PHYSICAL CHARACTERISTICS OF THE LIGHT WATER REACTOR CORE FUELLED WITH (Th+Pa+U)-CERAMICS (HIGH FUEL BURN-UP VIA DUPIC PROCESSES APPLICATION)

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ABSTRACT

Physical characteristics of light water reactor loaded with (Th-Pa-U)-fuel are studied. It is shown that the admixture of ^{231}Pa nuclide into fuel composition makes it possible to achieve high fuel burn-up due to stabilization of the neutron multiplying properties during fuel irradiation. The following chain is accomplished in such a fuel: $^{231}\text{Pa}\rightarrow{}^{232}\text{U}\rightarrow{}^{233}\text{U}$. Here, the first nuclide plays a role of an effective burning absorber, the second – a fissile nuclide with moderate neutron multiplying properties, and the third – a fissile nuclide with excellent neutron multiplying properties. Consecutive transmutations of these nuclides allow to improve neutron-multiplying properties of fuel under irradiation. So, the burn-up 25-30%HM can be achieved if in the fuel it is contained 10-15%: ^{231}Pa. Achieving high fuel burn-up corresponds to fuel residence time ~ 18 years for PWR-type reactor. It is shown that a fuel residence time of 50 years can be achieved due to reducing power density in the reactor core by a factor of 2.75. An appropriate displacements per atom (dpa) of structure material (stainless steel) for high burn-up fuel is as much as 63.5 dpa and fast neutron fluence $1.27\cdot 10^{+23}$ n/cm^2 (E_n>0.1 MeV). As it is known, such kind parameters have been already achieved in liquid metal fast breeder reactors quite successfully for fuel elements with stainless steel cladding. The long fuel residence time of the fuel containing ^{232}U (product of ^{231}Pa neutron capture) leads to essential helium accumulation in the fuel because of α-decays of ^{232}U and its daughter nuclides. It appears that this problem might be resolved by means of the DUPIC-type technology periodically removing gaseous substances from fuel and continuing the reactor operation.

1. INTRODUCTION

There is a well-known and very attractive idea, concerning the creation of a nuclear power reactor, which would not require refueling during the entire period of reactor operation («black box» reactor)[1]. However, there are many difficulties with this concept.

[1] Special nuclear reactors or reactors as an autonomous sources for remote small customers of energy are not had in view here.

Generally speaking, the fuel residence time might be prolonged due to reducing power density. Also, reduction of power density is a measure that enhances reactor safety (however, it can lead to increased capital investment). Nevertheless, some contemporary projects of advanced pressurize light water reactors imply reduced values of power density, and these reactors remain fairly competitive. So, for example, the project of the upgraded safety reactor VPBR-600 implies mean power density of about 69 MW/m^3 [1], while this value for the VVER-1000 reactor (PWR-reactor) is equal to 110 MW/m^3.

An almost sure action leading to prolongation of the fuel residence time is an increase of the fuel burn-up. It is well known that fuel burn-up is restricted by either reactivity margin or by integrity of fuel elements. In the power reactors under operation fuel burn-up is mainly restricted by the reactivity margin and equal to 4-6% HM. In fast reactors the achieved value of fuel burn-up exceeds 10% HM, in some experimental fuel elements loaded in the research reactor BOR-60 it was achieved the burn-up 32% HM [2]. So, it may be concluded that there are strict arguments in favor of the assumption that if fuel elements like those used in fast reactors would be used in thermal light water reactors, then the fuel residence time in these light water reactors might be substantially prolonged (by a factor of several times). In this case, the main problem is to form a fuel composition that application would provide to maintain its neutron multiplying properties at a level sufficient to achieve high fuel burn-up.

Below, a ceramic fuel based on the nuclide mixture ^{232}Th-^{231}Pa-^{233}U is considered, and the possible stabilization of neutron multiplying properties up to high fuel burn-up in pressurize water reactors is demonstrated. If neutron multiplying properties are stabilized for fuel burn-up higher than experimentally achieved values in fuel elements of fast reactors (20-30% HM [2]) the possible application of DUPIC-type technology (DUPIC is an abbreviation for Direct Use of Spent PWR Fuel In CANDU) to re-fabricate fuel elements and to continue irradiation in reactor is discussed.

