ADS FUEL CYCLE SUPPLEMENTED WITH DUPIC PROCESSES: ACHIEVEMENT OF HIGH FUEL BURN-UP

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ABSTRACT

The application of thermal-mechanical DUPIC-technology in the ADS fuel cycle to achieve high fuel burn-up has been studied. As it is known, one of the main ADS advantages is the improved safety caused by the sub-critical operation mode. The sub-critical operation mode also facilitates the possible use of the blanket design characterized by unfavorable reactivity effects and, hence, cannot be operated in a critical mode for safety reasons. Slightly sub-critical fast neutron ADS blanket loaded with oxide fuel containing minor actinides can be considered to be a power generating facility that is able to achieve ultra-high fuel burn-up if the DUPIC-type technology is applied repeatedly for thermal-mechanical regeneration of irradiated fuel. The DUPIC-type technology has potential to be extended for mono-nitride fuel regeneration. It is shown that high fuel burn-up at a level of 30-40% HM can be achieved in a slightly sub-critical fast ADS blanket, cooled by heavy liquid metal.

1. INTRODUCTION

In the 1990s the concept of a BREST fast reactor with heavy metal (lead) coolant was developed. As stated by the developers [1,2], a distinctive feature of the concept is the improved operational safety that excludes severe accidents at a deterministic level. It is achieved by the fact that all reactivity effects related to variation of the working reactor conditions are both small in value and favorable. Under any anticipated conditions reactivity effects cannot cause a prompt neutron criticality, with the possible exception of terrorist acts, which should be excluded through the proper organizational procedures. Duration of the fuel residence time and refueling scheme were chosen in such a way that appropriate reactivity changes during the fuel burn-up would not exceed β_{eff} – an effective fraction of delayed neutrons.

It is generally accepted that a sufficient sub-criticality level corresponds to the value of effective neutron multiplication factor K_{eff}, equal to about 0.95. However, after development of the BREST reactor concept, the essence of the sub-critical operation mode does not depreciate at all but, on the contrary, now opens new possibilities. Indeed, just the conceptual

development of the BREST reactor (critical system) with deterministic exclusion of severe accidents evidently means that the improved safety aspect is especially more appropriate for the sub-critical ADS blanket with K_{eff} sufficiently close to unity. This fact, in its turn, creates the possibility to attain substantially higher fuel burn-up as compared with that of the BREST reactor fuel. Then the advantages related to the reduction of the fuel cycle scope could be attained.

2. FEATURES OF THE BREST FAST REACTOR CONCEPT

According to the BREST reactor concept, the temperature effects of reactivity for fuel and coolant are negative and small in value (lower than the value of β_{eff}). The reactivity swing in the process of the fuel burn-up does not exceed β_{eff} because the core breeding ratio is close to unity (slightly more than 1). Actually, it is attained owing to the application of high-density fuel, namely, mixed uranium-plutonium mono-nitride (U,Pu)N, and by limitation of the fuel burn-up (7-9% HM) [3]. Under reduced power density (~150 MW/m^3) the fuel residence time lasts 5 years with partial refueling once a year. To enhance proliferation resistance of fissile nuclear materials, the concept implies application of the electrochemical fuel reprocessing facility *in situ* [4].

So, the need to maintain stable neutron multiplying properties of a critical system over fuel burn-up has enforced developers of the BREST fast reactor concept to reject conventionally used oxide fuel and select frequent electrochemical fuel reprocessing *in situ*.
As it is known, the impact of fission products on the balance of the chain fission reaction in fast neutron spectrum is rather small. So, it is possible, in principle, to achieve high fuel burn-up in fast reactor cores. However, the requirement to provide safety of the critical system (the BREST reactor) with the exclusion of severe accidents at deterministic level allowed the developers to use neither this advantage nor other advantages inherent to such a kind of fast neutron systems.

