https://doi.org/10.46813/2022-142-040 MULTIPLE RECYCLE FUEL CYCLE FOR SPENT NUCLEAR FUEL COMPONENTS INCINERATION IN FUSION-FISSION HYBRID

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A multiple recycle fuel cycle (MRFC) is analyzed using a simple numerical model. A straightforward approach to MRFC has some unfavorable features like strong variation of the neutron multiplication factor and accumulation of americium isotopes which would likely hamper its practical usage. A solution proposed here is addition of ²³⁸U both to initial fuel and the recycled fuel.

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INTRODUCTION

The problem of accumulation of spent nuclear fuel (SNF) is of global importance. This process continue and the amount of SNF is grow sincrementally since it remains almost untouchable in the geological storages. Such a treatment of SNF has serious disadvantages and risks. Some of them are:

- Risk of theft and usage of SNF components for antihumanitarian purposes, such as dirty bomb, etc.;
- Risks of accidents and consequent poisoning of surrounding environment;
- Extremely long storage time, about 300 000 years, and huge associated expenses;
- Waste of useful resources. Fissile and fertile elements which constitute the majority of SNF could be used for energy production in nuclear power plants;
- The current way of handling with SNF conflicts with basic principles of a sustainable society.

The alternative method of SNF handling is its nuclear incineration. There are several approaches to how to implement incineration. Here the transmutation of the transuranium content of SNF is considered. This content can be obtained after extraction of uranium, fission products and slug material from SNF. The transuranium content is the most radioactive component of the SNF. Uranium is almost not radioactive. The fission products decay relatively quickly, for about 300 years. They contain plenty of chemical elements including some quite valuable. Then, using the obtained transuranic substance, a nuclear fuel is made.

Burning of the transuranic mixture can be performed in fast reactors only. Non-moderated fission neutrons are necessary to provide efficient fission for all transuranic elements. In the MeV range of neutron energies which is occupied by fission neutrons, the fission cross-sections are much lower than for fission of ²³⁵U with thermal neutrons. This is the reason to use fuel with highest concentration of fissile material. It is expected that the percentage of burning will be less than in the light water reactors because of bigger influence of neutron capture by fission products and influence of high level DPA on physical properties of fuel.

In these conditions, the only way to achieve full SNF incineration is realization in a multiple recycle fuel cycle (MRFC) [1]. This fuel cycle consists in burning of the fuel in a fast nuclear reactor, then, after the normalization of heat release, reprocessing it with separation of fission products, adding fissile materials (in our case this is transuranium content of SNF), fabricating new fuel from this composition and repeating these all in a cyclic way.

All critical fission reactors are controlled essentially due to existence of the delayed neutrons. They are only in a small fraction, and this is a physical limitation for critical reactors which has a large negative impact on their safety. The fuel based on the transuranium content of SNF has much less delayed neutrons than the fuel for the light water reactors. So, the only possible solution that provides sufficient safety is a sub-critical system. Among other proposals, the stellarator-mirror (SM) [2] seems more suitable for industrial hvbrid transmutation. The device is aimed for steady-state operation. In a subcritical system, neutron reactions are controlled by an external neutron source (in our case by a plasma neutron generator), which substantially increases the safety and also provides more flexibility in the control. From a safety point of view, these externally generated neutrons essentially replace the actions of delayed neutrons and can drastically improve reactor safety.

The MRFC realization with the SM hybrid is considered in these studies. The paper is composed of the following sections. A simple environment-independent 0D model aimed to investigate the basic properties of the MRFC for fast reactors is presented in the next section. The result of calculations for straightforward application of MRFC are presented and analyzed in the Section 2. An improved version of MRFC is proposed and discussed in the Section 3. The last Section gives summary and proposals for future research.

1. NUMERICAL MODEL

A chain of nuclear transformations caused by neutron irradiation of uranium-238 is displayed in Fig. 1 (see, e.g. [3]).



