developing technologies for additional significant processing of the nuclear material extracted from fuel cycle of nuclear power generation because without implementation of additional processing such materials have...
comparatively large values of critical mass. On the other hand, it can be assumed that the purpose of sub-national and/or terrorist groups is to fabricate primitive nuclear charge from stolen nuclear materials without their preliminary serious technological processing, because deployment of such nuclear charge is intended on the territory of the country in question and its transportation to the place of the act of terror will be achieved using all available means [4,11–15].

Nuclear materials capable to sustain chain fission reaction with comparatively modest values of critical mass are, certainly, of interest for the present analysis. Existence of critical mass is the key property of the material and is the necessary but not sufficient condition of “attractiveness” in the selection of material for its unauthorized use [16].

Besides the critical mass which must have certain realistic value allowing performing transportation of the nuclear explosive by this or that available means to the place of criminal act, neutron background and level of heat release are the important characteristics of nuclear materials from the viewpoint of their diversion. Neutron background of nuclear material is generated due to the spontaneous fission of heavy nuclei, for the most part of uranium and plutonium. Additional neutron yield takes place due to (α-n) reactions on light elements, for the most part on oxygen. With high enough neutron background the probability of initiation of nuclear explosion accompanied with significant energy yield is very low because high neutron background will inevitably result in the premature initiation of chain fission reaction, i.e. in the pre-detonation, which practically excludes the possibility of obtaining the nominal energy yield [17].

Heat release, i.e. heating of nuclear material, takes place, for the most part, due to α-decay and other processes of decay of heavy isotopes contained in the material. Significant heating of the material makes it difficult to handle it, but the main factor here are the effects of high temperature on the layer of chemical explosive directly adjacent to the nuclear material. At sufficiently high temperature this explosive loses its properties and destroys functionality of the nuclear charge. Undertaking different tricks is possible for reducing the temperature, but, however, they inevitably lead to the loss of simplicity of the design [18].

Besides the above examined characteristics of nuclear materials affecting the dimensions, mass and functionality of nuclear charge, it is necessary to mention radioactive background which practically does not affect the dimensions and functionality but complicates handling such material by potential proliferators and, at the same time, makes it easier to detect such radioactive material.

Results of analysis of attractiveness of fuel compositions of fast sodium-cooled high-power nuclear reactor of BN-1200 type according to their main neutronics properties are discussed in the present paper. Technological factors associated with additional processing of materials within the NFC are the object for subsequent studies. Calculation studies were performed using the MMKKENO code.

**Input data**

Attractiveness of nuclear materials is determined by the neutronics properties inherent to these materials and allowing initiating self-sustained chain fission reaction [16]. The following properties were examined in the present study: critical mass (M); inherent neutron background (NB); heat release (HR); radioactivity (A). The following fuel compositions which may be utilized in loading the nuclear reactor under discussion were examined: uranium dioxide (UO$_2$), uranium nitride (UN), mixed oxide uranium-plutonium fuel ((U+Pu)O$_2$, MOX) and mixed nitride uranium-plutonium fuel ((U+Pu)N, MNUP).

Isotopic compositions were determined for the neutronics calculations of the fuel compositions for the following conditions: fresh fuel loaded in the reactor core and in the blankets (fresh fuel load); irradiated fuel unloaded from the reactor after the first completed fuel residence in the core (fuel unloading).

Isotopic composition of fresh UO$_2$ fuel (%) is following: 235 U 17.8; 238 U 82.2. Isotopic composition of UO$_2$ fuel unloaded from the reactor after the first completed fuel residence in the core (%): 235 U – 9.87; 236 U – 1.75; 238 U – 74.7; 239 Pu – 2.16·10$^{-2}$; 239 Pu – 4.6; 240 Pu – 0.354; 241 Pu – 1.74·10$^{-2}$; 242 Pu – 6.33·10$^{-4}$; 241 Am – 8.07·10$^{-4}$; 243 Am – 1.93·10$^{-5}$; 237 Np – 0.134; 239 Np – 1.45·10$^{-2}$; 235 U fission products – 9.56.

Isotopic composition of fresh UN fuel is following (%): 235 U – 14.4; 238 U – 85.6. Isotopic composition of UN fuel unloaded from the reactor after the first completed fuel residence in the core (%): 235 U – 7.81; 236 U – 1.26; 238 U – 79.0; 239 Pu – 1.36·10$^{-2}$; 239 Pu – 4.11; 240 Pu – 0.266; 241 Pu – 1.29·10$^{-2}$; 242 Pu – 3.91·10$^{-4}$; 241 Am – 6.08·10$^{-4}$; 243 Am – 1.11·10$^{-5}$; 237 Np – 9.59·10$^{-2}$; 239 Np – 1.27·10$^{-2}$; 244 Cm – 5.38·10$^{-7}$; 235 U fission products – 7.55.