3. POTENTIAL ADVANTAGES OF SUB-CRITICAL FAST NEUTRON BLANKET COOLED BY LIQUID HEAVY METAL

Conversion of the reactor core to sub-critical operation mode supported with neutrons emitted from the target bombarded by accelerated charged particles can preserve operational safety at a deterministic level and, in addition, can attain additional important advantages, as listed below:
1. Fuel burn-up can be essentially increased with the appropriate reduction of the scope of radio-chemical reprocessing. Thus, the potential danger related to uncontrolled proliferation of fissile materials can be decreased.
2. The fuel composition can be expanded by involving minor actinides, which allow the stabilization of neutron multiplying properties under high fuel burn-up [5].
3. The introduction of minor actinides (^{237}Np, ^{241}Am) into the fuel composition leads to a rapid increase of ^{238}Pu build-up. The presence of ^{238}Pu in plutonium isotope composition may

be considered to be an effective barrier against uncontrolled proliferation of nuclear materials [6].

4. Oxide fuel, applied widely in nuclear power and well compatible with many construction materials and coolants, could be used. For spent oxide fuel regeneration, so-called DUPIC technology [7] has been developed. This technology includes thermal & mechanical operations only and provides an essential increase of fuel burn-up in fast neutron spectrum, without radiochemical treatment.

5. Usage of heavy metal coolant, both in ADS blanket and as a material of ADS target, allows the exclusion of a stationary wall separating blanket and target regions. This is an important advantage because this stationary wall should be operated under hard radiation and temperature conditions.

4. THERMAL-MECHANICAL DUPIC TECHNOLOGY FOR OXIDE FUEL REGENERATION

Initially, DUPIC technology was developed for further burn-up of spent fuel discharged from light-water PWR-type power reactors in CANDU-type heavy-water power reactors. As known [7], spent fuel of PWRs contains 1.5-2.5% fissile nuclides (non-burnt ^{235}U and produced plutonium). Therefore, spent fuel of PWRs may be used in CANDU-type heavy-water reactors without additional enrichment. All that is needed is to arrange re-fabrication of oxide fuel with the subsequent manufacture of fuel elements and fuel assemblies for CANDUs. DUPIC technology implies only thermal and mechanical procedures. Spent fuel assemblies of PWRs are disassembled, and fuel elements are extracted and cut into sections to gain access to spent oxide fuel. Then, cycles of oxidation-reduction reactions are carried out for removal of fuel claddings, crashing of spent fuel and the release of gaseous and volatile fission products.

Oxidation takes place in air atmosphere at a temperature of about 450^0C. In this process, uranium dioxide transforms into uranium octa-oxide U_3O_8 with density decreasing by 30%. Reduction is carried out in an argon-hydrogen atmosphere ($Ar+4\%H_2$) at about 700^0C. In this operation, uranium octa-oxide U_3O_8 converts back into uranium dioxide. After multiple oxidation-reduction cycles, spent fuel is loosened and transformed into fine powder. Gaseous (xenon, krypton) and volatile (cesium, ruthenium) fission products escape the fuel. To produce sintered fuel with increased density later on, oxidation-reduction cycles are accompanied by procedures of fuel crushing in special mechanical mills. Thereafter, fuel powder is pressed and sintered into fuel pellets, which are then used to manufacture fuel elements and fuel assemblies for CANDU-type reactors.

So, DUPIC regeneration technology includes neither operations, nor equipment units for separation of uranium, plutonium and the major mass of fission products. Therefore, this technology may be considered to be a proliferation-resistant technology. In essence, application of this thermo-mechanical technology provides for the removal of gaseous and volatile fission products from spent fuel, the renewal of fuel element claddings and the manufacture of new fuel assemblies for the next stage in fuel irradiation. If regenerated fuel contains a sufficient amount of fissile material, the next irradiation step may be performed in

the same reactor from where spent fuel was taken. In the case under investigation, this means the next fuel irradiation step is performed in the ADS blanket.