2. MATHEMATICAL MODEL AND ADOPTED APPROXIMATIONS

The computer code package SCALE-4.3 [3] developed for neutronics analysis of light water reactors was used in numerical calculations. Using the code, a one-dimensional infinite lattice of fuel elements and time behavior of the fuel composition was analyzed. Neutron transport within a reactor cell was calculated in P_8-approximation with a number of internal iterations sufficient to assess K_∞ to an accuracy of 10^{-5}. The fuel composition was re-calculated with a time interval of 4 years in order to assess value K_∞ to an accuracy of 10^{-4}. Forty-three fission products set of higher reactivity impact were taken into account. The calculations were carried out in 44-group energy approximation using evaluated nuclear data file ENDF/B-IV [4], in which the number of fission neutrons $\nu_f(^{233}U)$ was reduced from 3.13 to 2.456 according to recommendations [5].

3. NEUTRONICS PREREQUISITES FOR HIGH FUEL BURN-UP

In this study a light water lattice of the VVER-1000 reactor type with high (20-30% HM) fuel burn-up is analyzed. To achieve such a fuel burn-up, certain problems have to be resolved. These problems include production of special fuel, analysis of reactor safety and technical support for high fuel burn-up. These problems are the subjects of further consideration.

Fresh fuel of currently operating power reactors always contains both fissile and fertile nuclides. Under neutron irradiation, nuclides with prevalent neutron multiplying properties – so called fissile nuclides (e.g. ^{233}U, ^{235}U, ^{239}Pu, ^{241}Pu) then alternate with nuclides that have prevalent neutron absorbing properties – so called fertile nuclides (e.g. ^{234}U, ^{236}U, ^{240}Pu, ^{242}Pu). This means that, if a fissile nuclide captures a neutron without fission, i.e. the nuclide is not used at its destination, it transmutes into fertile nuclide. The latter leads to a deterioration in neutron balance, to reduction of the reactivity and, as a consequence, to decrease of fuel burn-up. In order to change this situation efficiently a fissile nuclide should be converted into another fissile nuclide in case of neutron capture reaction. Such a pair of fissile nuclides represents a ^{232}U-^{233}U system, for example. To use these nuclides, fuel should contain sufficient quantities of ^{232}U itself or its predecessor ^{231}Pa. The following chain is accomplished in such a fuel: $^{231}\text{Pa}\rightarrow{}^{232}\text{U}\rightarrow{}^{233}\text{U}$. Here, the first nuclide plays a role of an effective burning absorber, the second – a fissile nuclide with moderate neutron multiplying properties, and the third – a fissile nuclide with excellent neutron multiplying properties. Consecutive transmutations of these nuclides allow to improve neutron-multiplying properties of fuel under irradiation. Worthwhile mentioning that these nuclides do not exist in nature. They should be produced, for example, in thermonuclear reactors [6].

4. ADVANTAGES OF HIGH FUEL BURN-UP

Time dependence of reactivity for light water reactor lattice of the VVER-type reactor, loaded with various fuel types, is shown in Fig.1. It can be seen that, in the case of traditional uranium dioxide fuel enriched up to 4.4% ^{235}U the neutron multiplication factor decreases monotonously from K_∞=1.38 to K_∞=1 till fuel burn-up of 4.2% HM (curve 1). Practically the same result can be attained for nitride ^{232}Th-^{233}U fuel containing 5.3% ^{233}U (curve 2). The situation changes drastically when ^{231}Pa being the initial nuclide for the ^{232}U-^{233}U couple, is introduced into thorium-based nitride fuel. Four fuel compositions, which differ from each other by content of ^{231}Pa, were analyzed.