Isotopic composition of fresh MOX-fuel (%) is following: 235 U – 8.18·10$^{-2}$; 238 U – 82.7; 239 Pu – 0.256; 240 Pu – 10.5; 242 Pu – 4.26; 241 Pu – 1.3; 242 Pu – 0.852. Isotopic composition of MOX-fuel unloaded from the reactor after the first completed fuel residence in the core (%): 235 U – 3.65·10$^{-2}$; 236 U – 8.99·10$^{-3}$; 238 U – 73.8; 240 Pu – 0.17; 239 Pu – 10.0; 240 Pu – 4.65; 241 Pu – 0.884; 242 Pu – 0.809; 241 Am – 0.146; 243 Am – 0.103; 239 Np – 1.7·10$^{-2}$; 244 Cm – 1.84·10$^{-2}$; 235 Pu fission products – 9.3.

Isotopic composition of fresh mixed nitride uranium-plutonium fuel (MNUP-fuel) (%) is following: 235 U 8.64–10$^{-2}$; 238 U 86.4; 239 Pu – 0.202; 240 Pu – 3.35; 241 Pu – 0.973; 242 Pu – 0.669; 241 Am – 4.76·10$^{-4}$. Isotopic composition of MNUP-fuel unloaded from the reactor after the first completed fuel residence in the core (%): 235 U – 0.785; 236 U – 4.18·10$^{-2}$; 238 U – 8.68·10$^{-3}$; 239 Pu – 0.145; 240 Pu – 8.78; 240 Pu – 3.63; 241 Pu – 0.672; 242 Pu – 0.63; 241 Am – 0.14; 243 Am – 7.23·10$^{-2}$; 239 Np 1.51–10$^{-2}$; 244 Cm – 1.11·10$^{-2}$; 239 Pu fission products – 7.43.

It has to be noted that in accordance with the calculation procedure accepted in the present calculation critical
Table 1
Characteristics of critical system with uranium oxide fuel.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Fresh load</th>
<th>Unloaded fuel</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Without reflector</td>
<td>Reflecter material</td>
</tr>
<tr>
<td></td>
<td>Be 10 cm</td>
<td>W 10 cm</td>
</tr>
<tr>
<td>$R_c$, cm</td>
<td>32.20</td>
<td>21.15</td>
</tr>
<tr>
<td>$M_{ur}$, kg</td>
<td>1454</td>
<td>412</td>
</tr>
<tr>
<td>NB, 1/s</td>
<td>$1.47 \times 10^4$</td>
<td>$0.43 \times 10^4$</td>
</tr>
<tr>
<td>HR, W</td>
<td>$2 \cdot 10^{-2}$</td>
<td>0.006</td>
</tr>
<tr>
<td>A, Ci</td>
<td>0.85</td>
<td>0.24</td>
</tr>
</tbody>
</table>

Table 2
Characteristics of critical system with uranium nitride fuel.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Fresh load</th>
<th>Unloaded fuel</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Without reflector</td>
<td>Reflecter material</td>
</tr>
<tr>
<td></td>
<td>Be 10 cm</td>
<td>W 10 cm</td>
</tr>
<tr>
<td>$R_c$, cm</td>
<td>39.90</td>
<td>26.8</td>
</tr>
<tr>
<td>$M_{ur}$, kg</td>
<td>3246</td>
<td>984</td>
</tr>
<tr>
<td>NB, 1/s</td>
<td>$3.35 \times 10^4$</td>
<td>$1.01 \times 10^4$</td>
</tr>
<tr>
<td>HR, W</td>
<td>0.04</td>
<td>0.013</td>
</tr>
<tr>
<td>A, Ci</td>
<td>1.71</td>
<td>0.52</td>
</tr>
</tbody>
</table>

Table 3
Characteristics of critical system with MOX-fuel.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Fresh load</th>
<th>Unloaded fuel</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Without reflector</td>
<td>Reflecter material</td>
</tr>
<tr>
<td></td>
<td>Be 10 cm</td>
<td>W 10 cm</td>
</tr>
<tr>
<td>$R_c$, cm</td>
<td>29.50</td>
<td>20.33</td>
</tr>
<tr>
<td>$M_{ur}$, kg</td>
<td>1118</td>
<td>366</td>
</tr>
<tr>
<td>NB, 1/s</td>
<td>$1.08 \times 10^8$</td>
<td>$3.54 \times 10^7$</td>
</tr>
<tr>
<td>HR, W</td>
<td>$1.96 \times 10^3$</td>
<td>$0.64 \times 10^3$</td>
</tr>
<tr>
<td>A, Ci</td>
<td>$1.38 \times 10^6$</td>
<td>$4.52 \times 10^5$</td>
</tr>
</tbody>
</table>

