Fast reactors offer better usage of fissionable materials and less nuclear waste production than thermal reactors. Uranium 235 becomes scarce, and uranium-plutonium fuel cycle should come to play. Without uranium-235, the major fissionable component of the fuel is plutonium, which is not a natural resource and should be bred artificially. Plutonium is the component of the nuclear waste from light water reactors (LWRs). A substantial amount of weapon-grade plutonium has been collected up to now, as well. It could also be used in fast reactor fuel. The problem of plutonium production may arise in future when the LWRs will leave the power production market. This problem must be solved by breeding plutonium at the place. Fast reactors also produce excessive neutrons and, therefore, can breed plutonium from the uranium. However, this technology is not fully developed yet to breed plutonium in sufficient amount. In a fast reactor a problem is that a plutonium fuel has a deficit in delayed neutrons which decrease the reactor controllability [1, 2] and safety. A negative feedback is useful in such a situation because the low portion of delayed neutrons results in narrower margins for reactor control. In thermal reactors the negative feedback is provided by the Doppler effect. The value of the Doppler effect at the fast reactors is reduced, which leads to weakening of nuclear safety in the case of accident situations, such as an increase of the temperature of the fuel in the reactor core. Therefore, to enable the Doppler effect, the spectrum of fast reactors should have a significant portion of low-energy neutrons in the energy range $10^5-10^6$ eV. This leads to decrease of the average energy of neutrons at the fast reactors to about $2\times10^5$ eV, which results in substantial decrease of fission rate for a number of transuranic isotopes. This decreases burning rate of heavy transuranic elements that constitute a mostly radiotoxic part of the nuclear waste. Impact of these negative issues may be softened if the number of delayed neutrons is close to such a number in LWR.

Another approach is brought with the breed-and-burn initiative [3, 4]. The key idea is to combine breeding and burning and perform them during a single fuel cycle. Within this initiative, the activity is mainly concentrated on the ideas which provide deep burnout of the nuclear fuel. The traveling wave reactor [2] idea is to burn out more than a half of the loaded fuel. The standing wave reactor project [1, 2] also provides deep burning and has an ambition to reach a commercial application.

The breed-and-burn concept could be applied to more common technologies in which burnout of the fuel is not deep and fuel is reprocessed after 5–10 % of burnup. In connection to this, the idea is to find the transuranic elements composition to which depleted uranium is continuously supplied, and the amount of all other transuranic fuel component remains unchanged in time [5]. This feature simplifies maintaining the closed fuel cycle and provides its periodicity. Such a fuel is called here ‘balanced fuel’. For each transuranic component, the balance is achieved by equating burnout and production rates. The production is due to neutron capture by the neighboring lighter isotope and consequent beta-decay. The burnup includes fission, neutron capture and decays. The “reconditioning” of the balanced fuel that reached its radiation damage limit may be performed without separation of its transuranic components. Only separation of fission products is necessary. The fission products need to be stored then in nuclear repositories. It is important that the storage time for the fission products is much less than for the spent nuclear fuel, just about 300 years.

**Key words:** fast reactor, spent nuclear fuel, MCNPX calculations, neutron spectrum, reaction rates.
is the safest because the gas almost does not absorb neutrons and, therefore, does not affect the reactor reactivity. In addition, gas-cooled reactor has a possibility to be refueled without being shut down. This on-load refueling is beneficial in commercial sense. It should be emphasized that in this paper the authors deal with the possibility of creating a balanced fuel taking into account only nine mostly important nuclei, but for more detailed consideration this set may be substantially expanded.

1. Isotope balance

The scheme of nuclear transformations of $^{238}$U under neutron irradiation (see e.g. [7]) is shown in Fig. 1. This chain is not complete, but its accuracy is quite sufficient for calculations which are used for conceptual analysis. It is rather often applied for studying fast reactors (see e.g. [8]).