The fraction of ^{232}Th is constant and equal to 69% (curves 3-6). ^{231}Pa and ^{233}U account for the remaining 31% of fuel. It can be seen from Fig.1 that high fuel burn-up (to 30% HM) may be achieved without ^{231}Pa at all (curve 3), but it can only be achieved with an extremely high value of initial fuel enrichment (31% ^{233}U). Evidently, this case is unlikely to be acceptable from the reactor safety standpoint because of the high reactivity margin of fresh fuel (K_∞=1.91). Substitution of 5% ^{233}U with 5%^{231}Pa (curve 4) reduces initial reactivity margin (K_∞=1.61) for the same value of fuel burn-up. Substitution of 10% ^{233}U with 10%^{231}Pa (curve 5) reduces initial neutron multiplication factor to K_∞=1.37, i.e. practically the same initial magnitude of K_∞ as that for traditional uranium dioxide fuel. If fractions of ^{233}U and ^{231}Pa are

approximately identical (curve 6), then initial multiplication factor is equal to a moderate value (K_∞=1.10), needed to compensate neutron leakage and for reactor control under operating conditions. It should be noted that neutron multiplying properties of such a fuel are stabilized until fuel burn-up ~17% and then they smoothly reduce to K_∞=1.0 when fuel is burnt-up to 30% HM. Such a fuel composition advantageously differs from previous compositions by a smooth variation of reactivity along with fuel burn-up. These cases demonstrate the stabilization of neutron multiplying properties in the process of fuel burn-up due to the increasing role of isotope transitions in the $^{231}Pa \rightarrow ^{232}U \rightarrow ^{233}U$ chain.

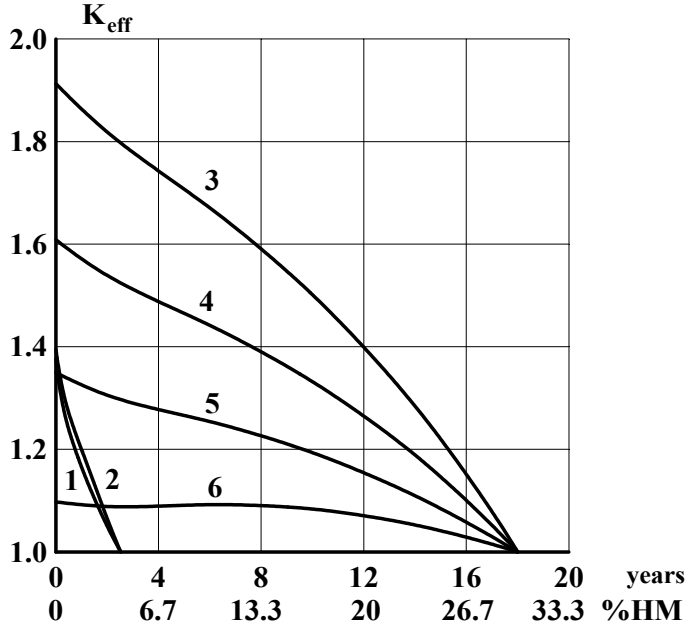

Figure 1. Dependence of multiplying properties on irradiation time (fuel burn-up) for different fuel types:

1 – (4.4%^{235}U+95.6%^{238}U) O_2, 2 – (5.33%^{233}U+94.67%^{232}Th) N,
3 – (0%^{231}Pa+31%^{233}U+69%^{232}Th) N, 4 – (5%^{231}Pa+26%^{233}U+69%^{232}Th) N,
5 – (10%^{231}Pa+21%^{233}U+69%^{232}Th) N, 6 – (15%^{231}Pa+16%^{233}U+69%^{232}Th) N,

The following benefits may be derived from using fuel with stabilized neutron multiplying properties up to high fuel burn-up. First of all, it drastically decreases the technological procedures related to fuel fabrication, fuel transportation and refueling (per 1 MW*day energy produced). So, for example, the number of procedures mentioned above will be less by a factor of 5-7, if traditional fuel of light water reactor with burn-up of 4-6% HM is changed to fuel with a burn-up of 30% HM. In addition to economical benefits, a reduction in the number of fuel reloads during reactor life-time will result in a substantial reduction of potential possibility of the switching fissile material from civilian usage to military purposes. It enhances proliferation resistance of the nuclear fuel cycle. If, in addition, power density in the reactor core is reduced by a factor of 2-3, then the reactor with high fuel burn-up is able to operate a full reactor life-time (40-50 years) without refueling. Such reactors may be supplied to developing countries as «black boxes» with minimal danger of nuclear proliferation, because all operations with nuclear fuel may be concentrated just at the fuel regeneration

plants. Also, such reactors will be simpler in manufacture and operation because they are equipped with tools for operative reactivity control only. The equipment needed for fuel reloading, cooling and transportation will not be installed at nuclear power plant.