5. MATHEMATICAL MODEL

Variation of neutron multiplying properties of the ADS blanket under high fuel burn-up with intermediate fuel regeneration by means of DUPIC technology was investigated on a cylindrical cell model with application of the computer code GETERA [8]. Neutron leakage from the ADS blanket was taken into account by the introduction of buckling values. Some parameters of the ADS blanket cell were taken from the publication [3] where design features of the BREST fast reactor are presented:

- Diameter of fuel element - 10.2mm;
- Thickness of steel cladding - 0.5mm;
- Stainless Steel - EP-823 Mark;
- Fuel - (^{238}U+^{239}Pu+^{237}Np)O_2 and (^{238}U+^{239}Pu+^{237}Np)N;
- Coolant – lead;
- Buckling - B^2=0.0005cm^{-2}.

The buckling value chosen for the calculations corresponds to axial neutron leakage into axial blanket regions of the BREST reactor (height of the reactor core - 110cm). Reduced fuel density was chosen to compensate the fuel swelling by solid fission products. Since gaseous and volatile fission products are removed by the DUPIC technology, the fuel swelling caused by solid fission products leads to a filling up of the porosity when the fuel burn-up of 35-40% HM [9] is achieved.

An achievement of ultra-high fuel burn-up (35-40% HM) will make an attractive concept of the Internationally Monitored Retrievable Storage System [10,11]. According to this concept, under international control a fresh MOX fuel can be manufactured (and spent fuel can be reprocessed after returning it back from NPP).

It was assumed that fresh MOX-fuel contains ^{239}Pu as a main fissile nuclide. Actually, it corresponds to the problem of weapons-grade Pu burn-up. To stabilize neutron multiplying properties during the fuel burn-up, 0-10% $^{237}NpO_2$ can be added to the fuel composition. A previous study [5] has shown that the admixture of ^{237}Np in fuel initiates the following reaction chain: ^{237}Np(n,γ)^{238}Pu(n,γ)^{239}Pu. Here, the first nuclide plays the role of burning absorber, the second is a fissile nuclide with moderate neutron multiplying properties, and the third is a fissile nuclide with excellent neutron multiplying properties. Thus, the reaction chain consequently improves neutron multiplying properties in the process of the fuel burn-up. This fact compensates the reactivity decrease caused by the accumulation of fission products, stabilizes neutron multiplying properties and, thus, facilitates an increase in fuel burn-up. It was assumed in calculations that the duration of the fuel irradiation cycle corresponds to the fuel burn-up of no more than 10% HM. Afterwards, the fuel is withdrawn from the ADS blanket and regenerated with the DUPIC technology. Then, fuel elements and fuel assemblies are re-fabricated. A new pitch of the fuel elements lattice is chosen in such a way to provide 10%HM fuel burn-up in the next irradiation cycle. Proper selection of the

pitch value effectively controls the initial neutron multiplication factor K_{multi} within the range $0.95<K_{multi}<1$. Here, K_{multi} represents the infinite multiplication factor K_{∞}, calculated under the assumption that neutron leakage from the ADS blanket is taken into account by using the value of buckling B^2 [8].

Two fuel irradiation regimes were analyzed. The first regime implied simultaneous and complete discharge and regeneration of fuel loaded in the ADS blanket. So, the initial value of K_{multi} was chosen from the range $0.95<K_{multi}<1$ and K_{multi} should not exceed unity, even if coolant density is decreased. The second regime implied continuous refueling of the ADS blanket where fuel assemblies with low and high fuel burn-up were present simultaneously. Such an irradiation regime allows the use of excess neutrons generated by the assemblies with low fuel burn-up for prolonged irradiation of the assemblies with high fuel burn-up. In this case the K_{multi} value of the fuel element lattice with high fuel burn-up may be significantly lower than 0.95, as fed by neutrons generated in high-reactivity fuel assemblies. Nevertheless the K_{multi} value averaged over the volume of the ADS blanket could be maintained at a level of 0.95. Then, the maximum value of the fuel burn-up (S_{max}) for such an irradiation regime can be determined from the following equation of integrated reactivity balance:

$$\int_0^{S_{max}} \left[K_{multi}(s) - 0.95\right] \cdot ds \quad = \quad \int_0^{S_{max}} \left[0.95 - K_{multi}(s)\right] \cdot ds$$