![Scheme of $^{238}$U Transformations](image)

Fig.1. Scheme of $^{238}$U transformations under neutron irradiation

Neutron captures with consequent beta decays result in increase of the nuclear number. In the model for the nuclei amount balance, the chain is started with $^{238}$U and ended at the americium isotopes $^{241}$Am and $^{243}$Am. The evolution of the isotope content is considered in frame of a single-group neutron model. The balance equations (Bateman OD equations) read:

$$
\frac{dN_i}{dt} = -\left(\sigma_f \Phi + \sigma_c \Phi + \Lambda_i\right) N_i + \\
(1-\delta_{ij})\left[(1-\delta_{1j})\sigma_{c,i}\Phi + \Lambda_{i-1}\right]N_{i-1} + \\
\delta_{ij}\Lambda_6 N_6 + \delta_{ij}\sigma_{c,j}\Phi N_3 + \sigma_{1,j} N_j. \tag{1}
$$

Here $N_i$ is the isotope content, index $i$ enumerates isotopes, $\sigma$ denotes the microscopic cross-section, indices $f$ and $c$ denote fission and neutron capture, $\Lambda_i = \ln2/T_i/T_{1/2}$ is the $\beta^-$ decay rate, $\Phi$ is the scalar neutron flux, $\delta$ is the Kronecker delta, $I$ is the fuel ($^{238}$U) supply rate. The cross-sections depend on the neutron spectrum. For this calculation, the spectrum of traveling wave reactor [9] is chosen. The element enumeration, cross-sections and decay rates are given in Table 1. Note that the calculations beta decays of $^{243}$Pu to $^{243}$Am are assumed in the calculations beta decays of $^{243}$Pu to $^{243}$Am are assumed immediate and, therefore, $^{243}$Pu is not considered.

The scalar neutron flux magnitude $\Phi$ determines the fuel burning rate. It is assumed not to vary in time. Calculations are made for two values: $\Phi = 3 \times 10^{-19}$ m$^{-2}$s$^{-1}$ and $\Phi = 3 \times 10^{-5}$ m$^{-2}$s$^{-1}$. The supply of $^{238}$U sustains the stationary content of depleted uranium in the reactor. Under such a condition the system of equations (1), which is linear in the isotope contents $N_i$, has a stationary solution.

The calculation results for two values of $\Phi$ are displayed in Table 2. The major component of the balanced fuel content in both cases is depleted uranium. Second and third in amount are the plutonium isotopes $^{239}$Pu and $^{240}$Pu. These isotopes constitute more than 99% of the fuel mixture. The stationary concentrations of them are almost the same for different scalar neutron fluxes. The low concentration of other isotopes is a consequence of the hard (highly energetic) neutron spectrum.

The infinite neutron multiplication factor is calculated following the formula:

$$
\eta_\infty = \frac{\sum_{i=1}^{9} \sigma_{f,i} N_i}{\sum_{i=1}^{9} (\sigma_{f,i} + \sigma_{c,i}) N_i}, \tag{2}
$$

where $\nabla_i$ is the number of fissile neutrons per fission reaction. For $\Phi = 3 \times 10^{19}$ m$^{-2}$s$^{-1}$, $\eta_\infty = 1.52$ is calculated and, for $\Phi = 3 \times 10^{18}$ m$^{-2}$s$^{-1}$, the neutron multiplication factor is $\eta_\infty = 1.5$. The effective neutron multiplication factor is higher than unity despite of the presence of the $^{238}$U isotope which rather captures neutrons than fissions. If the fuel consists only of three major components ($^{238}$U, $^{239}$Pu and $^{240}$Pu) and the corresponding neutron multiplication change is small: $\eta_e = 1.56$ in both cases.