Another factor of improvement of the fuel proliferation protection is a build-up of ^{232}U in fuel under irradiation, which can prevent nuclear fuel from switching to military purposes. This is explained by increased and long-term heat generation due to α-decay of ^{232}U (830 W/kg, $T_{1/2}$=68.9 years). Calculations show that spent fuel with an initial composition of 15% ^{231}Pa + 16% ^{233}U + 69% ^{232}Th with the good stabilization of reactivity over high burn-up (curve 6), contains 17.4% ^{232}U in a uranium fraction at the end of fuel residence time. It means that the specific decay heat generation of uranium, containing the proliferation-attractive fissile nuclide ^{233}U, is at a level of 145 W/kg, i.e. more by a factor of 14 than that of reactor-grade plutonium (10.5 W/kg) and by a factor of 63 than that of weapons-grade plutonium (2.3 W/kg) [7]. Such a level of decay heat generation in uranium makes it substantially complicated to apply uranium for military purposes. Certainly, at the beginning of fuel irradiation this parameter will be lower, and heat generation in fresh fuel is insignificant. To upgrade proliferation resistance, the introduction of a small ^{232}U amount into fresh fuel may be foreseen. Under fuel recycling conditions, a sufficient ^{232}U content in reloaded fuel will be provided in a natural way.

In the case of a closed nuclear fuel cycle, the use of fuel with high burn-up also leads to a substantial reduction in chemical reprocessing activities. So, for example, with fuel burn-up in light water reactor at a level of 5% HM, about 95% of non-burnt fuel is directed to chemical reprocessing, i.e. a 20-fold recycle is required to burn-up of fuel load. If fuel burn-up will be at a level of 30% HM, then only a 3.3-fold recycle is required, i.e. demands for radiochemical reprocessing activity can be reduced by a factor of about 6.

5. TECHNICAL FEASIBILITY OF HIGH FUEL BURN-UP

A question arises: how feasible is the regime of high fuel burn-up? As mentioned above, it was developed vibro-packed oxide fuel elements irradiated in fast neutron spectrum up to high burn-up as much as 32% HM without failure [2]. In present study it was assumed that fuel applied is a porous nitride fuel with reduced density: 9.8 g/cm^3, i.e. 31% lower than the density of uranium nitride fuel and 18% lower than the density of thorium nitride. There are no published data on the properties of protactinium nitride. Also, there is no information on the swelling of mixed nitride fuel (Th,Pa,U)N. However, it is known than uranium mono-nitride is characterized by a substantially lower value of swelling than that of uranium dioxide, owing to better structural integrity [8]. In addition, stainless steel is supposed to be used as a structure material (not zirconium-based alloys commonly used in thermal reactors). Stainless steels were and continue to be used as a material for fuel cladding in fast reactors [2]. Such a substitution of the structure materials appeared possible owing to a better neutron balance of the new fuel type and it was applied to protect nitride fuel against contact with water.

The proposed fuel contains a substantial fraction of ^{231}Pa. Under neutron irradiation, some part of ^{231}Pa transforms to ^{232}U, the initial nuclide for the chain of radioactive decays, culminating in stable ^{208}Pb. This chain includes six α-decays and result in helium accumulation in fuel. If the helium amount is a substantial fraction of the amount of gaseous fission products, the problem of fuel cladding integrity will be more complicated. It was evaluated the ratio of helium nuclei, generated in α-decays to a total amount of gaseous fission products generated. If fresh fuel contains 15% ^{231}Pa, the ratio is 25% (fuel burn-up - 30% HM). It can be concluded that the protection of the fuel cladding integrity will require proper selection of the fuel porosity.