Conditions of the system and its critical mass were determined in the first place. All remaining characteristics (heat release, neutron background, activity) were calculated as applicable to the critical mass of the fuel composition and with changing critical mass these values factually change proportionally to the fuel composition in question. If needed, obtaining specific characteristics is not difficult to accomplish.

Results of calculations of neutronics characteristics of critical systems with fast reactor fuel compositions

Results of calculations of neutronics characteristics of critical systems without neutron reflectors and with neutron reflectors containing fuel compositions of fast sodium-cooled nuclear reactor of BN-1200 type are presented in the present section for the preset conditions of fuel compositions under examination. Beryllium (Be) and tungsten (W) were examined as the reflector materials. Reflector thickness was accepted to be equal to 10 cm. Results are summarized in Tables 1–4.

Analysis of the calculated results obtained for “bare” systems demonstrates that critical mass exceeds one ton for the examined fuel compositions. Fresh MOX-fuel has the smallest critical mass equal to 1118 kg among the examined fuel compositions. For fresh and irradiated uranium nitride fuel critical mass is the largest among the examined fuel compositions and exceeds three tons. Under fuel irradiation in the reactor core the value of its critical mass increases due to the burn-up of main fissile isotopes and accumulation of fission products.

Comparison of fuel compositions on the basis of uranium demonstrates that for nitride fuel critical mass of system without neutron reflector is by approximately two times larger than that for oxide fuel. This can be explained by higher density of nitride fuel and, as the result, by its smaller enrichment as compared to oxide fuel. As it is known, for systems with fast neutron spectrum value of critical mass is inversely proportional to the square of the density of material of the system [19].

Approximately the same regularity is observed for mixed uranium–plutonium fuel compositions (MOX- and MNUP-fuel) as in the comparison of compositions of uranium oxide and uranium nitride fuel.

Neutron background was calculated taking into account the following two nuclear physics processes: spontaneous fission of heavy nuclei and ($\alpha$-n)-reactions on oxygen. Such reaction
on nitrogen does not produce any noticeable neutron yield. The main contribution in the neutron background is produced by even plutonium isotopes. As it was originally expected, neutron background for fresh uranium fuel for the critical system without neutron reflector has the smallest value equal to $\sim 10^4$ 1/s for the critical mass. For the examined case the value for nitride fuel is by more than two times higher than for oxide fuel. For fresh compositions of MOX- and MNUP-fuel neutron background for the critical system without neutron reflector amounts to $\sim 10^8$ 1/s for the critical mass. Such increase of neutron background as compared to uranium fuel is associated with presence of even plutonium isotopes in MOX- and in MNUP-fuel. Under irradiation of fuel compositions in the nuclear reactor values of neutron background of the critical system without neutron reflector increase for uranium fuel by approximately three orders of magnitude because of the appearance of plutonium isotopes and by approximately one order of magnitude for mixed uranium-plutonium fuel because of the increased fraction of even plutonium isotopes.

Heat release in fuel compositions is associated with presence in them of $^{238}$Pu as well as of americium and curium isotopes. However, contribution of the latter does not constitute a significant value because of their comparatively low concentration in the fuel. Because of the absence of plutonium heat release for the critical system without neutron reflector is fairly insignificant for fresh compositions of oxide and nitride uranium fuel and amounts to several hundredths of watt for the critical mass. Under irradiation of uranium fuel in the reactor heat release is significantly increased mainly due to the accumulated $^{238}$Pu isotope reaching several hundred watts for the fuel critical mass. For MOX- and MNUP-fuel level of heat release amounts for fresh fuel to the value of the order of several hundred watts and increases under burn-up reaching the highest values for spent fuel at the level of 300–500 W for the critical mass.

Activity of critical systems without neutron reflector containing fresh uranium fuel is not high and does not exceed one curie for critical mass of oxide fuel and two curies for critical mass of nitride fuel. With burn-up activity sharply increases reaching $\sim 10^8$ curies for the fuel critical mass. For systems containing fresh MOX- and MNUP-fuel activity amounts to $\sim 10^9$ curies increasing by approximately two orders of magnitude with burn-up which is valid to the same extent both for systems containing MOX-fuel and for systems containing MNUP-fuel.

