

## **INFLUENCE OF INTERMEDIATE CHEMICAL REPROCESSING ON FUEL LIFETIME AND BURN-UP**

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### **Abstract**

The influence of intermediate chemical processing of nuclear fuel with removal of fission products on the fuel burn-up and lifetime for heavy water CANDU type reactors operating with fuel on base of natural uranium is studied in this paper. Two types of nuclear fuel are considered: natural and slightly enriched uranium (with enrichment up to 1.4%) and thorium fuel on basis of  $^{232}\text{Th}$ - $^{233}\text{U}$ . Intermediate chemical processing permits to prolong lifetime and to increase fuel burn-up. However, the effect is not so high, the increase of burn-up is about 20%. More effect is gained by use of a fuel with increased enrichment.

## 1. Introduction

A heavy-water CANDU-type reactor has good neutron-physical characteristics due to the use of heavy water as moderator and coolant. In particular, it allows using natural uranium as nuclear fuel, whereas in other types of thermal neutron reactors, it is necessary to use uranium with enrichment of several percents. In natural uranium fuel, the relative role of plutonium produced from  $^{238}\text{U}$  is great. Nevertheless, fuel burn-up and lifetime are small because fuel multiplying properties at burning out are quickly reduced. One of the opportunities to increase the lifetime is the transition to nuclear fuel with slight enrichment. Another opportunity, intermediate chemical processing of fuel can be considered where the fission products are removed and fuel nuclides are recycled for further burning.

For the future atomic power, a nuclear fuel cycle on base of  $^{232}\text{Th}$ - $^{233}\text{U}$  can be rather perspective. Thorium cycle has essential advantages over traditional uranium – plutonium cycle because of considerably less amount of transuranium long-lived radioactive wastes (though considerably more amount of rather harmful  $^{232}\text{U}$ ). In CANDU reactors, operation in thorium cycle and, the intermediate fuel cleaning of the fission products could also prolong lifetime and lower the requirement of specially obtained  $^{233}\text{U}$ .

In this paper, results are given of a calculation study of the influence of intermediate chemical processing of nuclear fuel with removal of fission products on fuel lifetime and burn-up in CANDU-type reactor are given. Uranium fuel on basis of natural and slightly enriched uranium (with enrichment up to 1.4%) and thorium fuel on base of  $^{232}\text{Th}$ - $^{233}\text{U}$  are considered.

## 2. Calculation model

The reactor design is described in [1]. In an active core, 380 fuel assemblies are placed. Every assembly contains 37 uranium pins with zirconium cladding in zirconium tube. Heavy water is used as the coolant and moderator. Fuel assemblies are located in a square lattice with a pitch of 23.5 cm. Height of an active core is 594 cm. Natural uranium loading in reactor is 114 tonnes.

It was accepted in calculations that reactor multiplying properties can be approximately described by multiplication factor of an elementary cell as follows. Multiplication factor of an elementary cell  $k_{\text{eff}}$  varies in function of fuel burn-up. The reactor operates in a mode of continuous refuelling. Fuel assemblies with various burn-up from fresh fuel up to maximum burn-up are situated in core at every moment. The on-load refuelling is carried out independently in different channels after achieving the maximal burn-up. It allows accepting the value of multiplication factor in an elementary cell  $\langle k \rangle$  average over fuel lifetime with correction on neutrons leakage from reactor as approximate reactor multiplication factor. Such approximation is quite justified for comparative calculations of the effect of intermediate nuclear fuel cleaning.

At calculations of lifetime, it was considered that the neutrons leakage makes 1% and the value  $\langle k \rangle = 1.01$  was accepted.

In reactors with continuous refuelling at constant power, the value of neutron flux varies in time very slightly. It is necessary that the power of one fuel assembly varies in time because of changing of fissile nuclide amount, and the fuel assemblies with different burn-up have appreciably different power. Calculations of fuel burn-up and transformation of isotopes were carried out at constant neutron flux.

### 3. Natural or slightly enriched uranium fuel

The calculations of reaction rates and multiplication factor in an elementary cell were performed with the code [2]. A fuel assembly with pins was represented as a 4-ring coaxial assembly with the same volumes of all-structural materials and fuel loading. The enrichment of uranium from 0.714% up to 1.4% was considered. The amount of uranium in fresh fuel assembly was accepted the same for all enrichments, and the  $^{235}\text{U}$  amount corresponded to the enrichment. For natural uranium fuel, thermal neutrons flux was equal to  $\Phi = 5 \cdot 10^{13} \text{ n/cm}^2\text{s}$ . Neutron flux for the enriched fuel was determined in a way to get the same power of fresh fuel assembly as in the variant with natural uranium. At calculations of nuclide transformation, isotopes of uranium, neptunium, plutonium, americium and curium up to  $^{244}\text{Cm}$  were taken into account.

In uranium fuel,  $^{239}\text{Pu}$  is produced. This isotope gives the essential contribution in reactivity of CANDU-type reactor with natural or slightly enriched fuel already in the initial period of fuel lifetime. The dependence of multiplication factor  $k_{\text{eff}}$  in an elementary cell on irradiation time  $T$  was calculated for initial enrichment of uranium from 0.714% up to 1.4% without intermediate cleaning. The neutron capture by fission products was taken into account by means of an “effective fission fragment” [3]. A poisoning by  $^{135}\text{Xe}$ ,  $^{105}\text{Rh}$  and absorption of neutrons by  $^{149}\text{Sm}$  and  $^{151}\text{Sm}$  were additionally taken into account. The fuel lifetime  $T_f$  was determined by value  $\langle k \rangle = 1.01$ . Table 1, fuel lifetime  $T_f$  and burn-up FP defined by amount of fission products in 1 tonne of fuel without intermediate cleaning for different initial uranium enrichment  $C$ .