During reactor operation the structure materials are exposed to neutron irradiation, which can damage their crystal lattice, thus deteriorating strength properties and, finally, destroying the materials. The value of damage dose, or the number of displacements per one atom (DPA) of structure material for irradiation time was evaluated by the following equation:

$$DPA \approx \left[\sigma_{dpa}(0)\cdot\Phi(0)+\sigma_{dpa}(T)\cdot\Phi(T)\right]\cdot\left(T/2\right),$$

where: σ_{dpa} – displacement cross-section averaged over neutron spectrum; Φ – total neutron flux. The results presented in Table 1 show that even under high fuel burn-up, the dose rate changes moderately. So, it was assumed that this equation may be applied to evaluate the damage dose to an acceptable degree of accuracy using averaged displacement cross-section and integral neutron flux at the beginning and at the end of the fuel residence time as input data.

Table 1. Key neutronics parameters for damage dose accumulation in stainless steel (fuel cladding material in VVER-type reactor analyzed)

Fuel type			Mono-nitride (^{231}Pa,^{233}U,^{232}Th)N		Uranium dioxide
Fresh fuel composition, %	^{232}Th ^{231}Pa ^{233}U		94.67 0 5.33	69 15 16	4.4% ^{235}U 95.6% ^{238}U
Displacement cross-section σ_{dpa}, barn		t=0 t=T	261 252	318 283	241 252
Total neutron flux Φ, 10^{+14} n/(cm^2·s)		t=0 t=T	3.05 4.39	3.32 4.18	3.20 4.81
DPA (T)			7.56	63.5	7.87
DPA per 1% HM fuel burn-up		t=0 t=T	1.54 2.14	2.05 2.30	1.53 2.42
Fission cross-section, barn	^{233}U	t=0 t=T	31.3 38.2	9.1 22.2	33.4(^{235}U) 28.5(^{235}U)
	^{232}U	t=T	-	9.6	64.0(^{239}Pu)
Neutron fluence, 10^{+23} n/cm^2 (E_n > 0.1MeV)			0.15	1.27	0.16

It can be seen from Table 1 that displacement cross-sections and absolute values of neutron flux do not undergo substantial changes for all the fuel types and compositions analyzed. This is a result of compensating effects related to the fact that in process of fuel burn-up the

number of heavy atoms decreases because of transmutation into fission products. As a consequence neutron spectrum softens and the fission cross-section of ^{233}U substantially increases. So, even in the case of a reduced content of fissile nuclide ^{233}U, neutron flux does not increase significantly, and the damage dose rate (per 1% HM of the fuel burn-up) is appeared to be comparable with that for traditional oxide fuel. So, the dependence of DPA on irradiation time is mainly defined by the time of irradiation. In our calculations, the residence time for traditional uranium oxide fuel was assumed to be 2.5 years. The fuel residence time of high burn-up nitride fuel was shown to be 18 years. Then, DPA of structure material for high burn-up nitride fuel should be proportional to the time of irradiation: 63.5 dpa for a time range 18 years. Experimental data now confirm the fact that maximal DPA, up until a drastic loss of strength properties for some types of stainless steels applicable as a structure materials for nuclear reactors, can achieve a magnitude of about 180 dpa [10], so fuel elements may be used without re-fabrication of fuel cladding. According to Ref. [11], stainless steel cladding is able to withstand neutron fluence up to $4 \cdot 10^{+23}$ n/cm^2 (E_n>0.1 MeV). The evaluations show that the fluence of high-energy neutrons in the case of fuel burn-up (30% HM), does not exceed this magnitude (see Table 1). In case of necessity, it can be applied technology like the DUPIC-process to re-fabricate cladding of fuel elements and to prolong fuel burning-up.

6. RE-FABRICATION OF FUEL ELEMENTS BY MEANS OF THE DUPIC-TYPE TECHNOLOGY

If excellent neutron multiplying properties are maintained at a sufficient level for fuel burn-ups exceeding the limits defined by the capacity of fuel elements to work then, actually, there is no need to reprocess fuel itself. It is required to renew fuel elements only. In other words, it is required to insert the same fuel into a new fuel cladding. Thus, the reactor operation may be prolonged. As is known, a non-chemical technology, DUPIC-process has already been developed, and now is being tested [9], as applied to spent oxide fuel of a light water reactor (PWR-type) for further burn-up in CANDU-type heavy water reactors.