Fig. 1. Scheme of ²³⁸U transformations under neutron irradiation

The chain is generated and sustained due to neutron capture and beta-decay reactions. In this chain the most plutonium and americium isotopes of SNF are present. The ²³⁷Np isotope is not present, and this makes the scheme simplified. The main additional segment of the chain responsible for this isotope is ²³⁷Np \rightarrow ²³⁸Pu \rightarrow ²³⁹Pu. So, its influence consists in slight extra supply for major SNF isotope ²³⁹Pu. This does not have radical influence on the incineration process.

Another simplification is cut of the chain on two americium isotopes. It is worthwhile if the concentration of both of them is small. If not, the accuracy of the scheme is reduced.

To analyze possibilities given by the MRFC, we introduce a simple 0D isotope balance model [4]. The nuclear reactions are described by the following equations (1):

$$\frac{dN_i}{dt} = -\sigma_{c,i}\Phi N_i - \sigma_{f,i}\Phi N_i - \Lambda_i N_i + \Lambda_l N_l + \sigma_{c,m}\Phi N_m + I_i.$$
(1)

Here N_i is the isotope normalized content, index *i* enumerates isotopes, σ denotes cross-sections, indices *f* and *c* denotes fission and neutron capture, $\Lambda_i=\ln 2/T_{1/2,i}$ is the decay rate of *i* type nucleus, $T_{1/2,i}$ is the half-life for beta-decay, Φ is the scalar neutron flux, *I* is the fuel supply (sink) rate. Indices *l* and *m* have values that correspond to processes displayed in Fig. 1.

The chosen cross-sections [4] are representative for a fast reactor. The fission products quantity is counted by the number the source nuclei (2):



Fig. 2. Time evolution of k_{eff} *, fission products (FP),* ²³⁸U, ²³⁹Np, ²³⁹Pu (left chart), ²⁴⁰Pu, ²⁴¹Pu, ²⁴²Pu, ²⁴¹Am and ²⁴³Am (right chart) for first, fifth, twentieth and hundredth time periods (k=1, k=5, k=20, k=100)

Fuel isotope content

Isotope	FP	²³⁸ U	²³⁹ Np	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu	²⁴¹ Am	²⁴³ Am
Quantity	0.0	0.0	0.0	0.574	0.0226	0.0042	0.005	0.0098	0.0011

Note that in the calculations beta decays of ²³⁹U to ²³⁹Np and ²⁴³Pu to ²⁴³Am is assumed immediate. Thus, ²³⁹U and ²⁴³Pu are not considered. The neutron capture processes are blocked for two isotopes, ²⁴¹Am and ²⁴³Am, on which the nuclear chain ends.

The calculations were performed for fixed neutron flux $\Phi = 0.0002 \text{ barn}^{-1}\text{day}^{-1}$ with the periods of 600 days. At the end of each period a part the fission products (50 %) is substituted with the corresponding quantity of fuel. Also, the efficient neutron multiplication factor, k_{eff} , the ratio of neutron production and loss rates, arecalculated. In the computations, the computed k_{eff} values neglects neutron losses in slug material, coolant, etc, and therefore values of k_{eff} exceeding unity will result from the calculations.

2. MRFC WITH Pu-Am FUEL

The MRFCprocess is realized with the fuel made of spent nuclear fuel by deletion of uranium and fission products. Its isotope content is given in Table. With the same start-up fuel, the time evolution of fuel components is displayed in Fig. 2 for first, fifth, twentieth and hundredth time periods (k = 1, k = 5, k = 20, k = 100). These time evolutions do not reveal any complex behaviors.

The solution of the system of equations (1) and (2) depends on initial conditions. Nevertheless, all the solutions converge to the same periodical process in which the initial fuel for each cycle is identical. This happens not fast, just for k > 100.