(for fuel burn-ups with $K_{multi}(S) \geq 0.95$) (for fuel burn-ups with $K_{multi}(S) \leq 0.95$)

6. RESULTS OF CALCULATIONS

Variations of neutron multiplying properties in the process of high fuel burn-up and multiple application of the DUPIC technology for regeneration of fuel elements were calculated for dioxide fuel composition ($^{238}U+^{239}Pu+0\div10\%^{237}Np)O_2$ and for mono-nitride fuel composition ($^{238}U+^{239}Pu+0\div10\%^{237}Np)N$. Neptunium-free fuel and fuel containing up to 10% Np were analyzed. As known [5], a 10%-addition of neptunium in fuel makes it possible to stabilize neutron multiplying properties in the irradiation process and, thus, to increase fuel burn-up. After each 10% HM of the fuel burn-up (or more frequently, if neutron multiplying properties of the fuel element lattice degrade quickly) the DUPIC technology is applied for fuel regeneration and a new value for pitch-to-diameter ratio is chosen for the fuel element lattice, to maintain K_{multi} within the range $0.95<K_{multi}<1$.

Variations of neutron multiplication factor K_{multi} for the ADS blanket loaded with neptunium-free dioxide fuel are presented in Fig.1a for the cyclic irradiation regime with simultaneous discharge and regeneration of all the fuel assemblies. It can be seen that, at the initial irradiation stage of the ADS blanket with a rather wide fuel element lattice (pitch-to-diameter ratio p/d=1.51), reactivity reduces and achieves $K_{multi}=0.95$ at fuel burn-up of ~11% HM. Then, the fuel is re-fabricated with application of the DUPIC technology, new fuel elements are manufactured and sub-assemblies (with a new pitch-to-diameter ratio p/d=1.40 fuel element lattice) are assembled. These operations allow the upgrading of neutron multiplying properties. In recycles with fuel regeneration pitch-to-diameter ratio p/d consequently

decreases. Cyclic reloading of the entire ADS blanket requires that $K_{multi}(S)$ (S – a variable characterizing current burn-up) cannot be lower than 0.95. So, when the least acceptable value of pitch-to-diameter ratio p/d=1.17 is reached the corresponding value of the fuel burn-up is equal to ~27% HM.

In a continuous refueling regime it is assumed that, as a rule, those fuel assemblies which reach the next 10% HM of fuel burn-up, are discharged from the ADS blanket and regenerated by DUPIC processes. In this case, neutron multiplication factor K_{multi}, averaged over the ADS blanket volume, can be kept constant (K_{multi}=0.95) up to a highest value of fuel burn-up of ~25% HM (see Fig.1b). Thereafter, under the adopted assumptions, spent fuel should be reprocessed by application of radiochemical technologies with the extraction of accumulated fission products.