The fraction of delayed neutrons

$$
\beta_{\text{eff}} = \frac{\sum_{i=1}^{9} \sigma_{d,i} N_i}{\sum_{i=1}^{9} \Phi N_i}, \tag{3}
$$

Table 1. Cross sections and decay rates of transuranic isotopes.

| Index $i$ | Element | $\sigma_f$, b | $\sigma_c$, b | $\Lambda_i$, s$^{-1}$ |
|-----------|---------|--------------|--------------|---------------------|
| 1         | $^{238}$U | 0.0421678    | 0.174319     | 4.92×10$^{-10}$     |
| 2         | $^{239}$U | 0.41         | 0.0953       | 4.92×10$^{-4}$      |
| 3         | $^{239}$Np | 0.596865     | 0.958529     | 3.4×10$^{-6}$       |
| 4         | $^{239}$Pu | 1.64102      | 0.199029     | 9.12×10$^{-13}$     |
| 5         | $^{240}$Pu | 0.43464      | 0.28688      | 3.38×10$^{-12}$     |
| 6         | $^{241}$Pu | 1.88931      | 0.211814     | 1.54×10$^{-9}$      |
| 7         | $^{242}$Pu | 0.298487     | 0.43667      | 5.89×10$^{-14}$     |
| 8         | $^{243}$Am | 0.256523     | 0.43667      | 2.99×10$^{-12}$     |
| 9         | $^{241}$Am | 0.325332     | 1.01117      | 5.08×10$^{-11}$     |

Table 2. Equilibrium concentrations and radioactivity of transuranic isotopes for two values of scalar neutron flux.

| Index $i$ | Element | Conc. | Radioactivity, Bq/kg | Conc. | Radioactivity, Bq/kg |
|-----------|---------|-------|----------------------|-------|----------------------|
| 1         | $^{238}$U | 0.888 | —                    | 0.889 | —                    |
| 2         | $^{239}$U | 1.09×10$^{-6}$ | 1.19×10$^{15}$ | 1.09×10$^{-7}$ | 1.19×10$^{14}$ |
| 3         | $^{239}$Np | 1.57×10$^{-4}$ | 1.18×10$^{15}$ | 1.57×10$^{-5}$ | 1.19×10$^{14}$ |
| 4         | $^{239}$Pu | 8.39×10$^{-2}$ | 1.69×10$^{11}$ | 8.4×10$^{-2}$ | 1.69×10$^{11}$ |
| 5         | $^{240}$Pu | 2.31×10$^{-2}$ | 1.73×10$^{11}$ | 2.28×10$^{-2}$ | 1.71×10$^{11}$ |
| 6         | $^{241}$Pu | 2.6×10$^{-3}$ | 8.9×10$^{12}$ | 10$^{-3}$ | 3.42×10$^{12}$ |
| 7         | $^{242}$Pu | 1.04×10$^{-3}$ | 1.36×10$^8$ | 4.02×10$^{-4}$ | 5.25×10$^7$ |
| 8         | $^{243}$Am | 3.46×10$^{-4}$ | 2.29×10$^9$ | 1.31×10$^{-4}$ | 8.72×10$^8$ |
| 9         | $^{241}$Am | 8.55×10$^{-4}$ | 9.64×10$^{10}$ | 3.0×10$^{-3}$ | 3.38×10$^{11}$ |
of delayed fission neutrons per fission reaction) is almost the same for slow and fast fuel burning, i.e. $\beta_{\text{eff}} = 0.46\%$. This number is higher than the number of delayed neutrons for plutonium isotopes. Although $^{238}$U fission rate is small, $^{238}$U has a relatively large fraction ($b_{1/2,238} = 1.5\%$) of delayed neutrons and its contribution to $\beta_{\text{eff}}$ is substantial. Without uranium-238 the fraction of delayed neutrons is more than twice smaller, $\beta_{\text{eff}} = 0.21\%$.

The radioactivity of the fuel is high. The major radioactive components are $^{239}$U and $^{239}$Np, but they are short-lived. The major contribution to the radioactivity of the remainder is due to $^{241}$Pu (95% and 86% for two neutron flux values). Its decay time is medium, 14.3 years.

2. MCNPX calculations for nuclear reactor

To check a possibility to use the balanced fuel in a nuclear reactor, the MCNP calculations are employed. The nuclear reactor is modeled as a composition of its principal parts. The major simplification of the model is that each part is represented by a uniform mixture of its components.

The calculation are performed for a cylindrical reactor shown in Fig. 2. The feature of this reactor is usage of mixed-oxide fuel (MOX). The gas cooling is employed that simplifies fuel temperature control. In the model, the reactor core has 110 cm in radius and 400 cm in height. Such dimensioned reactor core is comparable in industrial reactors with large core.