The DUPIC-technology implies thermal and mechanical procedures only. In the process of oxide fuel milling and reduction-oxidation cycling the fuel is loosened, gaseous, volatile and semi-volatile fission products escape the fuel. Thereafter, the fuel powder is used again for re-fabrication of fuel elements and fuel assemblies to prolong their irradiation in the reactor.

The application of this technology (perhaps several times) could provide the prolongation of fuel irradiation in the same reactor and would achieve high fuel burn-up. All these operations might be accomplished directly at the nuclear power plant site, thus excluding fuel transportation and reducing the demands for radiochemical reprocessing. These circumstances and the fact that only thermal and mechanical operations are involved in the DUPIC processes by which it is impossible to separate the main mass of fission products from actinides and to separate actinides from each other, do enhance the proliferation resistance of such a fuel cycle. However, it should be noted that, till now, the DUPIC-technology has been developed for oxide fuel only, and there is no any information yet on its applicability for reprocessing other types of ceramic fuel (carbide, nitride). This stipulates the need to conduct neutronics

analysis of light water fuel lattices for mixed oxide (^{232}Th, ^{231}Pa, ^{233}U) O_2 as well. It will be performed at a later stage.

7. PROLONGATION OF THE FUEL RESIDENCE TIME IN REACTOR UP TO 50 YEARS

The results discussed above allow to discuss the possibility of creation of a light water reactor «black box»-type, i.e. reactor with a fuel residence time equal to the reactor life-time, i.e. about 50 years (see Table 2). Here, appropriate parameters of a traditional VVER-type reactor (uranium oxide fuel enriched up to 4.4% ^{235}U) are also presented as a reference option for comparison.

Table 2. Characteristics of traditional VVER-type and «black box»-type reactors

Characteristics	Reference option: VVER-reactor, fuel - UO_2	«Black box» reactor, (Th, Pa, U)-mono-nitride
Fresh fuel composition	4.4% ^{235}U	69%^{232}Th+15%^{231}Pa+16%^{233}U
Fuel density, g/cm^3	10.3	9.8
Fuel burn-up, % HM	4.2	30
Power density, MW/m^3	113	41
Fuel residence time, years	2.5*	50
DPA(T), (stainless steel)	7.9	63.5

* - time range for fuel reloading was not taken into account

It can be seen that a fuel residence time of 50 years can be achieved at fuel burn-up of 30% HM due to reduction of power density in the reactor core by a factor of 2.75. It is worth-while mentioning, an approach to reduce a power density in reactor core is applied to develop of upgraded reactor safety concepts [1, 11]. The calculations carried out for reduced power density showed that time behavior of the core reactivity only «stretches» in time, compared with the standard heat generation level.

The long fuel residence time of the fuel containing ^{232}U leads to significant helium accumulation in the fuel because of α-decays of ^{232}U and its daughter nuclides. Evaluations showed that for a 50-year fuel residence time the amount of gaseous substances (fission products and helium) increased by a factor of 1.87 for the fresh fuel contained 15% ^{231}Pa. It appears that this problem might be resolved by means of the DUPIC-type technology periodically removing gaseous substances from fuel and continuing the reactor operation. Needless to say, there is a possibility to relief gaseous products from fuel elements into primary coolant circuit.

CONCLUSIONS

The concept of light water reactor core fuelled with ^{232}Th-^{231}Pa-^{233}U ceramics was proposed. It is shown that such a reactor concept has a potential to achieve high (30% HM) fuel burn-up that may be restricted by the integrity of fuel elements. The application of the DUPIC-type technology is foreseen to achieve the high fuel burn-up. The neutron multiplying properties of

fuel lattice and safety parameters assessed have favorable character over high fuel burn-up. Certain reduction of power density in the reactor core would allow to achieve a fuel residence time of up to 50 years for fuel burn-up of about 30% HM.

In conclusion it is necessary to note that revision of ^{232}U nuclear cross sections and those of insufficiently known ^{231}Pa should be done as they differ significantly between the US library ENDF/B-IV and Japanese library JENDL-3.2.

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