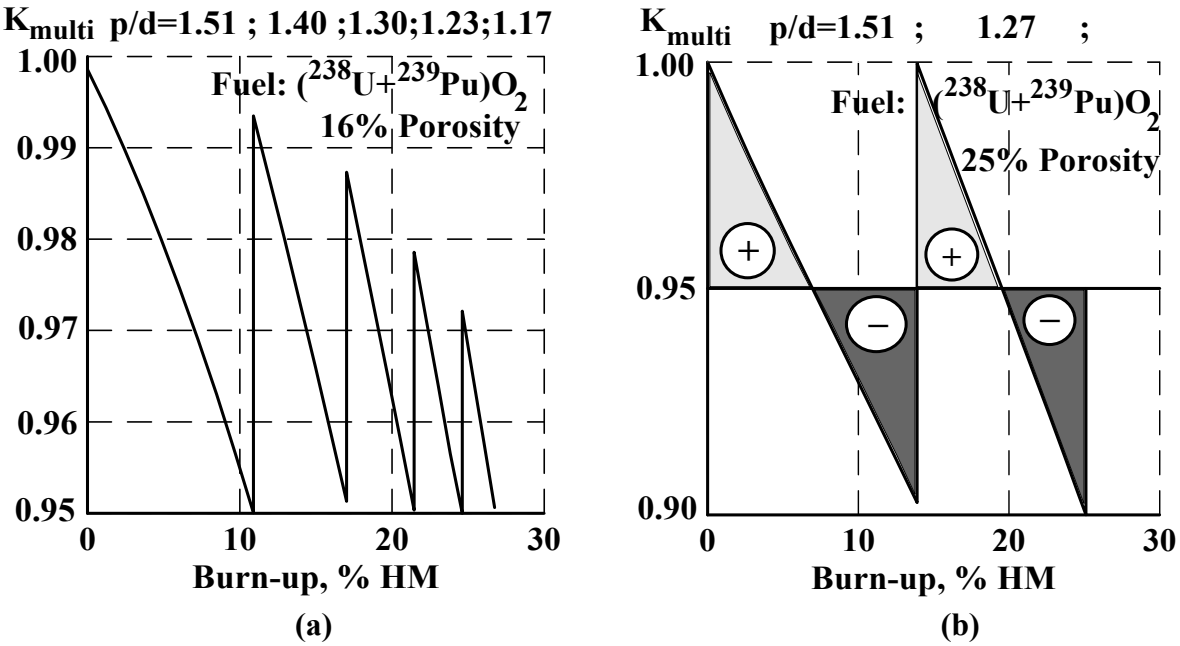

Figure 1. Dependence of neutron multiplication factor K_{multi} on fuel burn-up and applications of the DUPIC technology for neptunium-free dioxide fuel:
a) cyclic irradiation regime; b) continuous refueling regime

An addition of 10% ^{237}Np to the fuel on conditions of continuous refueling helps to achieve a maximum value of the fuel burn-up of 37% HM (see Fig.2a). The oxide fuel burn-up achieved is much more than that of the BREST reactor project. This can be accomplished due to stabilization of neutron multiplying properties over fuel burn-up due to ^{237}Np addition [5].