A shield is used to reflect neutrons and to reduce the neutron and gamma loads of the surrounding area. The shield contains a 60:40 vol.% mixture of the stainless steel alloy S30467 type 304B7 (see Ref. 11) with water. The steel contains 1.75 wt.% of natural boron.

The neutrons are absorbed by boron and, since boron absorbs mostly slow neutrons, they should first be slowed down, which is provided by the water in the vessel. Boron has a stable isotope $^{10}$B which absorbs neutrons very efficiently: the absorption cross section of thermal neutrons is about 4000 barn. After absorption of a neutron, the excited nucleus $^{11}$B is formed which immediately decays into the stable nucleus $^7$Li and an alpha particle.

As well as other parts of the reactor, the core is represented in the model as a homogenized mixture of MOX fuel and coolant. Since the coolant (helium) density is low, in the model its concentration will be small. In these calculations, contribution of helium coolant to nuclear processes is ignored for the reason of its too small density. Properties of oxide fuel ensuring its widespread usage in nuclear reactors are the high melting point (~3073 K), high endurance to the radiation damage and chemical stability over a wide temperature range. However, the MOX fuel has drawbacks — a low thermal conductivity and low density of fissile components. Low density of the oxide fuel results in necessity of increasing volume occupied by the fuel, and low thermal conductivity leads to high temperature of the fuel. These disadvantages of oxide fuel are less restrictive in industrial reactors with large core.

For calculations, a simplified fuel model which accounts for the nine above mentioned isotopes of uranium, neptunium, plutonium and americium is used. For initial calculations the fuel mixture given in Table 2 supplemented with oxygen is used (see Table 3, column “Initial fuel mixture”). For the reactor model, numerical calculations of kinetics of neutrons are performed with the code MCNPX [12].

| Element | Cross section (MCNPX) | Concentration, wt% |
|---------|-----------------------|--------------------|
| $^{238}$U | 0.052224 | 0.217036 | 0.784685 | 0.7571993 |
| $^{239}$U | 1.1125 | 0.449688 | 1.09×10^{-6} | 1.3098311×10^{-6} |
| $^{239}$Np | 0.518584 | 1.54867 | 1.57×10^{-4} | 1.8839721×10^{-4} |
| $^{241}$Pu | 1.691649 | 0.364 | 0.07414 | 0.079766757 |
| $^{240}$Pu | 0.423626 | 0.423626 | 0.020414 | 0.0408094876 |
| $^{241}$Pu | 2.26493 | 0.37749 | 0.002305 | 0.003794 |
| $^{242}$Pu | 0.2979 | 0.43332 | 0.00104 | 0.00221 |
| $^{241}$Am | 0.3114754 | 1.409836 | 8.55×10^{-4} | 1.0945665×10^{-3} |
| $^{243}$Am | 0.228758 | 1.30719 | 3.46×10^{-4} | 6.2385923×10^{-4} |
| $^{16}$O | — | — | 0.11605691 | 0.11425755 |
| $^{4}$He | — | — | 0.00005445 | — |

The isotope composition of the balanced fuel depends on the fuel spectrum, and refining of the component percentage is necessary because the neutron spectrum in the above-described reactors is not known beforehand. First, the initial fuel composition is used for calculations, and the fission and neutron capture cross sections (see Table 3) are computed for each isotope.

Next, these cross sections are used in the above-described zero-dimensional model instead of cross-sections taken from travelling wave reactor. Then, a new balanced fuel composition is found (see Table 3, column “Final fuel mixture”). This fuel composition was set in the program MCNPX again for further neutron calculations. Such an iteration could be repeated again, but even the single step results in the sufficient accuracy. Figure 3 shows dependence of the effective neutron multiplication factor on the density of the fuel and the gas coolant averaged over the reactor core. It is seen that the criticality of the system is achieved, $k_{\text{eff}} = 1$, when the fuel mass density of the homogeneous mixture equals $2.5×10^3$ kg/m$^3$, which is much less than the fuel mass density.