As it is seen in the Fig. 2, the main fissionable isotope ²³⁹Pu content decreases greatly. ²⁴⁰Pu and ²⁴¹Pu does not variate much. ²⁴²Pu and americium isotopes increase significantly. ²⁴¹Am reaches very high concentration, up to 25 % which is quite bad. This isotope is radioactive, as well as associated with it ²⁴²Cm and ²³⁸Pu which are not accounted for in this simple model (1) and (2). Very high concentration of ²⁴¹Am causes problems for efficient reprocessing of the fuel. Another serious deficiency of this version of MRFC is big variation of k_{eff} from cycle to cycle: it reduces from 2.36 at the first cycle to 1.88 at the hundredth cycle.



Fig. 3. Time evolution of k_{eff} , fission products (FP), ²³⁸U, ²³⁹Np, ²³⁹Pu (left chart), ²⁴⁰Pu, ²⁴¹Pu, ²⁴²Pu, ²⁴¹Am and ²⁴³Am (right chart) for MRFC with U-Pu-Am fuel

3. MRFC WITH U-Pu-Am FUEL

Decreasing the americium concentration in the fuel and mitigating the variation of k_{eff} can be achieved by adding ²³⁸U. We have consider a fuel consisting in 52 % of ²³⁸U and 48% of the isotopic content given in Table 1 as an addition made at each cycle. The start-up fuel at the first cycle has even more ²³⁸U, i.e. 75 %.

The calculation results for this case are presented in Fig. 3. The addition f^{238} U suppress k_{eff} variations. Now they are less between cycles than within an individual cycle.

However, the value of k_{eff} is decreased, and this is a result of the ²³⁸U presence in the fuel. The transient ²³⁹Np and the main fission making isotope ²³⁹Pu do not vary much which is connected to the k_{eff} stability. All heavier isotopes increase from cycle to cycle. This rise saturates at high cycle numbers k>100. As compared with the fuel cycle described in Section 2, the concentrations of the americium isotopes reduces approximately 4.5 times for each. This is because (i) their amount is twice less in the supplied fuel and (ii) the concentration of parent plutonium isotopes which generate americium is less.

CONCLUSIONS

The multiple recycle fuel cycle application seems the only possibility to incinerate fully the transuranic isotopes that reside in the spent nuclear fuel. The incineration is safer in a sub-critical fast nuclear reactordespite deficit of delayed neutrons in the transuranic fuel. One of the possibilities is usage of the stellarator-mirror fission-fusion hybrid. The MRFC had been analysed in this paper using a simple numerical model.

A straightforward approach to MRFC is to use transuranic mixture extracted from the spent nuclear

fuelboth as an initial fuel and the fuel addition during the cycling. This approach seems possible, but some unfavourable features like strong variation of the neutron multiplication factor k_{eff} and accumulation of americium isotopes would likely hamper its practicability.

A solution proposed here is addition of 238 U both to initial fuel and recycle fuel. In each sequence 238 U is added in different proportions. The variations of k_{eff} are reduced to minimum values. The americium content is decreased in 4.5 times in our calculations.

The proposed MRFC is likely prospective but needs more detailed studies.

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БАГАТОРАЗОВИЙ ПАЛИВНИЙ ЦИКЛ ДЛЯ СПАЛЮВАННЯ КОМПОНЕНТІВ ВІДПРАЦЬОВАНОГО ЯДЕРНОГО ПАЛИВА В ГІБРИДНОМУ ЯДЕРНО-ТЕРМОЯДЕРНОМУ РЕАКТОРІ

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Багаторазовий паливний цикл (БРПЦ) було проаналізовано за допомогою простої числової моделі. Прямий підхід до БРПЦ має деякі несприятливі особливості, такі як сильна варіація коефіцієнта розмноження нейтронів і накопичення ізотопів америцію, ймовірно, перешкодить його практичному використанню. Рішення, запропоноване в цій статті, полягає в додаванні ²³⁸U як до початкового палива, так і до рециркуляційного палива.