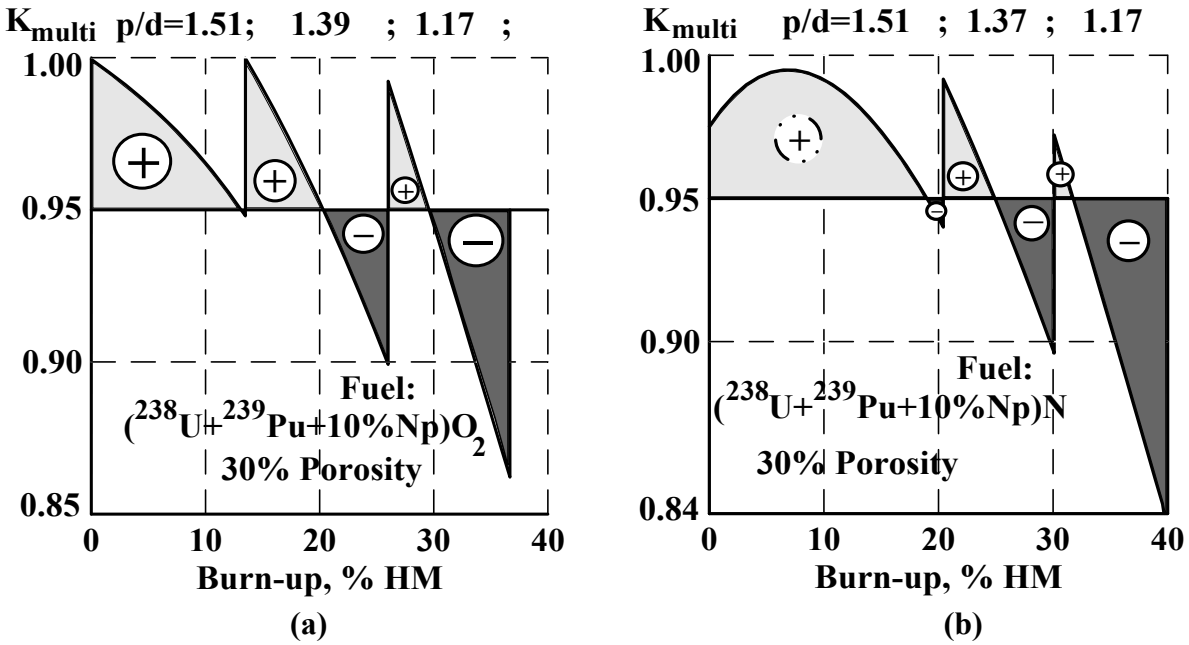

Figure 2. Dependence of neutron multiplication factor K_{multi} on fuel burn-up and applications of the DUPIC technology for fuel with 10% ^{237}Np on conditions of continuous refueling: a) dioxide fuel; b) nitride fuel

In the process of high fuel burn-up the content of ^{237}Np substantially decreases from 10% in fresh fuel to 0.5% at a fuel burn-up of 37% HM (see Table). So, after radiochemical reprocessing, ^{237}Np content should be made up again to the initial level of 10%, in order to provide a stabilizing effect on neutron multiplying properties of the fuel.

Table. Content of ^{237}Np in dioxide fuel and ^{238}Pu in plutonium (continuous refueling regime)

Fuel burn-up (% HM)	0	10	20	30	40
^{237}Np content (%)	10	5.1	2.52	1.15	0.47
^{238}Pu / Pu (%)	0	17.9	17.2	13.8	8.2
Heat generation in 10-kg plutonium charge (W)	25	1050	1010	815	500

Generation of ^{238}Pu in this chain leads to the fact that fissile plutonium contained in the fuel obtains an important barrier against uncontrolled proliferation. This barrier is created by an intense emission of spontaneous fission neutron emanation and by intense heat generation due to α-decay [12]. Data on variation of ^{238}Pu content in plutonium in the process of fuel burn-up are presented in the above Table. The heat generation in a 10-kg plutonium charge made of plutonium accumulated in the ADS fuel was also assessed. When fuel irradiation begins, the value of heat generation is close to that of weapons-grade plutonium [13]. However, in the process of fuel irradiation the decay heat generation increases by a factor of 20-40, a fact that can substantially affect the ability to transfer such plutonium to military purposes [12].

As it was mentioned above, when selecting the initial value of K_{multi} for each subsequent irradiation cycle it was taken into account that the reduction of coolant density could not lead

to the K_{multi} value exceeding unity. Since nuclides ^{237}Np, ^{238}Pu and fission products are present in the fuel composition the coolant effect of reactivity is positive and large in magnitude. Maximum values of this reactivity effect are as high as +0.0136 in the first irradiation cycle and +0.0275 in the last. Such positive reactivity effects are not acceptable for the BREST reactor project. However, the sub-critical ADS blanket under investigation is not able to achieve a critical state, even with an infinite lattice of fuel assemblies loaded. It is expected that if ^{237}Np content in fresh fuel exceeds 10% the attainable value of the fuel burn-up could be more than 40% HM. Also, a conservative requirement that neutron multiplication factor K_{multi} should not exceed unity can be avoided in detailed calculations of sub-critical blanket structure.

As known, a «typical» fuel cycle with the application of DUPIC technology implies some addition of low-enriched uranium prior to the fuel re-fabrication for CANDU reactors. The admixture of various materials into ADS fuel in the process of its regeneration and recycling might be considered as an additional factor, which, under conditions of reasonable use, could result in a further increase of fuel burn-up.