Figure 4 shows the energy group fluxes (neutron flux integrated over energy intervals) per one fission source neutron averaged over
the reactor core. As it is seen from the figure, the slow neutrons in the core are minimal, while the major part of the spectrum represents the fission spectrum with energy $10^3$–$10^7$ eV.

Besides the energy neutron spectrum, the rates of nuclear reactions are calculated. The MCNPX calculates a reaction rate following the formula:

$$R = N \int \sigma(E_n) \varphi(E_n) dE_n,$$

where $\varphi(E)$ is the energy-dependent fluence per one source neutron (m$^{-2}$), $\sigma(E)$ is the energy-dependent microscopic reaction cross section (barn), $N$ is the atomic density of material (atoms⋅m$^{-3}$). To find a negative contribution to the balance of an isotope, the sum of fission rate and neutron capture rate is calculated. It represents decrease of this isotope content, but without account for the spontaneous decay. Increase of the isotope content is represented by the neutron capture rate of lower isotope. The beta decay of lower isotope is assumed to be fast. The diagram of Fig. 5 displays the calculated decrease and increase rates per one fission neutron for isotopes contained in the fuel of the present model.

For $^{238}$U and $^{241}$Am isotopes, only decrease rate is given since uranium-238 is a primary element in the chain of transformations, and americium-241 has a different mechanism of appearance [13], see Fig. 1. For all other isotopes, decrease is compensated by increase to high degree, and amount of each isotope remains unchanged in time.

The estimate of reactor power is based on the cooling capabilities. Following them, the thermal power is about $5 \times 10^7$–$10^8$ W. With such a power the fuel with 10% of burnup should be reloaded every 5th year. It is assumed that temperature regime of the reactor is similar to other reactors with ceramic fuel.

### 3. Small nuclear reactor with metallic nuclear fuel

Besides the energy producing reactor, a small size fast reactor for research purposes is of interest. Its design is similar to the above considered, but the sizes are smaller. As shown in Fig. 6 the radius of the reactor core of the model was taken 0.5 m and the height was taken 1.5 m. Composition and size of the reflector and shield remain unchanged.

The core of the reactor is represented in this model as a homogenized mixture of the metallic fuel and the coolant. Metallic fuels have good thermal conductivity and a higher density compared with the oxide fuel. Therefore, this fuel can be used in small scale reactors rather than in industrial scale reactors. The metallic fuel has certain drawbacks:

- low melting point (~1293 K);
- when the fuel temperature ~973 K, it may form an eutectic with the steel cladding;
- swelling caused by the neutron irradiation.

Swelling rate of the metallic uranium caused by neutron irradiation is several times higher than of the oxide fuel that conditionality significant contribution swelling gas at temperatures of 823–873 K of fuel, due to the high creep of metallic uranium at these temperatures. High-speed fuel swelling leads to a high level of fuel pressure in the cladding, which significantly limits the acceptable level of fuel burnup.

Helium is also used as a coolant in the small reactor. The isotopic composition of the metallic fuel for small reactor was found in the same way as for bigger reactor (see Table 3). The difference in isotope concentrations between initial and
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Final fuel mixtures is smaller because the oxygen is not present in the core of the reactor.

Figure 7 shows the dependence of the effective neutron multiplication factor on the averaged mass density of the fuel and coolant in the reactor core. The criticality of the system is achieved ($k_{eff} \approx 1$) when the fuel-coolant mixture density equals $6.3 \times 10^3$ kg/m$^3$.

Figure 8 shows the energy group fluxes (neutron flux integrated over energy intervals) per fission source neutron averaged over reactor core. Figure 8 also indicates that the major part of the spectrum represents the fission spectrum with energy $10^3$—$10^7$ eV.

The diagram of Fig. 9 displays the calculated decrease and increase rates per one fission neutron for isotopes contained in the fuel of the present model. Figure 9 shows that the quantity of increase rate of different isotopes by dint of capture neutron of lower isotope is almost equal to the decrease rate of the same isotopes via fission and neutron capture reactions.

The estimate of reactor power is based on the cooling capabilities. Following them the thermal power is about $5 \times 10^6$ W.