Efficiency of material used as the reflector is determined by the mean free path of neutrons before scattering in this material. The smaller is this value the better is the reflector. Efficiency of neutron reflector depends not only on the material but, as well, on its thickness. The most efficient are thicknesses within the limits of three mean neutron free path values for the material in question. For larger thicknesses efficiency of neutron reflector is reduced and further increase of its dimensions does not produce noticeable gain in the reduction of critical mass of the system [20].

Beryllium is the most efficient neutron reflector. It especially efficiently reduces the critical mass for systems containing fresh fuel compositions – approximately to 30% of the critical mass for the “bare” system. For uranium oxide fuel critical mass with beryllium reflector is reduced to the values just a little larger than 400 kg. Taking into account the small neutron background, heat release and activity such fuel composition may be of certain interest for proliferators. Critical mass with beryllium reflector is even smaller for fresh MOX-fuel – it is slightly higher than 350 kg. And, although such fuel composition has significantly higher neutron background and heat release as compared with uranium fuel, it can also be of certain interest for proliferators. For nitride fuel compositions with neutron reflector made of beryllium critical masses are larger and amount to about 980 and 850 kg for uranium and for MNUP-fuel, respectively.

For fuel compositions which underwent burn-up (unloaded after the first residence in the reactor core) efficiency of beryllium as neutron reflector is somewhat lower – within $\sim 50\%$ of the critical mass for the “bare” system. Here, critical mass for practically all fuel compositions is approximately by two times larger as compared to their fresh compositions.

Efficiency of tungsten reflector in terms of reduction of critical mass is significantly lower as compared with beryllium.

It was noted in the investigation of neutron background and heat release for the examined fuel compositions that for fresh uranium fuel compositions (oxide and nitride) values of neutron background and heat release are comparatively not high and they, in all likelihood, would not be able to create any problems for potential proliferators. In this case the main hindrance will be the comparatively large value of the critical mass. However, if enrichment technology is available for the potential proliferators this hindrance can be overcome by additional enrichment of fresh uranium fuel.
to higher values with comparatively small expenditures for performing the separation work [3].

For fresh and irradiated mixed-fuel compositions (MOX- and MNUP-fuel), as well as for irradiated composition of uranium oxide and nitride fuel values of neutron background and heat release become fairly insignificant for critical assemblies with neutron reflectors and, together with increased critical mass, will represent practically insurmountable barrier if attempt is made by proliferators to use then for fabricating a nuclear explosive.

Activity of fresh uranium fuel (both oxide and nitride) is not high and does not exceed 1 Ci for critical mass of the system equipped with neutron reflector. Under burn-up activity sharply increases reaching $\sim 5 \cdot 10^7$ Ci for the fuel critical mass. For fresh mixed uranium–plutonium compositions activity of the critical system with neutron reflector amounts to $\sim 10^8$ Ci increasing by approximately one-two orders of magnitude with fuel burn-up which is true to the same degree both for critical mass of MOX-fuel and for critical mass of MNUP-fuel.

Conclusions

Calculation analysis demonstrated that the examined fuel compositions which can be used in loading high-power fast sodium-cooled nuclear reactor of BN-1200 type have large enough critical mass exceeding one ton and more. If simplest neutron reflectors are used critical mass for the examined fuel compositions is reduced by approximately two times but, nevertheless, remains to be fairly significant. Fresh oxide fuel with beryllium reflector with 10-cm thickness has the smallest critical mass among uranium fuel compositions equal to $\sim 400 \text{kg and}$, taking into account the small values of neutron background, heat release and activity, this fuel composition may represent certain interest for sub-national and/or terrorist organizations.

Attention must be attracted to the isotopic composition of plutonium accumulated in the fast reactor core when uranium fuel is used as the starting reactor fuel load. Plutonium with high concentration of $^{239}$Pu equal to $\sim 93\%$ is generated in the spent uranium fuel after the first fuel residence in the core. Rejection of the use of blankets in the fast reactor does not help resolving this problem and, factually, shifts breeding of conditioned plutonium into the reactor core with much higher productivity. Therefore, additional research of the problem under discussion is required prior to putting uranium-loaded fast reactors into operation.

As applicable to nuclear energy complex in Russia, the conditions during fabrication of uranium fuel for fast reactors, as well as during transportation of such fuel to NPPs including fuel handling operations conducted on NPP site prior to fuel loading in the reactor core and after it’s unloading from the core require special attention and protection.

References