Table 1. Fuel lifetime  $T_f$  and burn-up FP without intermediate cleaning

C, %	0.714	1.0	1.2	1.4
$T_f$ , years	2.0	2.8	5.38	6.67
FP, kg/tonne	11.4	17.4	20.8	23.4

Analogous time dependence of multiplication factor in an elementary cell  $k_{\text{eff}}$  with intermediate processing with cleaning from accumulated fission products was calculated for natural uranium fuel and fuel with enrichment 1%. The intermediate processing was carried out at time  $T_p = 0.8, 1.2, 1.6$  years. For enrichment 1%, the intermediate processing was carried out also at  $T_p = 2$  and 2.4 years. These data allow to estimate fuel lifetime corresponding to average over lifetime multiplication factor  $\langle k \rangle = 1.01$ . They are presented in Table 2.  $T_p = 0$  corresponds to the mode without intermediate cleaning.

Table 2. Fuel lifetime  $T_f$  and burn-up FP with intermediate processing

$T_p$ , years	C = 0.714%		C = 1.0%	
	$T_p$ , years	FP, kg/ton	$T_p$ , years	FP, kg/ton
0.0	2.0	11.4	3.9	17.4
0.8	2.52	13.8	4.71	19.9
1.2	2.4	13.3	4.76	20.1
1.6	2.4	13.3	4.69	19.8
2.0	–	–	4.57	19.5
2.4	–	–	4.41	19.0

These data show that fuel lifetime and burn-up in modes without intermediate processing essentially depend on fuel enrichment. At transition from natural uranium to enrichment 1%, the lifetime is increased 1.95 times (and 1.4 times because of reduction of flux density necessary to obtain the same power of fresh fuel assembly) and the burn-up grows 1.5 times. At transition from natural uranium to enrichment 1.4%, the lifetime is increased 3.3 times and the burn-up grows 2 times. In variants with intermediate processing, the lifetime increase is not so high. The maximal lifetime for natural uranium, 2.52 years, and burn-up, 13.8 kg/tonne, correspond to time point of processing  $T_p = 0.8$  years. The lifetime is longer by 26% and burn-up is greater by 21% than without intermediate processing. For 1% enrichment, the maximal lifetime is 4.76 years and burn-up makes 20.1 kg/tonne. Processing will be done at  $T_p = 1.2$  years. The increase in lifetime is 22% and that of in burn-up is 16% in comparison with a mode without processing.

#### 4. Thorium fuel $^{232}\text{Th}$ - $^{233}\text{U}$

In thorium mode of operation, all fuel assemblies were considered alike, containing identical fuel on basis of  $^{232}\text{Th}$  and  $^{233}\text{U}$ . The same elementary cell was studied as for uranium fuel. The thorium amount in fuel zones was accepted the same as  $^{238}\text{U}$  in uranium fuel. The share of  $^{233}\text{U}$  in fresh fuel was chosen 1.96% with respect to the amount of  $^{232}\text{Th}$ . That has ensured necessary over-criticality of a cell for appropriate fuel lifetime and burn-up.

During calculation of nuclide transformation, the production of isotopes of protactinium, uranium, neptunium, plutonium up to  $^{242}\text{Pu}$  was taken into account. The neutron flux is considered constant over a lifetime and equal to  $5 \cdot 10^{13}$  neutr/cm<sup>2</sup>s. In modes with intermediate processing, it was considered that short-lived  $^{233}\text{Pa}$  at an intermediate reactor shutdown completely decays into  $^{233}\text{U}$ . The intermediate processing is made at time points  $T_p = 0.4, 0.8, 1.2$  years. Fuel lifetime and burn-up corresponding to  $\langle k \rangle = 1.01$  for different points of intermediate cleaning are shown in Table 3.

Table 3. Fuel lifetime  $T_f$  and burn-up FP in thorium modes

$T_p$ , years	$T_f$ , years	FP, kg/ton
0	1.45	9.5
0.4	1.71	11.1
0.8	1.75	11.3
1.2	1.72	11.1

The lifetime without intermediate cleaning makes 1.45 years, burnup is 9.5 kg/ton. The maximal increase of fuel lifetime and burn-up at the expense of intermediate processing in comparison with a usual mode is achieved at processing at  $T_p = 0.8$  years and makes about 20%.

#### 5. Conclusion

The research performed has allowed to establish how it is possible to extend lifetime and to increase fuel burn-up at the expense of increase of uranium enrichment or at the expense of intermediate processing of uranium and thorium fuel with fission products removal. If a power of fresh fuel assembly remains constant with increase of uranium enrichment, it is necessary to reduce the neutron flux. At the expense of this effect, the lifetime is extended even at the same burn-up of fuel. Increase of burn-up and additional lifetime increase are caused by reactivity rise. At transition from natural uranium to

enrichment 1%, the burn-up grows 1.5 times, the lifetime is extended 1.95 times from 2 up to 3.9 years, the burn-up corresponding to natural uranium is achieved after 2.24 years. At transition from natural uranium to enrichment 1.4% the burn-up grows 2 times, the lifetime is extended 3.3 times. In modes with intermediate cleaning of fission products, an increase of lifetime is not so high. The lifetime for natural uranium raises by 26%, burn-up by 21%. For 1% uranium, an increase of lifetime is 22% and that of burn-up is 16%. The optimum time point of processing is somewhat less than half of lifetime without processing. In thorium mode, the maximal increase of fuel lifetime and burn-up at the expense of intermediate cleaning in comparison with a usual mode makes about 20%. Thus, the increase of burn-up and lifetime are obtained much more effectively at the expense of fuel enrichment.

## REFERENCES

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