4. Summary and conclusions

A uranium-based nuclear fuel and closed fuel cycle are proposed for energy production. The fuel composition is chosen so that during reactor operation the amount of each transuranic component remains unchanged since the production rate and burning are balanced for it. Only $^{238}$U content has a tendency to decrease and, to be kept constant, must be sustained by continuous supply. Other fuel components play the role of catalyst in process of $^{238}$U utilization for energy production.

The spent nuclear fuel of such a reactor should be reprocessed and used again after separation of fission products and adding

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**Table 4. Isotope composition**

| Element | Concentration, wt% |
|---------|--------------------|
|         | Initial fuel mixture | Final fuel mixture |
| $^{238}$U | 0.888 | 0.900 |
| $^{239}$U | $1.09 \times 10^{-6}$ | $2.0 \times 10^{-6}$ |
| $^{239}$Np | $1.57 \times 10^{-4}$ | $1.57 \times 10^{-4}$ |
| $^{239}$Pu | 0.0839 | 0.07181 |
| $^{240}$Pu | 0.0231 | 0.0231 |
| $^{241}$Pu | 0.0026 | 0.0026 |
| $^{242}$Pu | 0.00104 | 0.00104 |
| $^{241}$Am | $8.55 \times 10^{-4}$ | $8.55 \times 10^{-4}$ |
| $^{243}$Am | $3.46 \times 10^{-4}$ | $3.46 \times 10^{-4}$ |
| $^4$He | — | $0.000019$ |
depleted uranium. This process seems less costly than isotope separation. The composition of such a fuel (balanced fuel) can be found with a model for evolution of the isotope content. The problem is, in fact, self-consistent since the fuel composition depends on the neutron spectrum and vice versa. However, as our calculations show, one iterative step is sufficient to achieve decent accuracy. The variation of the fuel composition depending on the reactor chosen is not too big. The calculated fuel composition consists mainly of uranium with minority of plutonium isotopes. The effective neutron multiplication factor in infinite media is higher than unity, $k_{ef} > 1.5$. The portion of delayed neutrons is bigger than in plutonium fuel, which allows one to keep high-energy neutron spectrum without controllability and safety problems.

Using MCNPX numerical code, the calculations are performed for the reactor of industrial nuclear power plant size with MOX fuel and for a small reactor with metallic fuel. The calculation results indicate that it is possible to achieve it needs a separate consideration. Meanwhile, we give a few words about it. The base for the start-up fuel may be plutonium taken from the spent nuclear fuel of LWRs (light water reactors). However, the quantity ratio of the major isotopes of plutonium, $^{239}$Pu and $^{240}$Pu, in it differs from the necessary one. In such a case, addition of weapon-grade plutonium seems to be expedient. Usage of enriched uranium is an alternative.

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Ядерний топливний цикл з мінімальними відходами

Moisseenko V. E., Черницький С. В.

Для виробництва енергії пропонується ядерне топливо на основі урану та топливний цикл. Топливна композиція обрана так, що процес попередньої переробки ядерного топлива — переробки та використання — може бути здійснено без значних витрат, тому що вона не потребує додавання обедненого урану. Це спрощує замкнений ядерний топливний цикл і забезпечує його періодичність. Виходячи з цього, ми вважаємо, що наведені результати можуть бути застосовані до практики.

Ядерний паливний цикл з мінімальними відходами

Moisseenko V. E., Черницький С. В.

Для виробництва енергії пропонується ядерне топливо на основі урану та топливний цикл. Паливна композиція обрана так, що процес попередньої переробки ядерного топлива — переробки та використання — може бути здійснено без значних витрат, тому що вона не потребує додавання обедненого урану. Це спрощує замкнений ядерний топливний цикл і забезпечує його періодичність. У розрахунках паливного балансу використовуються 9 изотопів урану, плутоній та підгрупі урану, що дозволяє отримати точні результати. В результаті це дозволяє отримати точні результати. В результаті це дозволяє отримати точні результати. В результаті це дозволяє отримати точні результати. В результаті це дозволяє отримати точні результати.