In order to further reduce demands for radiochemical reprocessing it seems expedient to use more dense fuel (for example, mono-nitride fuel) instead of traditional oxide fuel. Improvement of neutron multiplying properties creates the chance to increase fuel burn-up. The calculations of neutron multiplication factor K_{multi} for continuous reloading regime of mono-nitride ($^{238}U+^{239}Pu+10\%^{237}Np$)N fuel with 30%-porosity are presented in Fig.2b. It can be seen that the average value of K_{multi}, equal to 0.95, may be maintained up to maximum fuel burn-up of ~41% HM. Nitride fuel with 30%-porosity allows the accumulation of solid fission products at a level corresponding with fuel burn-up of 50-60% HM, keeping in mind that thermal-mechanical DUPIC-type technology is applied for fuel regeneration with the release of gaseous, volatile and semi-volatile fission products.

Similar results are obtained if oxide fuel (porosity - 25%) is used first and, after achieving fuel burn-up of ~28% HM, the DUPIC technology is applied, complemented with a new technological operation for conversion of the oxide fuel into mono-nitride fuel with 30%-porosity (see Fig.3). However, the DUPIC technology should be further extended for regeneration of mono-nitride fuel. There are certain physical and chemical prerequisites for such a development, since it is known that mono-nitride fuel (mono-carbide fuel, carbide-nitride and oxide-carbide-nitride fuels, as well) can be produced from oxide fuel by means of dry thermal-mechanical procedures [4,9].

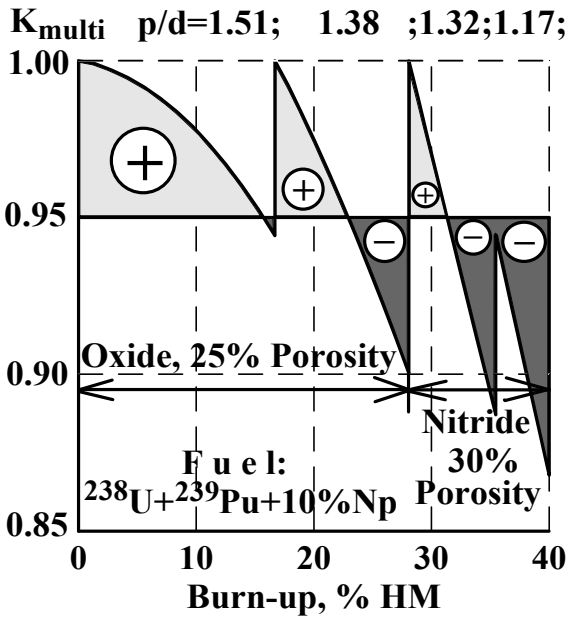

Figure 3. Dependence of neutron multiplication factor K_{multi} on fuel burn-up and applications of the DUPIC technology for dioxide and nitride fuel with 10% ^{237}Np (continuous refueling regime)

As the figures show, to maintain multiplying properties up to a high value of fuel burn-up, pitch-to-diameter ratio should be decreased using a small step 3-5% HM. In this case, just simplified DUPIC technology concerned with operation of pitch-to-diameter ratio variation may suffice.

CONCLUSIONS

Slightly sub-critical fast neutron ADS blanket loaded with oxide fuel containing minor actinides can be considered to be a power generating facility that is able to achieve ultra-high fuel burn-up if the DUPIC-type technology is applied repeatedly for thermal-mechanical regeneration of irradiated fuel. The DUPIC-type technology has potential to be extended for mono-nitride fuel regeneration.

Achievement of high fuel burn-up (35-40% HM) will make an attractive concept for the Internationally Monitored Retrievable Storage System due to a drastic decrease in the spent fuel flow rate. Under international control a fresh MOX fuel can be manufactured and spent fuel can be reprocessed after returning it back from NPP.

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